Electric transport studies on topological insulators

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Electrical Transport Studies of Topological Insulators

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School of Physical and Mathematical Sciences

A thesis submitted to Nanyang Technological University in partial
fulfillment of the requirement for the degree of Doctor of Philosophy

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Statement of Originality

I hereby certify that the work embodied in this thesis is the result of original research and has not been submitted for a higher degree to any other university or institution.

Date:

Signature:

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SUMMARY

Recently, a new type of topological condensed matter system characterized by Z_2 topological invariant, which is different from the Chern number topological invariant in quantum Hall system, has attract huge attention in condensed matter physics. This new phase is called topological insulator. Topological insulators are different from ordinary insulators in the sense that it is composed of an insulating bulk state and of an massless spin-helical Dirac cone formed two-dimensional surface states. Helicity means that the spin-up electrons propagate in one direction while spin-down electrons propagate in the opposite direction. A fantastic prediction of this material is that the surface state is their insensitivity to any non-ferromagnetic impurities scattering and disorder. Thus, the carriers on the surface are predicted to have a long mean free path.

Shortly after the theoretical prediction of 3D topological insulators in real materials, a number of surface spectroscopy measurements, such as spin and angle-resolved photoemission spectroscopy (ARPES) and scanning tunneling microscopy, have been used to detect the topologically nontrivial surface state in three-dimensional topological insulator $\text{Bi}_{1-x}\text{Sb}_x$, Bi_2Se_3 , Bi_2Te_3 , *etc.*

This thesis presents transport experiments performed on topological insulators systems of Bi_2Se_3 , $\text{Ca: Bi}_2\text{Se}_3$ and $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$. We first performed the temperature and magnetic dependent measurements of Bi_2Se_3 and discovered

Shubonikov-de Haas oscillations which verify the quality of single crystal. Due to the large bulk conduction contributions, we fabricate $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ single crystals whose bulk conduction are strongly suppressed. A “Spin-valve” effect has been observed in the $\text{CoFe}(5\text{nm})/\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ structure and it may be originated from the interactions between ferromagnetic layer and helical surface states of topological insulator. Moreover, we prepared the nanodevice based on $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ nanoflake and observed the two channel conduction mode in the samples. Weak-antilocalization effect and ambipolar field effect were also verified and studied in detail based on the nanodevice by the low temperature electrical transport measurements. Interestingly, the superconducting transition was obtained in the pure Bi_2Se_3 at the temperature below 4K. We propose that the defects formed during the single crystal growth induce such transitions.

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Chapter 1

Induction

1.1 Landau paradigm and Topological order in condensed matters

The most familiar condensed matters are solids and liquids and the solid state physics has been taking a lot attention and it has become one of the most popular and developed branches in physics due to its applications. In the past tens of years, researchers always used Landau's theory to classify matter by phases and symmetry. For example, crystalline solids contains the translational symmetry, ferromagnets contains a rotational symmetry and water contains universal symmetry. In the theory, the symmetry breaking results in a phase transition in the matter. We can take water as an example, water can be transited from liquid into crystallized solid which is called ice as the temperature decreases below zero degree while keeping at atmospheric pressure. In this process, the symmetry of water breaks. Similarly, metal insulator transition also happens in the metal oxide V_2O_3 . The phase diagram is shown schematically in Figure 1.1 and the point B is a critical point. The "Mott" transition between a metal and a insulator takes place across the line AB. It should be noted that the phase line AB goes from the metallic to the insulating state as the temperature increases.

Figure 1.1 Schematic phase diagram of V_2O_3 . [47].

Landau's paradigm is a powerful tool and it allows scientists to not only arrange the phases of matter on a chart but to understand the behaviors of known phases. Physicists were so pleased with Landau's theory that for a long time they believed that all phases of matter could be described by symmetries. However, beginning in the 1980s, condensed matter researchers investigated new quantum systems where numerous ground states existed with the same symmetry. It has been pointed out that these new states contain a new kind of order: topological order. Topological order is a quantum mechanical phenomenon which means it is not related to the symmetry of the ground state but instead to the global properties of the ground state's wave function. Therefore, it transcends the Landau paradigm which is based on the classical physics concepts. Topological order is a more general understanding of quantum phases and the transitions between them. In this new framework, the phases of matter were described not by the patterns of symmetry in the ground state, but by the patterns

of a decidedly quantum property, entanglement. When two particles are entangled, certain measurements performed on one of them immediately affect the other, no matter how far apart the particles are. The patterns of such quantum effect unlike the patterns of the atomic positions, could not be described by their symmetries.

1.2 Quantum Hall effect and TKNN invariant

In 1980, Von Klitzing et al. Discovered the quantum Hall effect in a high mobility 2D semiconductor under high magnetic fields [48]. The occurrence of this effect is usually limited to very low temperature, in their study the localized carriers and quantized Landau energy level lead to the vanishing of longitudinal conductivity together with quantized hall conductivity in the condition that the Fermi level locates between Landau levels. This quantized Hall conductance was clearly figured out by Laughlin's gauge argument [49].

In 1982, Thouless, Kohmoto, Nightingale and Nijs (TKNN) reported the topology meanings behind the quantum Hall effect [2]. The quantum Hall Effect shows a quantized conductivity as $\sigma_{xy} = ne^2/h$ while the integer n is an integral of the Bloch wave functions over the whole Brillouin Zone (BZ) which also corresponds to the first Chern number [3] or TKNN invariant. The integer n is expressed as,

$$n = \frac{1}{2\pi} \int_{BZ} d^2k [\nabla_k \times A(k_x, k_y)]$$

with

$$A = \sum_{m=1}^N \langle u_{m,k} | -i \nabla_k | u_{m,k} \rangle$$

where u is Bloch wave function and A is the Berry connection. Therefore, the Chern number n is associated to the Berry phase which appears in the Bloch wave function when the momentum k parameter in the band m moves around the closed loop of the Brillouin Zone. In the 2DEG system, since the conductive current are merely from the edge states as we discussed above, the Hall conductivity is independent of the occupied bulk states. Moreover, the TKNN invariant or Chern number is the number of edge states and the Hall conductivity is protected from backscattering caused by impurities. Thus, σ_{xy} measured in the experiment is quantized and robust.

Such quantum phenomena can also be understood by the semiclassical mode. As a current drives through a two dimensional (2D) electron system in strong perpendicular magnetic field, electrons execute circular cyclotron motions under the Lorentz force (Fig. 1.2(a)). However, the one dimensional conducting channel is formed by the skipping orbits effect along the 2D boundary (Fig. 1.2(b)).

Due to the average effect, the bulk of the 2D system behaves as a band insulator. In this system, the Hall conductivity obtained is quantized in strong magnetic field whose value is always a multiple of e^2/h and it is called quantum Hall effect.

Figure 1.2: (a) Illustration of the electron cyclotron motion under magnetic field. (b) Semi-classical picture of edge states by skipping orbits.

Figure 1.3: Landau levels in a strip of finite width. Energy levels are pushed up at the edges of the strip. The Fermi level crosses the Landau levels near the boundary position generating conducting states.

The quantum Hall effect originates from the Landau quantization of the two dimensional electron gases (2DEG) in a magnetic field [1]. As shown in Fig. 1.3, the Landau Levels have to bend steeply due to the boundary potential. The Fermi level crosses only occupied Landau levels in the sample near to the boundary. Therefore, only the edges present the conductive channel. The Landau levels spread upwards or downwards when the magnetic field is applied and swept and that Landau levels will cross the Fermi energy level of a discrete number of states. Therefore, the quantized

plateaus appear in the Hall measurement of 2DEG system is the crossings of Landau level by the Fermi energy. The quantum Hall system can be considered as the first topological insulator which has become known to researchers.

1.3 Quantum Spin Hall effect

As discussed above, the QHE appears only when a magnetic field is applied on 2DEG, which also indicates that the time reversal symmetry is broken. In other words, the Hall conductivity or the TKNN invariant will be zero if the time reversal symmetry is preserved. In such case, given the fact of electron spins, the TKNN invariant can be expressed as $n = n_{\uparrow} + n_{\downarrow} = 0$. However, when considering the spin Hall conductivity, it will be a finite and quantized number instead of zero because $n_{\uparrow} - n_{\downarrow} \neq 0$. Recently, Kane and Moore showed the topological order of such spin system which is protected by time reversal symmetry [4-6]. Such new topological invariant called Z_2 topological order predicts a new Hall Effect, quantum spin Hall Effect. The materials will show trivial insulating properties when the number of edge states is even which gives Z_2 number $\nu = 0$. However, if the number of edge states is odd, a new material called topological insulator or quantum spin Hall insulator is produced with Z_2 invariant $\nu = 1$. In this material the quantized spin Hall conductivity is predicted in the absence of magnetic field. Spin-orbit coupling interactions of electrons are introduced into the material and take a role of magnetic field without breaking time reversal symmetry. Due to the spin-orbit coupling effect,

electrons with opposite spin orientations will propagate in opposite ways as shown in Fig. 1.4.

Figure 1.4: In the 2D quantum spin Hall state, the bulk of the crystal is insulating and the currents are carried by counter-propagating, spin polarized edge states. The blue channel current propagates with spin up electrons with a clockwise direction. The red channel current propagates with spin down electrons with an anti-clockwise direction.

It should be noted that TKNN invariant $n = 0$ makes the normal Hall conductivity zero but Z_2 invariant $\nu = 1$ gives the spin Hall conductivity a finite number instead of zero. Thus there will be only spin current in the system but the whole system will show zero charge current. It should be noted that zero net fields will be generated from spin-orbit interactions due to the averaging effect.

However, the electrons in the two dimensional system still cannot transport along edges only due to the random propagation of the spin in the bulk of the system. The quantum spin Hall effect has been predicted in 2005 and 2006 by Kane, Mele and Zhang in the two dimensional system separately [23, 50-52]. Kane and Mele provided a model to realized the quantum spin hall insulator based on graphene model with spin-orbit coupling. In graphene, the band structure consists of two linearly dispersing

cones located at two different points in the Brillouin Zone near the Fermi level. In the model [50], the spin polarization edge state can be achieved which is also called spin momentum locking and breaking of time reversal symmetry by magnetic field is not mandatory for realizing a topological phase. Unfortunately, the spin orbital coupling in graphene is so weak that it is unable to experimentally achieve the quantum spin Hall effect.

Figure 1.5: the energy spectra of the $\text{Hg}_x\text{Cd}_{1-x}\text{Te}$ quantum wells. (a) $M/B < 0$, the system is a normal insulator. (b) $M/B > 0$, the system becomes the quantum spin hall insulator [24].

In the recent years, Bernevig, Hughes and Zhang [53] constructed a 2D model to produce a Z_2 topological phase based on the band structure of HgTe. In the study of $\text{Hg}_x\text{Cd}_{1-x}\text{Te}$ quantum wells, the models of two dimensional topological insulators have been built up and the theoretical study shows that such quantum wells can be tuned from normal insulator to quantum spin Hall insulator by varying the sign of M/B which is dependent on the strength of spin orbital interactions. In the two

subbands with the pseudo spin orbital $\text{Hg}_x\text{Cd}_{1-x}\text{Te}$ quantum wells study, the effective Hamiltonian can be expressed as:

$$H(K) = \varepsilon(k)\mathbf{1} + \begin{pmatrix} M(k) & A(k_x + ik_y) & 0 & 0 \\ A(k_x - ik_y) & -M(k) & 0 & 0 \\ 0 & 0 & M(k) & -A(k_x - ik_y) \\ 0 & 0 & -A(k_x + ik_y) & -M(k) \end{pmatrix}$$

$\varepsilon(k) = C + Dk^2$, $M(k) = M - Bk^2$, where the upper 2x2 matrix describes the spin up electrons in the s conduction and p valence bands, and the lower matrix describes the spin down electron in those bands [24]. $\mathbf{1}$ is the identity matrix and $k^2 = k_x^2 + k_y^2$. B is a negative parameter which is used to describe the curvature of the band and the energy gap is $2M$.

It has been proved that the system behaves like a normal insulator when $M/B < 0$ and the band structure is shown in Fig. 1.5(a) while the system transits to quantum spin Hall insulator or two dimensional topological insulator when $M/B > 0$ as shown in Fig. 1.5(b). As $M/B > 0$, the edge bands cross with each other between the energy gap which connects the conduction and valance bands. The dispersion of the energy bands is linear near the crossing point which indicates the existence of massless electrons. Such transition from normal insulator to quantum spin Hall insulator takes place only when the $k^2 = k_x^2 + k_y^2$ is bigger. In such topological insulator system, electrons can only propagate along the edges with a defined spin orientation called helical edge states as shown in Fig. 1.3.

The two dimensional topological insulators are also called quantum spin Hall

insulators which have been discussed in the above section.

Figure 1.6: Schematic representation of CdTe/HgTe/CdTe heterostructure where d is the thickness of HgTe layer.

As we know, both CdTe and HgTe are normal semiconductor with a positive energy gap and similar lattice constant, while the $\text{Hg}_x\text{Cd}_{1-x}\text{Te}/\text{HgTe}/\text{Hg}_x\text{Cd}_{1-x}\text{Te}$ structure shows a properties transition from normal semiconductor to topological phase as the thickness of HgTe thin film increases. The experimental observation of quantum Hall Effect was first achieved in mercury telluride quantum wells [7, 8]. In their experiment, the HgTe layer was sandwiched between CdTe thin films to form a quantum well and all the thin film layers were deposited by molecular beam epitaxy (MBE) technique under the ultra high vacuum. By tuning the thickness of HgTe layer, the whole structure's properties will transfer from normal insulator to quantum spin Hall insulator. When the thickness of HgTe is thinner than a certain value ($\sim 6.5\text{nm}$), the structure exhibits a normal insulator with a positive energy gap. However, it exhibits quantum spin Hall effect as the thickness of HgTe is thicker than 6.5nm .

In the system whose spin orbit interaction is strong enough, the bands can be inverted which means the p orbital band will be pushed above the s orbital band. The

Figure 1.7: Schematic of the HgTe quantum well. The blue and red curves represent the potential energy well experienced by the electrons in the conduction band and holes in the valance band separately. (a) The lowest energy conduction sub band E1 is above the highest conduction sub band H1 when the thickness $d < 6.5\text{nm}$. (b) The lowest energy conduction sub band E1 is below the highest conduction sub band H1 when the thickness $d > 6.5\text{nm}$ [24].

band inversion usually appears in the heavy elements systems such as Bi, Hg and so on. In the HgTe quantum wells, the strength of the spin orbit interactions can be increased by increasing the thickness of the HgTe thin film so that the band can be inverted and a negative energy gap will be formed. As shown in Fig. 1.7, the energy band becomes inverted as the thickness of the quantum well becomes larger than the critical thickness (6.5nm).

As shown in Figure 1.8, four samples were prepared as topologically trivial insulators and quantum spin hall insulators. The trivial insulator was found when the Fermi level was tuned into the energy gap by a gating voltage indicated as black I curve.

However, the nontrivial insulator predicted to have conducting edge channel shows a constant conductance with an exact value of $2e^2/h$ as the Fermi level is fixed into the

Figure 1.8: Evidence of 2D topological insulator or quantum spin hall insulator in HgTe system. Device I is a trivial insulator and shows a diverging resistance in the bulk band gap. Device III and IV are quantum spin hall insulator with a conductance of $G=2e^2/h$ which is consistent with edge transport mode. Device II is a nontrivial insulator the edge modes can't persist unscattered [8].

energy gap. Further study showed that such constant conductance is independent of the size and geometry of device. It is convincing that the current can only conduct on the edge instead of the bulk.

1.4 Three Dimensional Topological Insulators

As the two dimensional topological insulator has been predicted and achieved, it is natural to ask whether the three dimensional topological insulators exist. The three dimensional topological insulators were first proposed by Fu and Kane in 2006 [25]. Unlike the one dimensional conductive edge channel in two dimensional topological insulators, the protected topological states are two dimensional surface states in 3D topological insulators.

In the discussion of section 1.3, the quantum spin Hall insulator depends on only one Z_2 topological invariant. However, there are four Z_2 invariants (ν_0 , ν_1 , ν_2 and ν_3) that characterize the topology of the band structure in three dimensions. ν_0 , ν_1 , ν_2 and ν_3 can be determined by the inversion symmetry of the system. The theoretical study shows that the strong and weak topological insulators can be distinguished by the value of ν_0 [10]. The weak topological insulators with $\nu_0=0$ have the surface states which are not protected by the time reversal and the carriers on the surface can be localized by the strong disorder [26]. However, the strong topological insulators with $\nu_0=1$ have the surface states against the disorder. The surface states of three dimensional topological insulator have been well understood by the Rashba spin-orbit Hamiltonian [27] which is similar to the two dimensional topological insulator as discussed in the above section. The Hamiltonian of the surface and bulk states in the three dimensional topological insulators has been studied [28]. In Zhang's work,

some extraordinary properties of the surface states were predicted, such as chiral surface states, spin momentum locking effect. The unique band topology of the surface states generates the π Berry phase which provides the protection against coherent back scattering and weak antilocalization.

A series theoretical work has been done in the study of 3D topological insulator. Here, besides Berry phase and TKNN invariant, a few concepts will be briefly introduced. For the arbitrary linear operator A , the time reversal operator can be expressed as:

$$\langle \Theta \phi | \Theta A \Theta^{-1} | \Theta \phi \rangle = \langle \phi | A^+ | \phi \rangle$$

If the system is under the time reversal symmetry and we consider the time reversal invariant momentum point Λ , the below relation can be expressed after a series derivation:

$$\text{tr}[a(k)] = \text{tr}[a(-k)] + i \text{tr}[w^+(k) \nabla_k w(k)]$$

Where $a_{\alpha\beta}(k) = -i \langle u_{\alpha,k} | \nabla_k | u_{\beta,k} \rangle$ and it is called Berry connection matrix. $w_{\alpha\beta}(k) = \langle u_{\alpha,-k} | \Theta | u_{\beta,k} \rangle$. This relation becomes very important in the calculation of the Z_2 topological invariant. It has been showed that the physical consequence of a nontrivial Z_2 invariant is the appearance of topologically protected surface states. This is graphically shown in Figure 1.9, in which topologically trivial and nontrivial surface states are compared. For nontrivial situation, the surface state is guaranteed to cross any Fermi energy inside the bulk gap. Considering the inversion symmetry in

the system, the Z_2 invariant can be calculated simply by the parity eigenvalues ξ_{2n} at time reversal invariant momenta Λ_i .

Figure 1.9, Schematic pictures of the surface states between two surface TRIMs,

Λ_a^s and Λ_b^s , for (a) topologically trivial and (b) topologically nontrivial cases. In the latter, the “switch-partner” characteristic is a reflection of the change in the TR polarization. The shaded regions represent the bulk continuum states. To enhance the visibility of the Kramers degeneracy, surface state dispersions are shown from $-\Lambda_b^s$ to Λ_b^s through Λ_a^s .

After the theoretical predication and numerical calculation, a series of three dimensional topological insulators have been predicated, such as $\text{Bi}_{1-x}\text{Sb}_x$, Bi_2Se_3 , Bi_2Te_3 , *etc.* In the mean while, a few groups started to prepare the high quality samples and a number of surface sensitive probes, such as spin and angle-resolved photoemission spectroscopy (ARPES) [10-18] and scanning tunneling microscopy [19,20], have been used to detect the topologically nontrivial surface state in $\text{Bi}_{1-x}\text{Sb}_x$, Bi_2Se_3 , Bi_2Te_3 , *etc* single crystals. Fig. 1.9a-c is ARPES results for the

proposed topological insulator Bi_2Se_3 , while Fig. 1.9d shows a schematic band structure based on theoretical expectations. As shown in Fig. 1.10c, an energy gap of around 300 meV appears between the valence and conduction band, two linear dispersion bands coming from the sample surface cross with each other in the gap. The Fermi level crosses the surface bands in the gap also. Fig. 1.10b shows the spin-polarization experiments and it demonstrates that the electron spins are polarized at different k_x and opposite on each side of Dirac cone. To observe this momentum dependent spin effect more clearly, the ARPES results along z direction is selected as shown in Fig. 1.10a. The Dirac point is easy to obtain in the middle and the spin orientations near the point form a clockwise rotation and the electrons with opposite momentum have an antiparallel direction of spin. In another word, the spins in 3D topological insulator are locked perpendicular to the wave vector k .

To understand the spin-momentum locked effect, it is better to use a simple model of 2D surface states in a 3D material to explain it as shown in Fig. 1.11. On the upper surface, the electrons experience an electrical field $\mathbf{E}_{\text{trap}}$ which is also called trapping field originating from the broken of inversion symmetry. $\mathbf{E}_{\text{trap}}$ points down and restrains the electrons. As one electron moves on the upper surface with wave vector $\mathbf{v}$, an effective magnetic field is present and expressed as $\mathbf{B} = \mathbf{v} \times \mathbf{E}_{\text{trap}}$. Due to the

Figure 1.10: Angle-resolved photoemission spectroscopy results for TI Bi_2Se_3 [15]. (a) Dirac cone observed in z direction and the spin is noted by arrows. (b) Spin polarization measurement of electrons. (c) Bulk energy structure and surface state. 300meV bang gap and Dirac cone can be obtained directly. (d) Schematic of band structure of Bi_2Se_3 .

effective magnetic field B , the spins of the electrons will align with B to minimize the energy. This effect is Rashba-like Hamiltonian and it takes an important role in strong spin orbit coupling materials as discussed in the previous part. Thus, the spin current is produced at the upper surface as we drive a voltage bias through it. Similarly, another spin current with opposite orientation can be formed simultaneously at the bottom surface of the 3D material.

Figure 1.11: Schematic of spin momentum locked effect. Electrons on the top surface of a 3D topological insulator have a spin momentum locked states while the spin s pointed perpendicular to their wave vector v as determined by the effective B field.

When the time reversal invariant applies in such system, we will find that the electron with energy of $E(k, s)$ keep the constant as $E(-k, -s)$ after the electron revisers both momentum and spin. Thus, $E(k, s) = E(-k, -s)$. If the inversion symmetry is considered here, we can find that $E(k, s) = E(-k, s)$, together with time reversal symmetry, which would imply that $E(k, s) = E(k, -s)$. Therefore, the energy band is spin degenerated in the bulk since that the bulk of the material is inversion symmetry preserved. Now we consider the time reversal symmetry at $k=0$ point, we will find that there must be two spin states with opposite orientation at this point and it is called Dirac Point. The Dirac point will not be allowed to split apart and be protected from opening a gap by the topology protected continuity of the states.

The unique properties of the surface states of topological insulators are expected to form a playground of various quantum effects and show great potential in spintronics and quantum computation [17-19]. To fulfill the expectations, electrical transport study of significant surface of topological insulator systems is essential.

In the transport study of topological insulator, a number of evidences of two dimensional electron gas have been discovered in Bi_2Te_3 where the single crystal samples shows a nonmetallic properties after a tricky annealing process [47]. In their experiment, the angular dependent SdH oscillations have been obtained in the nonmetallic samples as shown in Fig.1.12. As the angular dependent SdH oscillations can only exist in the two dimension system, It is obvious that the nonmetallic samples contain a surface transporting layer which may origin from the topological surface states. However, in the metallic samples N1, the angular dependent SdH oscillations are unable to be obtained and it means such oscillations origin from the bulk carrier. It should be noted that it is the first and significant transport experiment show the existence of surface state in the topological insulator system. Similar experiments were also performed in Bi_2Se_3 and BiSb single crystal but most of the SdH oscillations observed originate from the bulk states due to the high bulk carrier concentration.

To confirm the surface transporting layer originate the topological surface states, the further Hall measurement is performed in the nonmetallic samples. As shown in Fig 1.13, an anomaly Hall effect was obtained in the low magnetic field. The distinct anomaly of the hall conductivity in Fig 1.13(A), appears around $B=1\text{T}$ and it is obvious to see the two different types of carriers coexist in the sample. The conductivity turns out to be electron like at low field (-1T to 1T), while it changes to hole like at high field beyond 1T . The measured Hall conductivity is summed by a

bulk hole like conductivity σ_{xy}^b and surface electron like conductivity G_{xy} .

Figure 1.12: (A) Angular dependence of $d\rho_{xx}/dH$ vs effective perpendicular magnetic field in sample Q1 which behaves as nonmetallic. (B) Angular dependence of $d\rho_{xx}/dH$ vs effective perpendicular magnetic field in sample H1 which behaves as metallic. (C) The minima field position of the $n=3$ Landau levels as a function of θ for Q1 where θ is the angle between H and the vector normal to the cleavage plane. (D) The minima field position of the $n=3$ Landau levels as a function of θ for N1. The experiment data blue circles is not fitted with the theoretical black curve [47].

The theoretical fitting analysis was performed to the measured results which is marked as the solid curve in Fig 1.12. After subtracting the bulk hole like conductivity, the surface conductivity G_{xy} is present as shown in Fig 1.13 (B). All the fitting parameters are figured out to further confirm the electron like conductivity coming from the 2D Dirac surface states.

Figure 1.13: (A) The measured hall conductivity vs magnetic field marked as open circles and theoretical fitting curve in black solid curve. (B) The surface conductivity vs magnetic field (red circles) after subtracting the bulk contribution (the solid blue curve) [47].

A series of materials have been predicted to be three dimensional topological insulators such as $\text{Bi}_{1-x}\text{Sb}_x$, Bi_2Se_3 , Bi_2Te_3 and Sb_2Te_3 [10-20]. They have long been known for thermoelectric applications. All the topological insulators mentioned above have the rhombohedral structure composed of quintuple layers by van der Waals forces and the energy gap of them is around 0.3 eV. BiSb , Bi_2Se_3 , Bi_2Te_3 and Sb_2Te_3 are supposed to have the chiral surface states based on the first principle calculations [29]. Below is a summary of the TI materials in Table 1 discovered so far.

Type	Material	Band gap	Bulk transport	Remark	Reference
2D, $\nu = 1$	CdTe/HgTe/CdTe	< 10 meV	insulating	high mobility	31)
2D, $\nu = 1$	AlSb/InAs/GaSb/AlSb	~4 meV	weakly insulating	gap is too small	73)
3D (1;111)	$\text{Bi}_{1-x}\text{Sb}_x$	< 30 meV	weakly insulating	complex S.S.	36, 40)
3D (1;111)	Sb	semimetal	metallic	complex S.S.	39)
3D (1;000)	Bi_2Se_3	0.3 eV	metallic	simple S.S.	94)
3D (1;000)	Bi_2Te_3	0.17 eV	metallic	distorted S.S.	95, 96)
3D (1;000)	Sb_2Te_3	0.3 eV	metallic	heavily p -type	97)
3D (1;000)	$\text{Bi}_2\text{Te}_2\text{Se}$	~0.2 eV	reasonably insulating	ρ_{xx} up to 6 Ωcm	102, 103, 105)
3D (1;000)	$(\text{Bi,Sb})_2\text{Te}_3$	< 0.2 eV	moderately insulating	mostly thin films	193)
3D (1;000)	$\text{Bi}_{2-x}\text{Sb}_x\text{Te}_{3-y}\text{Se}_y$	< 0.3 eV	reasonably insulating	Dirac-cone engineering	107, 108, 212)
3D (1;000)	$\text{Bi}_2\text{Te}_{1.6}\text{S}_{1.4}$	0.2 eV	metallic	n -type	210)
3D (1;000)	$\text{Bi}_{1.1}\text{Sb}_{0.9}\text{Te}_2\text{S}$	0.2 eV	moderately insulating	ρ_{xx} up to 0.1 Ωcm	210)
3D (1;000)	$\text{Sb}_2\text{Te}_2\text{Se}$	?	metallic	heavily p -type	102)
3D (1;000)	$\text{Bi}_2(\text{Te,Se})_2(\text{Se,S})$	0.3 eV	semi-metallic	natural Kawazulite	211)
3D (1;000)	TlBiSe_2	~0.35 eV	metallic	simple S.S., large gap	110–112)
3D (1;000)	TlBiTe_2	~0.2 eV	metallic	distorted S.S.	112)
3D (1;000)	$\text{TlBi}(\text{S,Se})_2$	< 0.35 eV	metallic	topological P.T.	116, 117)
3D (1;000)	PbBi_2Te_4	~0.2 eV	metallic	S.S. nearly parabolic	121, 124)
3D (1;000)	PbSb_2Te_4	?	metallic	p -type	121)
3D (1;000)	GeBi_2Te_4	0.18 eV	metallic	n -type	102, 119, 120)
3D (1;000)	PbBi_4Te_7	0.2 eV	metallic	heavily n -type	125)
3D (1;000)	$\text{GeBi}_{4-x}\text{Sb}_x\text{Te}_7$	0.1–0.2 eV	metallic	n (p) type at $x = 0$ (1)	126)
3D (1;000)	$(\text{PbSe})_5(\text{Bi}_2\text{Se}_3)_6$	0.5 eV	metallic	natural heterostructure	130)
3D (1;000)	$(\text{Bi}_2)(\text{Bi}_2\text{Se}_{2.6}\text{S}_{0.4})$	semimetal	metallic	$(\text{Bi}_2)_n(\text{Bi}_2\text{Se}_3)_m$ series	127)
3D (1;000)	$(\text{Bi}_2)(\text{Bi}_2\text{Te}_3)_2$	?	?	no data published yet	128)
3D TCI	SnTe	0.3 eV (4.2 K)	metallic	Mirror TCI, $n_M = -2$	62)
3D TCI	$\text{Pb}_{1-x}\text{Sn}_x\text{Te}$	< 0.3 eV	metallic	Mirror TCI, $n_M = -2$	164)
3D TCI	$\text{Pb}_{0.77}\text{Sn}_{0.23}\text{Se}$	invert with T	metallic	Mirror TCI, $n_M = -2$	162)
2D, $\nu = 1$?	Bi bilayer	~0.1 eV	?	not stable by itself	82, 83)
3D (1;000)?	Ag_2Te	?	metallic	famous for linear MR	134, 135)
3D (1;111)?	SmB_6	20 meV	insulating	possible Kondo TI	140–143)
3D (0;001)?	$\text{Bi}_{14}\text{Rh}_3\text{I}_9$	0.27 eV	metallic	possible weak 3D TI	145)
3D (1;000)?	$R\text{BiPt}$ ($R = \text{Lu, Dy, Gd}$)	zero gap	metallic	evidence negative	152)
Weyl SM?	$\text{Nd}_2(\text{Ir}_{1-x}\text{Rh}_x)_2\text{O}_7$	zero gap	metallic	too preliminary	158)

Table I. Summary of topological insulator materials that have been experimentally addressed. The definition of (1;111) etc. is introduced in Sec. 1.4. (In this table, S.S., P.T., and SM stand for surface state, phase transition, and semimetal, respectively.) [54]

1.5 Spin transport and two dimensional system

1.5.1 Giant Magnetoresistance

Giant magnetoresistance, GMR, is the phenomena where the resistance of the multi layer structure with more than one magnetic conducting layer changes when the relative orientation of the magnetizations change. The GMR effect was simultaneously discovered in the Fe/Cr multilayers structure by Prof. Albert Fert [30] and in Fe/Cr/Fe trilayers by Prof. Peter Grunberg [31] in 1988 and both of them were awarded Noble prize in physics in 2007.

Mott model is used for the qualitative analysis of the GMR effect [32,33]. In this model, the conduction carriers in the magnetic layer can be separated into two different groups, spin up and spin down electrons, whose scattering rates are different. When the spins of the electrons align with the net magnetization of the material, the scattering rate is low and the low resistance can be obtained. However, for antiparallel alignment of the electrons spins with the net magnetization, the scattering rate is high and the high resistance can be obtained. As shown in Fig. 1.14, the current in the GMR structure can be considered to be composed of two parallel channels, a spin up current channel and a spin down current channel.

In Fig. 1.14 (a), the spin-up electrons are aligned with the magnetic moments and have less scatterings in both top and bottom magnetic layers. However, the spin-down

Figure 1.14: Schematic of GMR effect with parallel (a) and anti-parallel (b) configurations. Arrows indicate the magnetization direction in the magnetic layer. The spin up and spin down electron trajectories are shown in both configurations assuming that the mean free path of the electrons is much larger than the thickness of the magnetic layer. $R_{\uparrow}$ and $R_{\downarrow}$ are resistances of up spin and down spin electrons resistance respectively. The small and large boxes correspond to the resistance valve in the magnetic layers for up and down spin electrons.

electrons are aligned anti-parallel to the magnetic moments in both top and bottom magnetic layers and therefore have high scatterings. The less and high scatterings mean the low and high resistance respectively. Based on the combination of parallel channels of the spin, the overall resistance is low. If we let the magnetization direction in the top layer flips through the applied magnetic field while keeping the magnetization direction in the bottom layer, a different phenomenon appear. Such anti-parallel magnetic layers structure can be achieved by composing two magnetic layers with different coercivities. As shown in Fig. 1.14 (b), the scatterings are

significantly enhanced when the spin and magnetic moments are antiparallel because higher scattering of spin up electrons from both top and bottom magnetic layers. Thus, the whole resistance of the GMR stack in this anti-parallel configuration is relatively higher than that in the parallel configuration.

The relative orientation of the magnetizations is tuned by the magnetic field. GMR can be obtained for both current traveling perpendiculars to the plane of the layers (CPP) and current traveling in the plane of the layers (CIP). The CPP and CIP can be comparable if the resistivity mean free path λ is much larger than the individual layer thickness. The CPP geometry can be described as the collection of layer resistance in series while the CIP geometry can be thought as the layer conductivities added in parallel. The GMR effect obtained in the experiment is usually named as GMR ratios and it significantly depends on the temperature. The GMR ratio increases with decreasing temperature.

After the discovery of GMR effect, the structure of ferromagnetic/normal metal (FM/NM) and ferromagnetic/normal metal/ferromagnetic (FM/NM/FM) have also been well studied [34]. The high GMR effect was obtained in Fe/Cr multilayers structure due to the band matching for the same orientation of spin [35]. In the structure, the minority spins have the similar band matching between Fe and Cr layers, while the majority bands are quite different. Therefore, such band matching gives rise to strong spin dependent scattering of electrons for antiparallel configuration of

magnetizations. Similarly, GMR effect in Co/Cr or Fe/Cu structures is neglected due to the band mismatching.

1.5.2 Anisotropic Magnetoresistance

Anisotropic magnetoresistance is usually named as AMR and the resistance of the material is closely related to the anisotropy of the magnetization. Usually, there are two types of magnetoresistance: positive and negative. Positive resistance refers to the increase in resistance with increase in magnetic field due to the Lorentz's force and it typically appears in the non-ferromagnetic metals. However, negative resistance refers to the decrease in resistance with increase in magnetic field and it is a common feature of ferromagnetic metals. If θ is the angle between current direction and magnetization direction, then the resistance will be proportional to $\cos^2\theta$. Anisotropic magnetoresistance, $\Delta\rho$, is defined as the difference between the resistance when the magnetization is parallel to the current versus perpendicular to the current.

In spite of the long history of AMR study, the mechanism of the AMR is still ambiguous. The AMR and Planar Hall Effect (PHE) are usually observed in various ferromagnetic materials, such as 3d ferromagnetic metals [36-38], magnetic semiconductors [39], and magnetoresistive oxide [40-42]. The AMR and PHE effect is due to the spin-orbit interaction in ferromagnetic materials. Electrical resistance originates from the electron scattering by phonons, magnons, impurities

and other electrons. The scattering probability is different along different directions because the wavefunctions of the electrons are not symmetric, and therefore the resistivity along different directions is different. Because of the spin-orbit interaction, the symmetry of the electron wavefunction in magnetic materials is lower than cubic. The electron wave function with $S=1/2$ (spin up) prefers to align parallel to the magnetization, while the electron wave function with $S=-1/2$ (spin down) prefers to align perpendicular to the magnetization. Suppose the current is positively spin polarized, the carrier will have more chance to be scattered when the current is parallel to the magnetization, because many spin up states are available in the transport direction (small angle scattering dominates), therefore positive AMR is realized. If the current is negatively spin polarized, the carrier will have less chance to be scattered when the current is parallel to the magnetization, and thus negative AMR is obtained. The above clarification was demonstrated by McGuire and Potter's detailed calculations [35]. Recently, AMR is also investigated in the combined Rashba and Dresselhaus model in the presence of electromagnetic impurity. It is proposed that one of the essential conditions for the occurrence of AMR is the existence of polarized magnetic impurity. In the presence of the magnetic field, the density will be quite different for spin up and spin down electrons in ferromagnetic materials.

1.5.3 Weak Antilocalization Effect

In the perfect periodic crystal, the transport electrons suffer a periodic potential and move ballistically without any loss in momentum. However, such perfect crystal is extremely difficult to be realized in the experiment. Defects and impurities will be introduced into the crystal during the sample preparation period. Therefore, diffusive transport should be commonly observed.

In the disordered system, the electrons will propagate and be scattered away with randomized momentum and phase. The phase of electron wavefunction is very important in the studying of transport properties. For example, in the single elastic scattering, the electrons will keep the phase after each elastic scattering and interfere with each other during the transport procedure. However, in the inelastic scattering such as electron-phonon scatterings, electron-electron scatterings and electron-magnetic impurities scatterings, the phase of electron will be affected. To discuss the quantum interference effect in the electron transport, a few concepts should be defined. We define the τ_ϕ as the dephasing time of an electron which is the time of an electron keeping the original phase. In another way, τ_ϕ is also treated as the lifetime of electron before inelastic scattering. The dephasing time is defined as

$$\tau_\phi^{-1} = \frac{1}{\tau_{e-ph}} + \frac{1}{\tau_{ee}} + \frac{1}{\tau_{imp}} + \dots \text{ where } \tau_{e-ph}, \tau_{ee} \text{ and } \tau_{imp} \text{ are the dephasing time for}$$

electron-phonon, electron-electron and inelastic impurity scattering. Similarly, the coherence length l_ϕ is defined as the distance of the electron traveled before losing

the original phase or before the inelastic scattering and $l_\phi = \sqrt{D\tau_\phi}$ for 2D system, where $D = v_F^2 \tau_m / 2$ is called diffusion coefficient.

Considering an electron wave that diffuse back through a closed loop to the initial point by the elastic scatterings, the phase of the back-scattered electron is the same with the phase of time reversed back-scattered electron. Therefore, two electron waves will interfere with each other constructively at the initial point and it means there is an enhanced probability that the electron will have a zero net displacement which is also called localization. This phenomenon is called weak localization (WL) [43]. Such constructive interference can only be obtained at the finite temperatures and will be destroyed by increase of the temperature. Because the elastic scattering can be only valid when $L_m \gg L_e$. As the temperature increases, the electron phonon scattering will play a more and more important role. In the presence of a magnetic field normal to the plane of carrier motion, it breaks the time reversal symmetry and suppresses the Weak-Localization and therefore, leads to a negative magnetoresistance at low magnetic fields around $B=0$ [44].

The conductivity correction from the weak localization is give by:

$$\Delta\sigma = -\frac{4e^2 D}{h} \int_0^\infty P(0,t) dt$$

Where $P(x,t)$ is the probability of the electron moving with a path x in time t .

When the spin orbit interaction is introduced into the above situation, a more interesting effect will be obtained. In the spin orbit interaction system, the spin of the carrier is coupled with the momentum. The interference of time reversed paths has consequences beyond the weak localization effect. As the spin experiences a sequence of scatterings along the path, the spin orientation is randomized in the scale of l_{SO}

Figure 1.15: An electron starting at point 0 can return to 0 through the unprimed path or primed path. The primed path is simply the time reversal of the unprimed path.

which is the length of electron traveled in spin orbital interaction time. The stronger the spin orbital interaction, the smaller is l_{SO} . When $B=0$, the interference between the path of electron and the time reversed paths will lead to a reduction of the backscattering and this is called Weak-Antilocalization. Weak-Antilocalization was experimentally obtained by Bergmann in thin film [45] and the transition from weak localization to Weak-Antilocalization has been observed as increase the spin orbital interaction strength [46]. The field dependent magnetic conductance for the weak anti-localization in 2D systems can be expressed as:

$$\Delta\sigma_{xx}(B) = \alpha \frac{e^2}{\pi h} \left[\Psi\left(\frac{\hbar c}{4eL^2\phi B} + \frac{1}{2}\right) - \ln\left(\frac{\hbar c}{4eL^2\phi B}\right) \right]$$

where Ψ is the digamma function and L_ϕ is the phase coherence length.

For the Dirac fermion with π Berry phase, it also leads to the weak antilocalization effect. With the magnetic field, the two time reversed paths interfere with each other destructively due to the π phase shifts in the closed loop. The localization of electrons will be further reduced in this case. The prefactor α will be $-1/2$ for each transport channel that either carries the π Berry phase or bears a strong spin orbital coupling. Since both effect can lead to the weak antilocalization, it will be difficult to judge the existence of Dirac fermions by the observation of the weak antilocalization effect in the experiment. The weak antilocalization with $\alpha = -1/2$ has been observed in the Bi_2Se_3 thin films which was fabricated by MBE. It has been showed that the top and bottom surfaces are connected through the bulk channel to form a single diffusive transport channel [55-59]. For the thickness of the thin film shorter than the coherence length, electrons can always diffuse from the surface to the other surface without losing the phase memory. For the low bulk carrier density topological insulators, the gating technique can be used to decouple one of the surface states from the rest by creating a gapped layer. Therefore, α can be found to become -1 which is consisting of two independent diffusive transport channels [57,58] and the π Berry phase can be proven in such experiment.

1.5.4 Universal Conductance Fluctuations

In the two dimensional electron gas system, the conductance can be well studied by the Drude model in the classical. In such model, the conductance is expressed as: $G = \frac{W}{L} n_s e \mu$, where L and W is the length and width of the device, n_s is the sheet density of electron and μ is the mobility. In this model, the electron phase breaking length l_ϕ is much smaller than the size of device thus electrons are treated as completely incoherent due to the inelastic scattering during the transport.

However, in the nanodevice whose dimension is even smaller than the phase breaking length of electron, the transport procedure should be treated as wave transport. The wave function coherence issue must be counted in the electron transport. The electron wave can be scattered from the impurities and the subwaves will be generated and interfere with each other after the scattering event. It has been shown that the interference between partial wave functions of a conduction electron will affect the conductivity of a disordered metal in the last part. However, the weak localization is dependent on the specific types of paths and is not directly dependent on the orientation of scattering sites.

The Universal Conductance Fluctuations (UCF) is magnetofingerprint effect that the root mean square magnitude of the conductivity is to be order of e^2/h . For each system, it has a unique arrangement of scattering sites. Thus the interference pattern

is sample specific. An example of such magnetofingerprint at different temperatures is given in Figure 1.16.

Figure 1.16: The UCF of a 125nm wide Ag wire. The features of the magnetofingerprint are reduced at higher temperatures due to the temperature dependence of the coherence length.

In the phase coherent 1D metal wire, the coherence length is larger than the length of

the sample, the rms magnitude of UCF can be expressed as $\text{rms}(G_N) \approx 0.730 \frac{e^2}{h}$

when the magnetic flux $\Phi < \frac{h}{e}$, otherwise, $\text{rms}(G_N) \approx 0.516 \frac{e^2}{h}$. More generally, the

rms magnitude has such expression: $\delta G = e^2 / h [L_\phi / L]^{4-d}$ where d is the dimensions of the sample.

1.6 Organization

In this thesis, the transport experiment of topological insulator in Bi_2Se_3 and $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$. Chapter 2 describes the general experimental apparatus while further details relevant to each experiment will be given at the beginning of the appropriate chapter. Chapter 3 describes the surface dominated transport in single crystalline nanoflake devices of topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$. Chapter 4 describes the anisotropic magnetoresistance in topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}/\text{CoFe}$ heterostructures. Chapter 5 describes some other topological insulator single crystals we discovered before $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$. The superconductor-like Bi_2Se_3 will also be reported in this chapter. Finally, I briefly summarize our results and propose the future work in Chapter 6.

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Chapter 2

Experimental Setup

This Chapter presents the sample preparation and the experimental techniques used to characterize topological insulators. I start by introducing the modified Bridgeman method for sample preparation and device fabrication, followed by the experimental setup for electrical transport measurements.

2.1 Sample preparation

The Bridgeman method is a popular way to fabricate semiconductor single crystals. The polycrystalline material sources will be heated over their melting point and then cooled down from one end of its container to produce a seed crystal first. The temperature gradient is formed near the single crystal growth region, so that the bottom side is a few degrees cooler than the upper side of the vertical furnace. Thus, the seed crystal can be formed first at the bottom region and then the single crystal with the same crystallized orientation as the seed crystal will be formed progressively.

We choose high temperature quartz tube as the container of the sample. All the elementary sources such as bismuth, selenium powders (for Bi_2Se_3 single crystal) are precisely weighted in glove-box and put into the cleaned quartz tubes. Turbo pump station is used to evacuate such quartz tubes to a vacuum level of 10^{-6} Torr. All the

quartz tubes containing stoichiometric mixtures of elementary powders were sealed under acetylene jet-burner with a temperature as high as 2000 °C in the mechanical workshop. The sealed quartz tubes which are shown in Fig. 2.1 are put into furnace to be pre-melted at 950 °C for one week. In this step, all the elementary powders are melted and mixed together. Then we put the tubes with annealed powder vertically in the single crystal furnace. We modified the single crystal furnace to make sure that the temperature gradient inside is more even around the sample region. The samples in the furnace are kept at 850 °C for three days and slowly cool down to 550 °C over 5 days and then cool down to room temperature quickly.

Figure 2.1: Picture of evacuated quartz tube after sealing by acetylene jet-burner.

The crystals were cleaved very easily along the basal plane and were cut into 1 x 5 x 5 mm³ rectangular size samples for X-ray diffraction experiment. The crystal structure of Bi₂Se₃ is hexagonal structure and the periodic stacking of Bi-Se quintuple layers along the c axis are shown in Fig. 2.2. The unit cell spans the quintuple layers

with lattice constant of $a = 0.41$ nm, $c = 2.86$ nm which agrees with the XRD experiment patterns very well. The XRD patterns of Bi_2Se_3 are also shown in Fig. 2.3.

Figure 2.2: Schematic crystal structure of Bi_2Se_3 , the dashed panel shows a unit of quintuple layers. The crystal structure with primitive lattice vectors denoted as $t_{1,2,3}$. A quintuple layer with $\text{Se1-Bi1-Se2-Bi1}'\text{-Se1}'$ is indicated by the red square [1].

The XRD results show that the single phase of (001) is detected from the single crystal sample which indicates the good quality of the sample. A series of samples with different Se composition were fabricated and resistivity characterized results are shown in Figure 2.4.

Figure 2.3: X-ray diffraction patterns of Bi_2Se_3 single crystal which was prepared by classical Bridgeman method used for transport measurements. No impurity phases are discovered from XRD patterns.

Figure 2.4: Temperature dependence of longitude resistivity in single crystal of Bi_2Se_x which was prepared by classical Bridgeman method with x varying from 3 to 6. All

samples behave as metals of resistance decreasing with temperature but saturate below 30 K.

2.2 Experiment setup of electrical transport measurements

All of the transport measurements in this thesis were performed in Physical Properties Measurement System (PPMS). It can supply a temperature range between 2 K to 400 K and a magnetic field range between -9 T to 9 T in the sample space. The picture of our PPMS is shown in Fig. 2.5. For bulk single crystal transport measurement, a constant DC or AC current of 0.2 mA are used usually.

Figure 2.5: Setup of PPMS in our lab. The probe covered by aluminum foil is inserted into the black dewar which filled by liquid helium in the inner jacket. Temperature and magnetic field are supplied by this container.

The schematic structure of sample space chamber is shown as Fig. 2.6. The superconductor coils submerged in liquid Helium are enclosed by a vacuum isolation. Outside of the bore of the insulation, there is a stainless steel container consisting of liquid nitrogen to protect the evaporation of helium. The experimental probe with a thermal sensor is put into the sample space and all the space is kept at low pressure (28 torr) evacuated by an mechanical vacuum pump. We use DC and AC sample puck to do the electrical transport measurement of single crystal bulk samples while the nanodevice sample is mounted on the multifunctional probe for measurement as shown in Fig. 2.7.

Figure 2.6: Schematic cross section structure of major components of the PPMS probe.

During the measurement, the sample can be tuned to perpendicular or parallel to magnetic field. Sweeping angle is also achieved by the motor associated to PPMS. The angular dependent magnetoresistance measurement is very important in the topological insulator study which is going to be described in the following chapters in Bi_2Se_3 , $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ single crystals and nanoflake devices.

Figure 2.7: Multifunctional probe used in the device measurement. To achieve the low noise measurement, we modified the multifunctional probe and use external shielding cable to connect the device to the instruments outside. The cable connection is covered by the aluminum foil as shown in the picture.

To achieve the precise and low noise measurement, we use external instruments, SR830 lockin amplifier, Keithley 6221 DC source, 2182 voltage meter, 2400 voltage source and preamplifier. The schematic electric circuit for nanodevice measurement is shown in Fig. 2.8. A constant voltage of 0.1 V comes out with a certain frequency (677.7 Hz) from sine-out of the lockin amplifier and then the signal is transformed into a constant current (100 nA) after a standard huge resistor of 1 M Ω . The measured

potential difference is filtered and amplified by Low-Noise preamplifier (SR-560) and then received by lockin amplifier. The measured voltage is divided by the current through the device and the resistance is calculated by software and shown on the monitor as the y-axis. In the meantime, the values of temperature and magnetic field at the sample's position are recorded by the labview program and plotted as x-axis on the monitor. The sweeping or stable gating voltage is supplied by Keithley instrument 2400 voltage source meter. Step-sweeping mode is used for low temperature Ambipolar effect measurement and all the measurement can be controlled by the labview program.

Figure 2.8: Schematic of electric circuit in nanodevice measurement. Signal is activated and received synchronously and controlled by labview programming.

2.3 Device fabrication and electrode deposition

The devices are fabricated by photolithography method and all the procedures were carried out in clean room Nanoscale Device Fabrication Centre in School of Electrical and Electronic Engineering, Nanyang Technological University.

Spin coat photoresist AZ5214

AZ5214 is the most popular photoresist used in the device fabrication. The substrates were first spin-coated with photoresist by spin coater and the spin speed was 4000 rounds per minute (rpm). The coating time was 30 seconds. The thickness of the photoresist coated was controlled by the spin speed and coating time and it is about 1.4 ± 0.2 micrometers for our experiment [1]. The photoresist was spread over the substrate due to the centrifugal force during the fast rotation of the spin coater. Air suction was applied so that the substrate stay firm during spinning. Each substrate was prebaked at 105 °C for 105 seconds immediately after spin-coating. The substrates were then placed on the chuck of MJB4 mask aligner, such that the nanoflake and the electrode pattern image from the photo mask can be aligned.

Alignment of substrate and photo mask in MJB4

The photo mask was secured by vacuum suction, and placed on the mask holder with the chrome side facing up. The mask holder was then inserted into the mask holder frame, and secured by screws. The desired electrode pattern from the mask was selected by adjusting the Top-Side Alignment (TSA) microscope. The photo mask was removed after alignment, and the substrate was placed on the chuck, followed by pushing up the contact level at the side. Caution was taken to ensure that TSA microscope remain in the same position after the alignment was done. The chuck holder was aligned manually so that the nanoflake on the substrate was in correct position and orientation with the electrode pattern from photo mask. Following this,

Figure 2.9: Mask Aligner MJB-4.

the photo mask was inserted again, and the alignment was fine tuned until the

nanoflake was perpendicularly across the selected electrode pattern on the photo mask. The substrate was brought as close to the photo mask as possible for better resolution.

Developing electrode pattern

After alignment, all areas of photoresist except electrode pattern image were exposed to UV light (320 nm) for 2.2 seconds (Fig. 2.10). After exposure, reversal baking of 7 minutes 30 seconds at 110 °C was carried out. The unexposed area-the electrode pattern image-still behaved like a normal positive photoresist (Fig. 2.11). After reversal baking, flood exposure (without mask) of 22 seconds was carried out on the substrates. The photoactive compound (PAC) in the exposed area and the special cross-link agent in AZ5214E would lead to an almost insoluble (at development stage) and no longer light sensitive substance [1]. The developer for the substrates was prepared by mixing AZ 400 K with deionized water in the ratio of 1:2. The developing time was (90 ± 10) seconds. The unexposed area (electrode pattern), covered with positive photoresist, was dissolved in developer after flood exposure. The exposed/cross-linked areas remained (Fig. 2.14). The overall result was a negative image of the mask pattern-electrode pattern. The develop time was longer sometimes, to ensure that all the photoresist on the pattern was dissolved in the developer, so that it would not insulate the nanoflake from the electrodes. The finished pattern was checked under microscope (100X) in the clean room.

Figure 2.10: Side view of the substrate with photoresist. UV light exposure for 2.2 seconds, all area of photoresist are exposed, except those covered by the electrode pattern. Nanoflake on Si substrate is coated with photoresist.

Figure 2.11: Side view of the substrate with photoresist. The exposed area is soluble in developer. The unexposed area is still active.

Figure 2.12: Side view of the substrate with photoresist. After image reversal bake at 110°C , the cross-linked agent makes the exposed area insoluble in developer.

Figure 2.13: Side view of the substrate with photoresist. With flood exposure, the previously unexposed areas are exposed to UV light, and are developable in developer.

Figure 2.14: Side view of the substrate with photoresist. Under developer solution, the normal positive photoresist is removed, while photoresist in other area still remained, leaving behind negative image of the electrode pattern.

Chromium (5 nm)/gold (120 nm) were deposited on the wafer covered by the developed resist using magnetron sputtering system with a base pressure of 10^{-8} torr. Before the sputtering process, the high pure argon gas is feed into the chamber up to a

Figure 2.15: JGP 560 Six-Gun Magnetron Sputtering System used for Cr/Au contact deposition.

pressure to 0.2 Pa first. The argon ions are generated by the high voltage on the target. In the DC sputtering a negative potential up to a few hundred volts is applied to the target and the argon ions are accelerated towards the target and hit the material free. In this procedure, the secondary electrons can be produced and they will cause a further ionization of the argon gas. The motion of the ions will be controlled by the magnetic field from the strong magnet under the target so that a semicircle hitting will be formed. The surface atoms on the target are bombarded out and sputtered on the substrate. After a time of growing, a continuous thin film will be formed on the substrate. In insulator or oxide thin film growth, radio frequency (RF) sputtering is used and the temperature will be set to a certain value which can make the thin film crystallized. The thicknesses of electrodes can be controlled by the depositing time.

After the deposition, the substrates with Cr/Au thin films were immersed in acetone to lift off the Cr/Au layer off the pattern. The photoresists remaining on the substrates would dissolve in acetone after 2- 3 minutes. Acetone was used to blow off the unwanted Cr/Au deposition. The substrate was then dried using N₂ gas.

Figure 2.16: SEM image of nanoflake device with Cr/Au electrodes after lift-off procedure.

2.4 Bibliography

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Chapter 3

Indications of surface dominated transport in single crystalline nanoflake devices of topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$

In this chapter, we report experimental evidences in surface dominated transport in single crystalline nanoflake devices of topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ (BSTS). The resistivity measurements show dramatic difference between the nanoflake devices and bulk single crystal. Based on a two-channel model, the analysis on the resistivity and Hall resistance indicates that $\sim 99\%$ surface transport contribution can be realized in 200 nm-thick BSTS nanoflake devices. Using a standard back gate with SiO_2 as a dielectric layer, a pronounced ambipolar electric field effect was observed in devices fabricated with 100–200 nm thick flakes. Moreover, angle-dependent magnetoresistances of a nanoflake device with a thickness of 596 nanometers are fitted to a universal curve for the perpendicular component of the applied magnetic field. The value of phase coherence length obtained from two-dimensional weak antilocalization fitting further confirmed the surface dominated transport. Our results open a path for realization of electric and spintronic devices based on the topological helical surface states.

I will give a brief instruction of this chapter first. Then it is followed by the two channel model for the electron transport in the system with both surface and bulk contributions. The two channel mode will be used for the numerical analysis in the following experiments. Then the bulk single crystal and nanodevice preparation will be briefly introduced and the detailed experiments will be followed.

3.1 Introduction

Topological insulators (TIs) are gapped bulk insulators with gapless Dirac surface states [1–4]. The surface states of these topological insulators are spin polarized and protected by time-reversal symmetry. A number of surface spectroscopy measurements, such as spin and angle-resolved photoemission spectroscopy (ARPES) [5–13] and scanning tunneling microscopy [14,15], have been used to detect the topologically nontrivial surface state in three-dimensional (3D) topological insulator $\text{Bi}_{1-x}\text{Sb}_x$, Bi_2Se_3 , Bi_2Te_3 , etc. The exotic surface states of topological insulators are expected to form a playground of various topological quantum effects and show great potential in spintronics and quantum computation [16–18]. To fulfill the expectations, realizing topological insulator systems with significant surface transport is essential. However, due to the defects or impurities in the samples, it is extremely difficult to eliminate the bulk contribution to electron transport [19–24]. Realizing surface-dominated transport in current topological insulator systems is still a challenge despite extensive efforts involving chemical doping [22,33–39], thin film or

nanostructure fabricating [25–32], and electrical gating [21,26,32,43]. In this chapter, we present strong evidence for surface dominated transport in nanoflake devices fabricated with topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ (BSTS) flakes with a thickness of several hundred nanometers. We performed electron transport measurements of both bulk single crystals and nanoflake devices of high-quality single crystalline BSTS. Nanoflake devices show a transition from semiconductor to metal near 100–150 K with decreasing temperature, while bulk crystals shows semiconductor behavior in the measured temperature range from 300 K to 10 K and only present resistance saturation at very low temperature (<40 K). At 10 K, the resistivity of the nanoflake devices decreases with the sample thickness and can get to hundreds of times smaller than that of the bulk single crystal. Back-gated devices fabricated with 150–200 nm thick BSTS nanoflakes show pronounced ambipolar electric field effect, which demonstrates significant topological surface transport. It was also found that the angle-dependent magnetoresistances of a 596-nanometer thick nanoflake devices are fitted to a universal curve for the perpendicular component of an applied magnetic field. The phase coherence length obtained from a two dimensional weak antilocalization fitting is much smaller than the sample thickness (596 nm), which clearly proves the 2D surface transport in the device. All the experimental results suggest that surface dominated transport has been realized in BSTS devices with a thickness of several hundreds of nanometers.

3.2 A simple Two-channel model for calculating the sample resistivity

Since the conduction of topological insulator originates from both bulk and surface channel, it is essential to figure out the relationship between total conduction and sample thickness.

To calculate the thickness dependence of the resistivity of BSTS single crystal, a simple two- channel model is utilized.

Figure 3.1: Schematic diagram of a two-channel model for resistivity of BSTS nanoflake device.

R_b , R_s , ρ_b , ρ_s , l , w , t_b , $t_s = 0.5$ nm are the bulk resistance, surface resistance, bulk resistivity, surface resistivity, sample length, sample width, the thickness of the bulk channel, the thickness of the surface channel, respectively.

$$R_b = \frac{\rho_b \cdot l}{w \cdot t_b} \quad (1)$$

$$R_s = \frac{\rho_s \cdot l}{w \cdot t_s} \quad (2)$$

$$R = \frac{1}{\frac{1}{R_b} + \frac{1}{R_s}} = \frac{1}{\frac{1}{\frac{\rho_b \cdot l}{w \cdot t_b}} + \frac{1}{\frac{\rho_s \cdot l}{w \cdot t_s}}} = \frac{\rho_s \cdot \rho_b \cdot l}{(t_s \cdot \rho_b + t_b \cdot \rho_s) \cdot w} \quad (3)$$

$$\rho = \frac{R \cdot w \cdot (t_s + t_b)}{l} = \frac{\rho_s \cdot \rho_b \cdot (t_s + t_b)}{t_s \cdot \rho_b + t_b \cdot \rho_s} \quad (4)$$

To accurately determine the thickness and width of the nanoflake devices, atomic force microscopy (AFM) and scanning electron microscopy (SEM) were used, respectively. The microscopy was performed after the transport measurements.

The conductance contribution of the surface state can be easily calculated as

$$\sigma_b = \frac{W \cdot t_b}{\rho_b \cdot l} \quad \sigma_s = \frac{W \cdot t_s}{\rho_s \cdot l} \quad (5)$$

$$\frac{\sigma_s}{\sigma_s + \sigma_b} = \frac{\frac{W \cdot t_s}{\rho_s \cdot l}}{\frac{W \cdot t_s}{\rho_s \cdot l} + \frac{W \cdot t_b}{\rho_b \cdot l}} = \frac{1}{1 + \frac{\rho_s \cdot t_b}{\rho_b \cdot t_s}} \quad (6)$$

Suppose $t_b = 200$ nm, $t_s = 5$ nm, the surface contribution can be easily calculated. The value is 98.9% if $\rho_b = 8.55 \, \Omega \cdot \text{cm}$ and $\rho_s = 2.27 \times 10^{-3} \, \Omega \text{cm}$.

For calculating two channel mode Hall effect, we use Drude model:

$$m \frac{d\vec{v}}{dt} = -e(\vec{E} + [\vec{v} \times \vec{B}]) - \frac{m\vec{v}}{\tau} \quad (7)$$

$$\vec{j} = ne\vec{v} \quad (8)$$

At steady state:
$$e\vec{E} = -e[\vec{v} \times \vec{B}] - \frac{m\vec{v}}{\tau} \quad (9)$$

$$\vec{E} = -\frac{1}{ne} [ne\vec{v} \times \vec{B}] - ne\vec{v} \cdot \frac{m}{ne^2\tau} \quad (10)$$

Define
$$R_H = \frac{1}{ne}, \text{ and } \rho = \frac{m}{ne^2\tau} \quad (11)$$

We obtain
$$\vec{E} = \rho \cdot \vec{j} - R_H (\vec{j} \times \vec{B}) \quad (12)$$

When the magnetic field $\vec{B}$ points to z direction, we obtain

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix} = \begin{pmatrix} \rho & -R_H H \\ R_H H & \rho \end{pmatrix} \begin{pmatrix} j_x \\ j_y \end{pmatrix} \quad (13)$$

Then the conductivity

$$\hat{\sigma} = \begin{pmatrix} \rho & -R_H H \\ R_H H & \rho \end{pmatrix}^{-1} = \frac{1}{\rho^2 + (R_H H)^2} \begin{pmatrix} \rho & -R_H H \\ R_H H & \rho \end{pmatrix} \quad (14)$$

For more than one channel transport,

$$\hat{j} = \sum_i \hat{\sigma} \vec{E} \quad (15)$$

Using $\frac{1}{\rho} = ne\mu$ and $\frac{R_H}{\rho} = \mu$,

$$(16)$$

We obtain

$$\hat{\sigma} = \begin{pmatrix} \sum_i \frac{ne\mu_i}{1+\mu_i^2 B_i^2} & \sum_i \frac{ne\mu_i^2}{1+\mu_i^2 B_i^2} \\ \sum_i \frac{ne\mu_i^2 B_i}{1+\mu_i^2 B_i^2} & \sum_i \frac{ne\mu_i^2}{1+\mu_i^2 B_i^2} \end{pmatrix} \quad (17)$$

Using $\hat{\rho} = \hat{\sigma}^{-1}$

$$(18)$$

For a two channel system, we obtain

$$\rho_{xy}(B) = -\frac{B (n_1\mu_1^2 + n_2\mu_2^2) + B^2\mu_1^2\mu_2^2(n_1 + n_2)}{e (n_1\mu_1 + n_2\mu_2)^2 + B^2\mu_1^2\mu_2^2(n_1 + n_2)^2} \quad (19)$$

The formula is for two dimensional system channel, if channel 2 is a two dimensional transport channel (thickness=0), the formula should be written as,

$$\rho_{xy}(B) = -\frac{B (n_b\mu_b^2 + n_s\mu_s^2 / t) + B^2\mu_b^2\mu_s^2(n_b + n_s / t)}{e (n_b\mu_b + n_s\mu_s / t)^2 + B^2\mu_b^2\mu_s^2(n_b + n_s / t)^2} \quad (20)$$

which is the equation in the main text. Here, t is the sample thickness.

Equation 18 is clear for understanding physics concept. However, n_b, n_s, μ_b and μ_s are very large numbers. It is difficult to use equation 18 for curve fitting. By using equation 14 or derive from equation 12 directly, we transfer equation 18 into another form:

$$\rho_{xy}(B) = -\frac{(R_b\rho_s^2 + R_s\rho_b^2) + B^2R_b^2R_s^2(R_b + R_s)}{(\rho_b + \rho_s)^2 + B^2(R_b + R_s)^2} \quad (21)$$

Here, R_b and ρ_b are the Hall coefficient and resistivity of the bulk electrons.

$R_s = t / en_s$ and $\rho_s = \rho_{sheet} / t$, where ρ_{sheet} is the surface sheet resistance.

3.3 Experiment

High quality BSTS single crystals were grown using modified Bridgeman methods. High purity (99.9999%) Bi, Sb, Te, and Se with a molar ratio of 1.5:0.5:1.8:1.2 were first thoroughly mixed and then reacted at 950°C for 1 week in an evacuated quartz tube in a box furnace. We then located the quartz tube vertically in a specially designed furnace with large temperature gradient. The temperature is then decreased to room temperature over three weeks, with different cooling speed in different temperature regions. The obtained crystals are easily cleaved and revealed a flat and big shiny surface as shown in the inset of Fig. 3.2(a). An as grown single crystal is a cylinder with $\sim 0.7 \text{ cm}^2$ (the cross section area) $\times 2 \text{ cm}$ (height). Large cleaved crystals can be cut from different positions of the crystal.

The X-ray diffraction pattern indicates the high quality of our samples. Fig. 3.2(a) shows the wide angle x-ray diffraction from a bulk crystal oriented with the scattering vector perpendicular to the (001) family of planes. No peaks from other plane families can be observed. It should be noted that the count of the (006) peak is larger than 300 k. Only high quality single crystals can get to this value in XRD facility. The inset of Fig. 3.2(a) shows the large and shining (001) surface of the single crystal. Energy dispersive X-ray spectroscopy (EDS) was employed to probe the homogeneity of the samples. The small area EDS mapping as shown in Fig. 3.2(b)

indicates the element distribution in a small area is homogeneous. We also performed the EDS measurements at various points of the bulk single crystal used in our measurements. As shown in Fig. 3.2(c), the molar ratio of the 4 elements at various points of the big crystal only shows small fluctuations in the $\sim 8 \text{ mm} \times 8 \text{ mm}$ single crystal, which means a large crystal cut from the same position is quite homogeneous. Small crystals cut from this $8 \text{ mm} \times 8 \text{ mm}$ big crystal show very similar transport behaviors.

Figure 3.2: Summary of structural characterizations. (a) shows the wide angle x-ray diffraction from a bulk crystal oriented with the scattering vector perpendicular to the (100) family of planes. Inset is a photograph of a typical crystal. (b) The EDS mapping of a small area in a BSTS single crystal. (c) Molar ratio of Bi, Sb, Te, and Se at ten random positions in a big crystal like the one shown in the inset of (a). (d) A typical EDX spectroscopy. The insert is a schematic diagram of an as grown single crystal.

The molar ratio of (Bi+Sb)/(Se+Te) and Bi/Sb in crystals cut from different positions of the crystal cylinder shows near-constant molar ratios of 2/3 and 3/1, respectively, while the Te/Se ratio changes from $\sim 1.7/1.3$ to $\sim 1.9/1.1$ for crystals obtained from different positions. Transport measurements show that, although the molar ratio of Te/Se varies with position as shown in the inset of Fig. 3.2(d), all the crystals show similar semiconducting $\rho(T)$ curve at high temperature and saturation behavior at low temperature and large resistivity ($>1 \text{ } \Omega\text{cm}$) at 10 K. The value of $\rho(10\text{K})/\rho(300\text{K})$ can vary from 30 to 250. This characteristic of BSTS is very different from the condition of grown Bi_2Te_3 single crystals. Bi_2Te_3 single crystals obtained from different part of the cylinder usually show different transport behaviors, from metallic to semiconducting. We choose BSTS crystals with $\rho(10 \text{ K})/\rho(300 \text{ K}) > 100$ and $\rho(10 \text{ K}) > 5 \text{ } \Omega \cdot \text{cm}$ for our experiments.

Figure 3.3 are the measurement results of this device. The thickness of the nanoflake used in this device is 596 nm. There are four steps [Fig 3.3(a)] from the bottom to the top in the circled region. The thickness of the sample is the total height of the four steps as shown in the AFM image.

For the transport measurements on bulk samples, the contact were made with silver paste and cured at room temperature. Photolithography was used to pattern electrodes on the nanoflake devices. Cr/Au (5 nm/120 nm) contacts were deposited in a magnetron sputtering system with a base pressure of 1×10^{-8} torr. The typical devices are shown in Figs. 3.4(g) and 3.4(h). Standard lock-in technique was employed in

performed four-terminal magnetoresistance and Hall measurements in a 9 Tesla Quantum Design PPMS system. All the transport measurements were performed with the applied current in the (001) plane.

Figure 3.3: Image of typical device for weak antilocalization, angle dependent magnetoresistance measurements and universal fluctuation conductance measurements.

Figure 3.4. (Color online) Temperature dependence of resistivity in zero field of (a) bulk single crystal with a thickness of $102\ \mu\text{m}$ and (b) nanoflake devices with various thicknesses. (c) The resistivity values of nanoflake devices and bulk crystals with various thicknesses. The red line is the fitting curve based on the two-channel model. (d) The zoom in of Fig. 3.3(c). It shows the thickness of the resistivity of nanoflake devices. (e) The Hall measurement results of the $102\text{-}\mu\text{m}$ thick BSTS single crystal and the fitting (the red curve) based on a two-channel model. (f) The Hall resistance of a BSTS nanoflake device with a thickness of 134 nm. The red line is a drawn straight for comparison. (g) and (h) The image of two typical devices for transport measurements.

3.4 RESISTIVITY, HALL RESISTANCE, AND AMBIPOLAR ELECTRIC GATING EFFECT

The temperature dependence of the resistivity of the bulk single crystal and three nanoflake devices measured in zero field are shown in Figs. 3.4(a) and 3.4(b), respectively. The 102- μm thick bulk crystal shows semiconductor behavior in the measured temperature regime and reaches a saturation behavior when $T < 40$ K, which is due to the increasing transport contribution from surface[37,38]. The resistivity at 10 K is more than 200 times larger than that at room temperature. Although the nanoflake devices also show the semiconductor behavior in the high-temperature range, the transport characteristics transfer to a metallic type when $T < 150$ K. It should be noted from Figs. 3.4(a) and 3.4(b) that the resistivity of nanoflakes are hundreds of times smaller than that of the bulk single crystal although the nanoflake is exfoliated from the bulk crystal used in the transport measurement [Fig. 3.4(a)]. As shown in the inset of Fig. 3.4(b), the resistivity of eight nanoflake devices with varying thickness smaller than 600 nm and the nine bulk single crystals with thickness between 1 μm to 157 μm at 10 K decreases with decreasing sample thickness. The eight nanoflake devices and 102- μm thick bulk sample are obtained from the same piece of single crystal, while the other eight bulk samples are obtained from another piece of single crystal. From the systematic decrease of resistivity with decreasing sample thickness and the energy-dispersive x-ray spectroscopy (EDS) as shown in Fig.

3.2, we can conclude that the dramatic decrease of resistivity with decreasing sample thickness is not due to chemical inhomogeneity. As the material system has already been proved to be a topological insulator[36,37], the dramatic decrease of resistivity with decreasing sample thickness indicates that the contribution from the surface states plays a more and more important role as the thickness reduces. As shown in Fig. 1, supposing that the total thickness of the two surface states is 5 nm, we use a simple model to fit the resistivity of the surface states and bulk. The formulas are shown below,

$$R = \frac{1}{\frac{1}{R_b} + \frac{1}{R_s}} = \frac{1}{\frac{1}{\frac{\rho_b \cdot l}{w \cdot t_b}} + \frac{1}{\frac{\rho_s \cdot l}{w \cdot t_s}}} = \frac{\rho_s \cdot \rho_b \cdot l}{(t_s \cdot \rho_b + t_b \cdot \rho_s) \cdot w}$$

$$\rho = \frac{R \cdot w \cdot (t_s + t_b)}{l} = \frac{\rho_s \cdot \rho_b \cdot (t_s + t_b)}{t_s \cdot \rho_b + t_b \cdot \rho_s}$$

where ρ , ρ_b , ρ_s , t_b , t_s , are the resistivity of the sample, the resistivity of the bulk part, the resistivity of a 5 nm thick surface state, the thickness of the bulk, and the thickness of the surface state (5 nm), respectively. The fitting curve is shown as the red line in the inset of Fig. 3.4(c), which yields $\rho_b = 8.036 \text{ } \Omega\text{cm}$ and $\rho_s = 2.19 \times 10^{-3} \text{ } \Omega\text{cm}$. For a device with a thickness of 200 nanometers, the conductance contribution of the surface states is 98.9% at 10 K. Fig. 3.4(d) is the zoom in of Fig. 3.4(c), which shows the resistivity of the eight nanoflake devices more clearly. As shown in Fig. 3.4(e), the Hall measurement of the single crystal (102 μm) is also fitted using a standard two-band model [32,38] (details in section 3.2),

$$\rho_{xy}(B) = -\frac{B (n_b \mu_b^2 + n_s \mu_s^2 / t) + B^2 \mu_b^2 \mu_s^2 (n_b + n_s / t)}{e (n_b \mu_b + n_s \mu_s / t)^2 + B^2 \mu_b^2 \mu_s^2 (n_b + n_s / t)^2}$$

where ρ_{xy} , B , n_b , n_s , μ_b , μ_s , and t are the Hall resistivity, magnetic field, bulk charge density, surface charge density, bulk mobility, surface mobility, and sample thickness, respectively. The fitting yields $n_b = 1.1 \times 10^{17} \text{ cm}^3$, $n_s = 2.5 \times 10^{12} \text{ cm}^2$, $\mu_b = 25 \text{ cm}^2/\text{V s}$, $\mu_s = 1767 \text{ cm}^2/\text{V s}$. Based on the fitting results, the surface conductance contribution of a 200-nm thickness sample is 98.7%, which agrees well with the results obtained from resistance measurements. It should be noted that the results obtained from the fitting of Hall resistivity has certain arbitrariness due to the multiple free parameters. To relieve the arbitrariness and further prove the surface dominated transport in nanoflakes with a thickness of hundred nanometers, we measured the Hall resistance of a nanoflake device (134-nm thick) from 9 T to -9 T. As shown in Fig. 3.4(f), the shape of Hall resistance vs magnetic field curve is very different from that of the bulk sample in Fig. 3.4(e). The Hall resistance of the nanoflake is always linear over the entire range of magnetic field, indicating all channels contributing to the transport have similar mobility and the same carrier sign. The transport channels of nanoflake devices include the top and bottom surfaces, the side surfaces, and the bulk channel. As it is known that the mobility of the bulk channel of BSTS is much smaller than that of the surface transport channels, from the perfect linear behavior of the Hall resistance, it can be concluded that the bulk transport is negligible in the nanoflake device at 2 K. Otherwise, the Hall resistance vs magnetic field curve will show clear curvature due to the transport channels with very different mobility. The linear Hall

resistance also shows that thermal ability of electrons on the top, side, and bottom surface are similar, which indicates that the device fabrication process does not significantly change the mobility of the top and side surfaces of BSTS samples. The Hall resistance of the nanoflake device is $207 \text{ } \Omega/T$, which corresponds to a Hall carrier density of $3.01 \times 10^{12} \text{ cm}^{-2}$. The sheet resistance of the device at 2 K is $1260 \text{ } \Omega$, therefore the mobility of the surface transport can be calculated as $1647 \text{ cm}^2/\text{V s}$. Both the charge density and mobility of the surface state obtained from the direct measurements agrees well with the value obtained by the fitting on the bulk Hall measurements [Fig. 3.4(e)], which further support the $\sim 99\%$ surface transport in a BSTS device with a thickness of 200 nm.

Figure 3.5: (a) The gate voltage dependence of resistance (R_{xx}) and Hall resistance (R_{xy}) at 1 Tesla filed of a BSTS nanoflake device at 2 K. (b) The temperature evolution of the $R_{xx}(V_g)$ curve.

If the electron transport is mainly due to the topological surface transport, we should be able to observe the ambipolar electric effect even in devices fabricated with BSTS nanoflakes with a thickness of several hundred nanometers, which is indeed the case in our experiments. Figure 3.5 shows the gatevoltage dependence of longitudinal resistance (R_{xx}) without applied magnetic field and Hall resistance (R_{xy}) at 1 Tesla magnetic field of a 172-nm thick device. To eliminate any possible nonsymmetry effect of the sample and electrode contacts, all the Hall resistances were obtained from low field Hall measurement with magnetic field scanning from -1 Tesla to 1 Tesla. As the sample is 172-nm thick, the electrostatic gating can only shift the Fermi level of the bottom surface. The key finding is the resistance maxima appears near $V_g = -10$ V. The resistance change near 35% with ± 60 V applied gate voltage. The quite sharp charge neutrality point at 2 K and the gradual decrease of resistance with V_g deviating from the charge neutrality point indicate that the ambipolar behavior should be due to the Dirac cone of the topological surface state, which has been observed by ARPES[36]. If it were due to the normal band banding, there would be a wide constant resistance region when the Fermi level is in the band gap and a much dramatic resistance decrease when the Fermi level enters into conduction or valence band. The Hall resistance can also be changed from n type to p type with applied gate voltage as shown in Fig. 3.5(a), which further confirms the ambipolar electric gating effect in this device. From the measured Hall resistance R_H ($150 \Omega/T$ at 2 K), if we neglect the Hall contribution from the bulk, the charge density can be estimated to be

about $4.2 \times 10^{12} \text{ cm}^{-2}$ (bottom + top), which approximately agrees with the bulk Hall fitting results as aforementioned. For a 300-nm SiO_2 dielectric layer, a gate voltage of 60 volts can induces $n = 4.4 \times 10^{12} \text{ cm}^{-2}$, therefore the 300 nm SiO_2 dielectric layer should be able to generate $\sim 50\%$ resistance change because we can only tune the bottom surface state. However, considering the 176-nm thick side surface of the devices, which can contribute $\sim 5\text{--}10\%$ surface conductance contribution, and the small amount of charge from the bulk, it is very reasonable that we only get $30\text{--}40\%$ resistance change. It should be pointed out that we have observed similar ambipolar behavior in many devices fabricated with BSTS flakes with a thickness of several hundred nm. Figure 3.5(b) shows the gate voltage dependence of resistance at various temperatures. It is observed that the shape of the $R(V_g)$ curves become more broad with increasing temperature, which is a standard ambipolar behavior as shown in graphene [40]. It also agrees well with the decreasing surface conductance contribution with increasing temperature. The shift of resistance peaks with increasing temperature indicates the shift of chemical potential of the bottom surface with increasing temperature.

The ambipolar gating effect of a device stored in vacuum for two months is shown in Fig. 3.6, which indicates that the charge neutrality point shifts to near -80 V and it disappears when the temperature is higher than 50K . From the Hall Effect results, we can conclude that around 40% of concentration has been enhanced after the long time store. Such changes can be explained by the surface contamination effect. Although

Figure 3.6. (Color online) (a) The $R_{xx}(V_g)$ curves of a BSTS nanoflake devices at various temperatures. The devices had been stored in vacuum for two months. The peak shifts to 80 volts. (b) The low field Hall resistance of the device.

sample were stored in the vacuum with pressure of 10^{-8} Torr, the limited molecular like oxygen and other atoms are still able to be absorbed by the surface of the device which highly enhance the carrier concentration. Therefore, the topological surface states are changing with time even in the vacuum condition. In spite of this, we are still able to observe the ambipolar behavior due to the two dimensional electron gases and it shows the reliability of BSTS. Moreover, to make sure of the reproducibility of the observed ambipolar effect, another batch of devices were prepared. We perform the results of two devices (D4 and D5) in Fig 3.7 and Fig 3.8.

Similar gating voltage dependent resistance behavior appears in both two samples at 2 K. Sample D4 doesn't show the sharp peak in the ambipolar curve and the transition voltage is around -15V. However, the sharp peak appears in Sample D5 and locates at

Figure 3.7: (a) The weak antilocalization curves of a 198 nm thick devices (D4) fitted to a universal curve when the x axis is the perpendicular part of the magnetic field. (b) The weak antilocalization curve of the device at various temperatures.

-30V. The difference of the ambipolar behavior must be from the different conditions of the device surfaces. The temperature dependent resistances of these two samples are also shown in Fig 3.7(b) and Fig 3.8(b) which agree with other nanodevices we obtained. The two $R(T)$ curves show the similar behavior.

Figure 3.8: (a) The $R(V_g)$ curves of a 212 nm thickness BSTS nanoflake device (D5) at 2 K. (b) The $R(T)$ curve of the device.

Based on above repeatable experiments, we conclude that the ambipolar behavior we obtained in the BSTS nanodevice is reliable.

3.5 Weak Antilocalization

To further confirm the surface-dominated electron transport in the nanoflake device, we have performed the angle-dependent magnetoresistance measurements using a very thick (596 nm) nanoflake device. To eliminate any effect from Hall resistance, all the magnetoresistance measurements were performed from -2 Tesla to 2 Tesla and carried out corresponding calculation process to obtain intrinsic magnetoresistance. The magnetoresistance was measured by tilting the bulk crystal and nanoflake device with respect to the applied magnetic field from 0° to 90° . $\theta = 0^\circ$ means the magnetic field parallel to the (001) surface while the $\theta = 90^\circ$ means the magnetic field perpendicular to the (001) surface. Figures 3.9(a)–(d) show the field dependence of magnetoconductances $G = G(B) - G(0)$ with various θ for the bulk single crystal at 2 K and nanoflake device at 2 K, 10 K, and 30 K, respectively. Both the bulk crystal and nanodevice show weak antilocalization behavior in the low magnetic field region. Such a weak antilocalization at low temperature has been attributed to both strong spin-orbital coupling and topological π Berry phase of two-dimensional surface states[41,46]. It is very obvious that G decreases with decreasing θ for both bulk single crystal and nanoflake device and it is more pronounced for the nanoflake device. The variation of magnetoconductance as a function of the perpendicular

component of the magnetic field for the bulk single crystal at 2 K and nanoflake device at 2 K, 10 K, and 30 K are shown in Figs. 3.9(e)–3.9(h), respectively. The G vs $B \sin \theta$ (the perpendicular component of the applied field) curves of the nanoflake device are perfectly merged into one universal curve at all temperatures. It should be noticed that a misalignment between the sample and applied magnetic field has a large effect when θ is near 0 while it has a negligible effect when θ is a large value, which can be inferred from the small difference between the $\theta = 80^\circ$ and $\theta = 90^\circ$ curves and the large difference between $\theta = 0^\circ$ and $\theta = 5^\circ$ curves. Based on this, we speculate that the very small magnetoconductance at $\theta = 0^\circ$ may be due to the small unavoidable misalignment between the sample and the applied magnetic field in experiment. Therefore, the perfect fitting into a universal curve for all θ value (except $\theta = 0^\circ$) as shown in Figs. 3.9(f)–3.9(h) is a support of the conclusion obtained from the resistivity and Hall measurements, surface-dominated transport (96.9% in a 596-nm thick device). For the bulk single crystal, it is not a surprise that the weak antilocalization is observed for all the angles and G vs $B \sin \theta$ curve cannot be fitted to one universal curve because of the coexistence of both the bulk spin-orbit coupling effects and helical surface state contribution.

Magnetoconductances ΔG at different temperatures (2 K, 4 K, 7 K, 10 K, 15 K, 35 K, and 45 K) have been obtained in a magnetic field perpendicular to the (001) plane ($\theta = 90^\circ$), as shown in Fig. 3.10(a) and 3.10(b) for bulk single crystal and nanoflake device, respectively. The Hikami-Larkin-Nagaoka (HLN) formula

Figure 3.9. (Color online) Angle-dependent conductance of a bulk BSTS single crystal and a nanodevice fabricated with a 596-nm thick BSTS flake. θ is the angle between the direction of the magnetic field and the (001) plane. (a), (b), (c), and (d) show the magnetoconductance of bulk BSTS single crystal at 2 K, nanoflake device at 2 K, 10 K, and 30 K, respectively. (e), (f), (g), and (h) show the curves of magnetoconductance vs the perpendicular component of the magnetic field of bulk BSTS single crystal at 2 K, nanoflake device at 2 K, 10 K, and 30 K, respectively.

$$\Delta\sigma_{2D} = \left[-\frac{\alpha e^2}{2\pi^2 \hbar} \right] \left[\ln\left(\frac{\hbar}{4eL_\phi^2 B}\right) - \Psi\left(\frac{1}{2} + \frac{\hbar}{4eL_\phi^2 B}\right) \right]$$

is used to fit the magnetoconductance observed in both bulk and nanoflake device, where $\Delta\sigma_{2D}$ is the two dimensional conductivity ($\Delta\sigma_{2D} = \frac{\Delta G \cdot W}{L}$, W and L are the width and length of the sample, respectively.) Ψ is the digamma function, L_ϕ is the phase breaking length, and α is a prefactor which contains information about the nature of the electrons in topological insulators [42, 46]. The fitted curves are plotted in Fig. 3.10(a) and 3.10(b) in solid lines. Due to the three dimensional bulk contributions, the fitting curves do not agree well with experimental data for bulk single crystal samples. For the nanoflake device, the two dimensional fitting curves agree very well with the experimental data for both fitting range from 0 to 2 Tesla and from 0 to 0.5 Tesla at all temperatures from 2 K to 45 K. The fitting results of α and L_ϕ of nanoflake device in both fitting ranges are shown in Fig. 3.13(c) and 3.13(d) respectively. It is clear that similar results are obtained from different fitting ranges. The power law fit gives $L_\phi \propto T^{-0.66}$ and $L_\phi \propto T^{-0.19}$ (for the fitting of 0-0.5 Tesla field range) in the temperature regime 10 K-40 K and 2 K -7 K, respectively, which indicates the effect of phonon scattering at high temperature and paramagnetic impurity at low temperature [44, 45].

It should be noted that the fitted L_ϕ at all temperatures are much smaller than the thickness of the sample (596 nm), which strongly indicates that the two dimensional electron transport characteristics are due to the surface states. From this point of view,

Figure 3.10: Temperature-dependent magnetoconductance of the BSTS single crystal and 596 nm thick nanoflake device with $\theta = 90^\circ$. (a) Magnetoconductance of the bulk single crystal at different temperatures. The HNL fitting curves are plotted in solid lines. (b) Magnetoconductance of the nanoflake device at different temperatures. The HNL fitting curves are plotted in solid lines. (c) The fitted values of α at different temperatures for the nanoflake device in the fitting range of 0 - 2 Tesla and 0 - 0.5 Tesla, respectively. (d) The power-law fit gives $L\phi \propto T^{-0.66}$ and $L\phi \propto T^{-0.19}$ (0.5 Tesla) in the temperature regime 10–45 K and 2–7 K, respectively

our experiment is very different from previous measurements on ultra-thin Bi_2Se_3 and Bi_2Te_3 films. The two dimensional transport behaviors in ultra-thin Bi_2Se_3 and Bi_2Te_3 films cannot be completely attributed to the surface states, because the L_ϕ is larger than the thickness of the film and therefore the film itself, including the bulk and surface states, form a two dimensional transport system. Since the thickness (596 nm) of our nanoflake BSTS device is much larger than L_ϕ (~ 180 nm at 2 K, ~ 110 nm at

10 K, and ~ 60 nm at 30 K), it is a three dimensional transport system. The two dimensional transport behavior of weak antilocalization can only originate from the helical surface states. The coefficient α takes a value of $-1/2$ for a traditional 2D electron system with strong spin-orbit coupling and one helical surface with a single Dirac cone. Since carriers on both up and bottom surface can contribute the conduction in topological insulator samples, ideal double transport channel should add give $\alpha = -1$. As shown in Fig. 3.10(c), the perfect fitting using HLN formula generates α values between -0.7 to -0.8 at various temperatures, which also supports the 2D surface transport.

As a supportive study, Another device (D5) with a thickness of 142 nm was also performed to further confirm the above conclusion. Not surprisingly, the weak antilocalization is observed for all the angles in this device and G vs. $B \sin \theta$ curves are perfectly fitted to one universal curve as shown in Fig 3.11(a). Moreover, the temperature dependent weak antilocalizations were also performed where the $\theta = 90^\circ$ as shown in Fig 3.11(b). At 2 K, the fitted value of L_ϕ is 194 nm and the value of α is -0.68 . Therefore, all the results show that the ambipolar behavior and WAL effects appear in all the nanodevices and it strongly supports the existence of the topological surface states.

Figure 3.11: (a) The $R(V_g)$ curves of a 142 nm thickness BSTS nanoflake devices at 2 K. (b) The $R(T)$ curve of the device.

3.6 Time dependent Universal Conductance Fluctuation

We also observed a novel conductance fluctuation phenomenon in the nanoflake device under both low and high magnetic fields. The low magnetic field conductance fluctuation is very clear as shown in Fig. 3.9(b) and 3.9(d). Fig. 3.12(a) shows the

conductance fluctuations at different temperatures below 10 K between 4 and 9 Tesla perpendicular fields. Different from normal universal conductance fluctuations (UCF) discovered in topological insulator Bi_2Se_3 [22, 31], the conductance fluctuation in our nanoflake device evolves with time. Such time-dependent UCF can only be observed below 10 K and the magnitude of those fluctuations increases with decreasing temperature. The top 3 curves shown in Fig. 3.11(a) were measured continuously with different field sweeping direction. As shown in the circled region in the figure, the curves show very similar fluctuation behaviors in the same magnetic field region because the time interval between the measurements is short. With the time evolution, the fluctuation patterns become different in the same magnetic field region. Time-dependent UCF is due to the sensitivity of the conductance to the motion of individual scatters and such phenomena have been reported. The variation of conductance pattern is due to the motion of scattering sites which changes the interference pattern of all the intersecting electronic paths in the coherent volume of the scattering sites. Since the motion of scattering sites is related to the time, the observed fluctuations are also time-dependent which allows the non-retraceable results. As the electron transport in the nanoflake is almost fully surface transport as discussed before, the motion of the scattering site might be due to the time dependent surface contamination in the measurement chamber. Each magnetoresistance curve from 4 to 9 Tesla takes about 1 hour. According to the results, we can conclude that the scattering site pattern on the topological surface changes after about 30 minutes.

The amplitude of fluctuation detected is around $0.2e^2/h$. The phase coherence length of electron L_ϕ observed at 2 K is $\sim 0.18 \mu\text{m}$ (Fig. 3.10) and the thermal length, $L_T = \sqrt{\hbar D / k_B T}$ is around $1 \mu\text{m}$ at 2 K. As $L_\phi < L_T < L$, the fluctuation of δG can be calculated as $\delta G = e^2 / h [L_\phi / L]^{4-d}$, where L is the sample size. The calculated δG is $0.3e^2/h$ which is quite close to our experiment data $0.2e^2/h$ at 2 K. Furthermore, the root-mean-square (rms) of δG vs. temperature curve is also plotted as shown in Fig. 3.12(b). The rms of δG is usually following the power law $\text{rms}\delta G \propto T^{-0.5}$ for two dimensional systems. Our fitting shows $\text{rms}\delta G \propto T^{-0.43}$, which also indicates that the time-dependent UCF observed in the BSTS device sample is mainly from the surface states.

Figure 3.12: (a) Magnetofingerprint (UCF) signal δR vs magnetic field for the nanoflake device. At 2 K, we scan the field for three times with alternative field sweeping direction (the arrows). Seven UCF curves measured at temperatures between 2 K to 10 K were shown in the figures. (b) Temperature dependence of root mean square of the fluctuation of conductance. The solid line is the fitting curve using power law and the fitted power is -0.43.

3.7 Summary

In conclusion, we demonstrate surface-dominated transport in single crystalline nanoflake devices of topological insulator BSTS. The analysis on the resistivity and Hall resistance based on a two-channel model indicates that $\sim 99\%$ of the surface transport contribution can be realized in 200-nm thick BSTS devices. The pronounced electric gated ambipolar behavior of BSTS devices with a thickness of several hundred nanometers proves the dominated topological surface transport. Moreover, the angle-dependent weak antilocalization effect of bulk single crystals and nanoflake devices strongly suggest the surface dominated transport in a nanoflake device, which is further confirmed by the fact that the phase coherence length $L\phi$ obtained by 2D HNL fitting is much smaller than the thickness of device.

3.8 Bibliography

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Chapter 4

Anisotropic magnetoresistance in topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ /CoFe heterostructures

As discussed in the previous chapter, the pronounced electrical field ambipolar effect has been realized in gated devices fabricated by 100 - 200 nm thick nanoflakes exfoliated from $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ crystals. We also observed 2D weak antilocalization behavior in a 596 nm thick $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ single crystalline flake. These results indicate that dominated surface transport can be realized in $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ single crystalline flakes with a thickness of several hundred nanometers. These materials offer a well characterized playground for studying the topological surface state.

There are a few ways to study the spin polarization properties of topological surface state, such as Andreev reflection between superconductor and TI surface states, tunneling magnetoresistance study and anisotropic magnetoresistance study. Here, we report a novel in-plane anisotropic magnetoresistance in topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ /CoFe heterostructures. To explain the novel effect, we propose that the $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ /CoFe heterostructure forms a spin-valve or Giant magnetoresistance device due to spin-momentum locking. The novel in-plane

anisotropic magnetoresistance can be explained as a Giant magnetoresistance effect of the $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}/\text{CoFe}$ heterostructures.

Topological insulator/ferromagnet interface is a potentially very interesting system to investigate. Various novel devices based on ferromagnets and topological insulators have been proposed in recent years[1-6]. For example, spin and electron transport on topological insulators with ferromagnetic electrode has been studied theoretically[1]. Inverse spin-galvanic effect has been predicted to occur at the interface between a topological insulator and a ferromagnet[2]. The thin film heterostructure based on topological insulators and ferromagnet was believed to generate giant magneto-optical Kerr effect and universal Faraday effect[4]. However, as far as we know, the transport experiment on topological insulator/ferromagnet has never been attempted so far. In this work, we measured the magnetoresistance of a $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}/\text{CoFe}$ heterostructure. We observed unusual in-plane anisotropic magnetoresistance effect that is determined by the angle θ between the applied magnetic field and the current direction. With a model based on Giant magnetoresistance (GMR) effect due to the peculiar characteristic of the surface states of topological insulator, the spin-momentum locking, we successfully explain the novel in-plane anisotropic magnetoresistance. In this model, the two spin polarized electron transport channels are the spin polarized topological surface states and the ferromagnetic CoFe layer.

4.1 Huge bulk resistivity-Bi_{1.5}Sb_{0.5}Te_{1.8}Se_{1.2} and BSTS/CoFe structure

High quality single crystals of Bi_{1.5}Sb_{0.5}Te_{1.8}Se_{1.2} (BSTS) were grown by a modified Bridgeman method. High purity (99.9999%) Bi, Sb, Te, and Se with a molar ratio of 1.5:0.5:1.8:1.2 were first melted at 850°C in an evacuated quartz tube which was located vertically in a specially designed furnace with large temperature gradient. The temperature was then decreased to room temperature over 3 weeks. The characterization of structure and chemical homogeneity of these crystals are shown in Ref. [7]. The cleaved crystals were characterized by standard four contact electrical transport measurements. Ohmic contacts were made by room temperature curved silver paste. The magnetoresistance was measured on a horizontal rotator of a Physical Property Measurement System (PPMS, Quantum design). To study the exotic helical surface of the topological insulator with significant surface transport, 5 nm CoFe (Co_{0.9}Fe_{0.1}) and 3 nm Al₂O₃ protecting layer were sputtered on (001) surfaces of 50-100 μ m thick single crystals at room temperature in a chamber with base pressure of 1×10^{-8} Torr. Standard four contact electrical transport measurements were employed to measure the magnetoresistance of the BSTS/CoFe heterostructures. Both the current and magnetic field were applied in the (001) plane. The BSTS single crystals and BSTS/CoFe interface were examined by high resolution transmission microscopy (HRTEM).

We have measured the $R(T)$ curve of more than 15 pieces of 50-100 μm thick cleaved single crystals. All of them show similar $R(T)$ behaviors. The resistance increases with decreasing temperature at high temperature regime, which indicates that the Fermi level is in the band gap. Only the $R(T)$ curves of three samples are plotted in Fig. 1. We name the samples as S1, S2 and S3. The resistivity at low temperature of every sample is higher than 1 Ωcm . Based on the recent electric transport results, the topological surface conductance contribution can get to $\sim 10\%$ in a $\sim 100 \mu\text{m}$ thick BSTS sample at 10 K.[7-8] As shown in the inset of Fig. 4.1, the temperature dependence of the magnetoresistance of the sample S3 changes from weak

Figure 4.1: Temperature dependence of the resistivity of four BSTS single crystals. The inset shows the temperature dependence of the magnetoresistance of sample S3.

Figure 4.2: (a) and (d) the high resolution cross section TEM of CoFe/BSTS. (b) The high resolution cross section STEM of the CoFe/BSTS. (c) The high resolution TEM of bulk BSTS single crystals.

anti-localization type to normal parabolic type. The weak anti-localization type magnetoresistance is due to the π Berry phase of the helical surface state and the strong spin orbit interaction in the bulk[9-11]. The thickness dependence of weak anti-localization behavior indicate that the helical surface state contribute significantly to the weak anti-localization in a $\sim 100 \mu\text{m}$ thick BSTS sample [7].

As shown in the cross section STM and STEM of BSTS/CoFe heterostructure (Fig. 4.2), the interface between BSTS single crystal and CoFe thin film is not sharp, which may be due to a thin oxide layer covered on the BSTS single crystal or new compounds formed between BSTS and CoFe. The unknown compound layer formed at the interface between BSTS and CoFe may result in the novel anisotropic magnetoresistance in the magnetic field.

4.2 In-plane anisotropic magnetoresistance measurements

To study the exotic helical surface of the topological insulator with significant surface transport, we performed in-plane anisotropic magnetoresistance measurements on BSTS/CoFe (5 nm) heterostructures. For every resistance vs. magnetic field measurement, the angle θ between the current and applied magnetic field was fixed. A series of the measurements were performed by changing θ from 0° to 360° with 15° interval. In the measurements, the samples were cooled down to 10 K with zero field. After that, the magnetic field was set to 10 kOe to saturate the ferromagnetic moment in CoFe layer, then swept to -10 kOe and finally drove back to 10 kOe. When the magnitude of the magnetic field was larger than 800 Oe, the data was collected every 1 kOe. Between -800 Oe and 800 Oe, the resistivity was measured every 10 Oe. Fig. 4.3(A)a–3(A)p show sixteen magnetoresistance measurements of a BSTS/CoFe sample at 10 K at sixteen different θ s, respectively. The BSTS crystal of this sample was cut from the same crystal of the sample S1 (Fig. 4.1). The current was applied to

the direction B on the (001) plane as shown in diagram 2q below the Fig. 4.3(A) and Fig. 4.3(B). The measurement setup is shown in diagram 2r. When the magnitude of the magnetic field is larger than 800 Oe, the system shows positive magnetoresistance with no special characteristics at all θ values. Very unusual magnetoresistance behavior is observed in a field range between -800 Oe and 800 Oe. The two most striking characteristics are the disappearance of magnetoresistance at 0° and 180° (subtracting the background from the single crystal BSTS) and the square shape magnetoresistance shown at several θ s (30° , 120° , 150° , 210° , 300° and 330°). The novel anisotropic magnetoresistance was confirmed in nine BSTS/CoFe samples. As the (001) plane has a C6 symmetry, the current was applied along the A, B or C direction with 20° interval as shown in the diagram 2q below Fig. 4.3(B). For each current direction, three samples were measured. S3a and S3b were exfoliated from the same crystal of sample S3, of which the $R(T)$ curve is shown in Fig. 1. The magnetoresistance measurements of the BSTS (sample S3a)/CoFe heterostructure are shown in Fig. 4.3(B), in which the current is applied to C direction. Its anisotropic magnetoresistance shows the same characteristic as that of the BSTS (sample S1)/CoFe (Fig. 4.3(A)). The current flow in BSTS (sample S3b)/CoFe is along direction A, which also shows similar anisotropic magnetoresistance behavior. From these measurements, we can obtain two important conclusions. Firstly, the novel square shape magnetoresistance can be confirmed in BSTS/CoFe structures fabricated from different BSTS crystals. Secondly, the current

Figure. 4.3: Anisotropic magnetoresistance curves of BSTS/CoFe heterostructures at sixteen different θ s when current is applied to B direction (sample 1, Fig. 4.3(a)) and C direction (sample S3b, Fig 4.3(b)). The red curves are the scans from positive field to negative field. The black curves are the scans from negative field to positive field. The diagram 4.3q below the figure shows the A, B and C directions. The diagram 4.3r shows the set up of our experiments.

flowing along different crystalline direction shows negligible effect on the anisotropic magnetoresistance.

We also studied the temperature dependence of the square shape magnetoresistance (sample S1) as shown in Fig. 4.4. All the measurements were carried out with $\theta = 30^\circ$, because the evolution of the square shape magnetoresistance is more easily to be studied. It is found that the square shape magnetoresistance disappears when the temperature is higher than 50 K. The high temperature magnetoresistance ($T > 50$ K) is composed of a positive magnetoresistance from BSTS single crystal and two very broad peaks (negative magnetoresistance) which may be originated from the CoFe layer. The square shape type anisotropic magnetoresistance and its temperature evolution are phenomena unforeseen in normal single layer ferromagnetic systems.

To study the exotic magnetoresistance behavior, we measured the anisotropic magnetoresistance of $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ single crystal and 5 nm thick CoFe film on Si (111) (with the same crystal structure and similar lattice constant as BSTS (001) surface), respectively (Fig 4.5(A) and 4.5(B)). The magnetoresistances were measured under different angles and no spin valve effect was obtained in all the measurements. The CoFe film on Si (111) shows standard anisotropic magnetoresistance of ferromagnetic material [12-14]. Its magnetoresistance value almost keeps constant for all θ s although the shape of the magnetoresistance curve evolves with the variation of θ . This behavior is very different from that of CoFe/BSTS heterostructure, of which the magnetoresistance shows pronounced

change with θ and is zero near 0° and 180° (subtracting the background from the weak anti-localization of the single crystal BSTS).

Figure 4.4: The magnetoresistance evolution of BSTS/CoFe (sample S1) with increasing temperature. The range of magnetic field is from -800 Oe to 800 Oe. The magnetoresistance curves were measured at (a) 4 K, (b) 6 K, (c) 10 K, (d) 20 K, (e) 50 K, and (f) 100 K, respectively. θ is equal to 30° for all measurements in this figure.

The magnetoresistance of CoFe films on Si(111) also shows little change with increasing temperature due to the high Curie temperature. As shown in Fig. 4.5(A), the magnetoresistance of non-magnetic BSTS single crystal shows no special feature at all θ s except the weak anti-localization behavior. Therefore, we can conclude that the unforeseen magnetoresistance of BSTS/CoFe is not the addition of the normal anisotropic magnetoresistance of BSTS single crystal and CoFe thin film. The peculiar characteristics of the novel anisotropic magnetoresistance of the BSTS/CoFe

Figure 4.5: (a) The anisotropic magnetoresistance of single crystalline BSTS with various θ_s at $T = 10$ K. (b) The anisotropic magnetoresistance of CoFe(5 nm)/Si(111) with various θ_s at $T = 10$ K. The red curves are the scans from positive field to negative field. The black curves are the scans from negative field to positive field.

heterostructure can be summarized.

Firstly, the magnetoresistance from ferromagnet disappears at certain angles (0° and 180°) and only the magnetoresistance from BSTS is shown. Secondly, square shape magnetoresistance is shown at certain angles. Thirdly, the novel magnetoresistance disappears when $T > 50\text{K}$ and the magnetoresistance of BSTS single crystal changes from weak anti-localization type to parabolic type in the same temperature range. All these characteristics are different from the magnetoresistance of CoFe thin film on Si (111), although the Si (111) has the same crystal structure and very similar lattice constant comparing with the (001) surface of BSTS single crystal. There is no disappearance of magnetoresistance at any θ for CoFe/Si (111). The magnetoresistance of CoFe/Si(111) also shows small change when the temperature increases from 10 K to 100 K due to the high Curie temperature of CoFe.

4.3 A model to explain the novel anisotropic magnetoresistance: spin-momentum locking induced Giant magnetoresistance

As shown in the cross section TEM of BSTS/CoFe heterostructure (Fig. 4.2), the interface between BSTS single crystal and CoFe thin film is clear although not very sharp. From the STEM picture, we can conclude that the element distribution between BSTS and CoFe is pretty sharp, which indicates that the chemical reaction between

BSTS and CoFe is negligible. As we grew the CoFe thin film at room temperature, this result can be expected. Therefore, the effect of an unknown compound formed at the interface can be ruled out.

In order to explain the novel anisotropic magnetoresistance, we propose that, due to spin momentum locking, the BSTS/CoFe heterostructure in fact form a spin valve or Giant magnetoresistance (GMR) device. The novel anisotropic magnetoresistance is a GMR effect. As shown in Fig. 4.6(a), - standard spin-valve or GMR device is composed of two ferromagnetic layers (FM) separated by a nonmagnetic metallic layer (NM). The ferromagnetic moment in one of the ferromagnetic layers is pinned by an adjacent antiferromagnetic layer (AFM), usually fixed during growth or cooling down process in a magnetic field (the pinned layer), while the ferromagnetic moment in the other layer can be flipped freely by magnetic field (the free layer). Based on Mott's two-current model, the resistance of the system is determined by the relative orientation of the ferromagnetic moments in the two layers. Low resistance is realized if the spin of the free layer is parallel to the spin of the pinned layer. When the spin of the free layer is switched to antiparallel configuration to the pinned layer, the spin-valve is at its high resistance state. For more general condition, the GMR effect is determined by $\cos \delta$, where δ is the angle between the spin orientations in pinned layer and free layer. The parallel condition of spin orientation means $\delta = 0^\circ$, while the antiparallel condition of spin orientation means $\delta = 180^\circ$. There will be no GMR effect if δ keeps as 90° when we scan magnetic field. This condition can be realized when

we sweep a magnetic field perpendicular to the fixed spin orientation in the pinned layer. Furthermore, if the magnetic field is along the easy axis of the free layer, almost all the spins in the layer will flip at the coercive field, therefore the resistance shows a sudden jump at the coercive field (square shape $R(H)$ curve). On the other hand, if the magnetic field is not along the easy axis, the spin flipping is a process of gradual evolution, therefore the resistance of the spin valve structure varies slowly with changing magnetic field. The applied current in a GMR device is usually in-plane.

Figure 4.6: The schematic diagrams of (a) the normal spin-valve device and (b) the spin-momentum helical local induced spin-valve effect in a topological insulator/ferromagnet device.

The key point here is that the BSTS/CoFe system forms a spin-valve or GMR system with one fixed spin-polarized layer and one free spin-polarized layer. As shown in Fig. 4.6(b), for the BSTS/CoFe heterostructure, there are one ferromagnetic layer and one

adjacent helical surface layer. The ferromagnetic moment in the free CoFe ferromagnetic layer can be flipped by the applied magnetic field (the free layer), while in the top helical surface layer, the transporting electrons show fixed spin orientation (the fixed layer) due to the *spin-momentum helical locking*. In general GMR effect, the required conditions for realizing GMR effect is: a “free” ferromagnetic layer and a layer with fixed spin orientation. In the presence of spin-momentum locking, the requirements for GMR are fulfilled in the BSTS/CoFe heterostructure system. One important point is that the topological surface state will not be destroyed by the magnetic moments in CoFe, because the ferromagnetic moments in CoFe are in-plane and therefore parallel to the BSTS surface. The thin oxide layer at the interface can also separate the CoFe layer and the BSTS surface states and therefore protect the spin-momentum locking. In this system, there are four electron transport channels, the CoFe ferromagnetic layer, the top helical surface contacting the CoFe, the bulk BSTS, and the bottom helical surface. Only the electrons scattered in the CoFe layer and the adjacent top helical surface contribute to the GMR effect. Based on the above model, we can easily explain the aforementioned characteristics of the novel anisotropic magnetoresistance. The three characteristics of the anisotropic magnetoresistance CoFe/BSTS are the disappearance of anisotropic magnetoresistance at 0° and 180° , the square shape magnetoresistance at 30° , 210° , 150° , and 330° , and the temperature evolution of the anisotropic magnetoresistance. As aforementioned, the GMR effect is determined by the angle between the spin orientation in the pinned layer and free layer.

As shown in the schematic diagram (Fig. 4.6(b)), the spin orientation in helical surface is fixed and perpendicular to the current flowing direction. When $\theta = 0^\circ$ or 180° , the applied magnetic field (the spin orientation in the CoFe layer) is perpendicular to the fixed spin orientation in the helical surface state (the pinned layer), therefore there is no GMR effect except the background weak anti-localization effect from the bulk BSTS single crystal. With increasing θ (from 0° to 90° and from 180° to 270°), the projection of the applied magnetic field on the fixed orientation of spins in the helical surface increases and therefore GMR effect gradually enhances. At $\theta = 90^\circ$ or 270° , the applied magnetic field and the fixed spin orientation are parallel or antiparallel to each other and therefore generate the largest GMR effect. If we further increase θ , the GMR will decrease again and get to zero at $\theta = 360^\circ$ and 180° . These phenomena are just what we observed in our measurements (Fig. 4.3). It should be noted that the GMR effect is only determined by the relative orientation of the current flowing and magnetic field. As the magnetic field is in-plane, the 180° rotation of the sample is equivalent to the switching the current flowing direction to opposite direction, which will switch the spin orientation to opposite direction due to spin-momentum locking. As for the square shape magnetoresistance at 30° , 210° , 150° , and 330° , we believe that it is due to the magnetic anisotropy of the CoFe layer. As shown in Fig.4.6(b), the BSTS/CoFe system has biaxial magnetic easy axes near $\theta = 30^\circ$ (210°) and $\theta = 150^\circ$ (330°). As discussed in the last paragraph, when the applied magnetic field is along the easy axis, all the spins in the free layer switches to the

opposite direction simultaneously at the coercive field and therefore a square shape R (H) curve is obtained. The origin of the easy axis is still not clear yet. It may originate from the coupling between the helical surface state of $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ and CoFe, which diminishes at high temperature. As the novel magnetoresistance behavior is due to the spin-momentum locking at the helical surface, it is also easy to explain the temperature dependence of the spin-valve type magnetoresistance as shown in Fig. 4.4. It should be noticed that the weak anti-localization behavior of BSTS (the inset of Fig. 4.1) and the GMR type magnetoresistance (Fig. 4.3) appear in the same temperature range and disappear with increasing temperature. The interconnected results indicate that the disappearance of GMR magnetoresistance with increasing temperature may be due to the loss of significant helical surface transport at high temperature. The high temperature magnetoresistance ($T > 50$ K) is composed of a magnetoresistance from BSTS single crystal and an anisotropic magnetoresistance from the CoFe layer. It also should be noted in Fig. 4.4 that the magnetoresistance shows two downward sharp peaks at 20 K. It might be due to the competing effect of thermal agitation energy and coupling between momentum-locked spin and magnetic moment of CoFe. It is well known that the GMR effect is only determined by the average spin polarization of electrons moving from high potential to low potential. A larger current density only means that the percentage of electrons moving to a certain direction becomes larger. The average spin polarization of the current will remain the same. As larger current density will not change the spin polarization of the current, the

anisotropic magnetoresistance should remain the same with changing current density, which is also confirmed by our measurement as shown in Fig. 4.7.

Figure 4.7: Anisotropic magnetoresistance curves of BSTS/CoFe heterostructure (sample S3b, Fig. 4.1) and at $\theta = 255^\circ$ with applied current of (a) 0.01 mA, (b) 0.025 mA (c) 0.05 mA (d) 0.1 mA (e) 0.15 mA and (f) 0.2 mA. The red curves are the scans from positive field to negative field. The black curves are the scans from negative field to positive field.

For further study of such spin valve effect, we did a comparison of Bi_2Se_3 and BSTS.

Firstly, we measured both Bi_2Se_3 and BSTS pure single crystal with the magnetic field parallel and perpendicular to current direction at 2 K separately as shown in Figure 4.8.

Figure 4.8: Magnetoresistance of pure single crystal BSTS (a, c) and Bi_2Se_3 single crystals (b, d) at 2 K. (a) and (b) show the result when the current direction is perpendicular to the magnetic field while (c) and (d) show the result when the current direction is parallel to the magnetic field.

The parabolic MR curves observed in BS single crystals are quite different with that of BSTS samples. Since both of these two single crystals are strong spin orbital interaction materials, it is impossible to explain this difference using spin orbital interactions induced weak antilocalizations (WAL). The WAL effects cannot produce the two different shape MR curves. One possible reason caused such different MR may be due to the 2D electron surface states. As mentioned in the beginning, the

Figure 4.9: The isotropic in-plan MR resistance measured for CoFe/BS at different angles at 2 K.

Bi_2Se_3 samples always show the heavily doped n-type semiconductor behavior because of the selenium vacancies. Such doping effect results in that the protected surface states conduction contribution will be negligible compared with the bulk contribution. However, in BSTS samples, due to the high bulk resistivity, the surface states contribution will be largely enhanced and therefore the obtained MR curves in (a) and (c) are WAL effects which are induced by 2D surface states. WAL effects are going to be detailed discussed in the following chapter. We also obtain in in-plane MR measurement that the magnetoresistance is isotropic.

We also try to obtain the spin valve effect in CoFe/BS structures. Unfortunately, due to the high bulk conduction contribution, most of the current flow through the bulk channel and we cannot observe such interesting effect. All the measurement results of CoFe/BS structures are shown in Fig. 4.9.

4.4 Summary

In summary, novel in-plane anisotropic magnetoresistance has been observed in topological insulator BSTS/CoFe heterostructure.

To explain the novel effect, we propose that the BSTS/CoFe heterostructure forms a spin-valve or GMR device with one fixed spin polarized layer and one free spin polarized layer. The ferromagnetic moment in the free CoFe ferromagnetic layer can be flipped by the applied magnetic field (the free layer), while in the adjacent helical

surface layer, the transporting electrons show fixed spin orientation (the fixed layer) due to the *spin-momentum helical locking*. As the magnetic moment of CoFe is parallel to the BSTS surface and the protection of the thin oxide layer, the CoFe cannot break the time reversal symmetry and therefore the spin-moment locking of the helical surface states is preserved. The novel in-plane anisotropic magnetoresistance is originated from the GMR structure formed by the CoFe layer and the adjacent helical surface of BSTS. Further theoretical and experimental research is required to fully understand the novel anisotropic magnetoresistance.

4.5 Bibliography

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Chapter 5

Other Topological Insulators and Superconducting transport

Before the BSTS single crystals, we tended to fabricate other topological insulator single crystals which is able to be electrically studied on the helical surface state, such as, Bi_2Se_3 , $\text{Ca}:\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Te}_{1.95}\text{Se}_{1.05}$ single crystals. All these samples are proved as topological insulators by ARPES and STM experiments and they are supposed to supply a platform to study the surface states through electrical transport method. However, we didn't continue such research on them due to different reasons which are going to be discussed below. In spite of this, we still did the preliminary work and get some interesting results.

5.1 Shubnikov-de Haas (SdH) oscillations in Bi_2Se_3

Bi_2Se_3 , a conventional thermal electric material, has been predicted to be a topological insulator [1]. Recent ARPES measurement shows an almost fully spin polarized surface state [2]. Various electrical transport measurements on Bi_2Se_3 have been carried out [3-9]. Although Bi_2Se_3 is expected to have a 300meV bandgap [10, 11], all of these transport experiments on the bulk Bi_2Se_3 materials still cannot directly indicate the topological properties due to the high bulk carrier density. Only a

few transport experiments on the nanoscale Bi_2Se_3 materials can indicate the two dimensional surface states [12-15] because of the highly enhanced surface to volume ratio and it is also our motivation of achieving BSTS nanoflake devices which has been detailed discussed in Chapter 4.

As one of the prototype topological insulators, the study of angular dependent Shubnikov de Hass oscillations on bulk Bi_2Se_3 single crystals has been achieved and the anisotropy of the Fermi surface in all directions has been mapped out [16]. In their study, they have shown that the observed angular dependent SdH oscillations originate from the 3D bulk Fermi surface. Although the two dimensional signatures haven't been observed in the bulk Bi_2Se_3 , the method of magnetoresistance study in low temperature and high magnetic field to obtain the 2D topological surface states in other topological insulators is important. In the mean time, observing SdH oscillations in Bi_2Se_3 single crystals is still very important and meaningful in our experiment as this phenomenon can be used to verify the qualities of material.

High quality Bi_2Se_3 single crystals are grown using a modified Bridgeman method which has been described in Chapter 3. The obtained crystals are easily cleaved and revealed a flat shiny surface. The X-ray diffraction pattern indicates a rhombohedral structure with no impurity phase. The resistivity was measured by a standard four probe method with current flowing in the (1000) surface of a 10 mm (x direction) x 3 mm (y direction) x 0.1 mm (z direction) sample. Figure 5.1 shows the resistivity

measured at 2 K and 10 K with field perpendicular to the (001) surface. The Shubnikov-de Hass oscillations, which usually can only be clearly seen in dR/dH vs. H curve, are observed directly in R vs. H curve. As the magnitude of SdH oscillations is determined by the shape of the Fermi surface, temperature, the life time of electrons (Dingle temperature), and the quality of single crystal, the large SdH oscillations indicates very high quality of the single crystal.

Figure 5.1: Magnetoresistance of the Bi_2Se_3 single crystal under a magnetic field perpendicular to (001) surface at 2 K and 10 K respectively.

Since Bi_2Se_3 has been well studied, the research on it becomes less attractive but the technique of fabrication high quality single crystals encourages us to develop other topological insulator single crystals with lower bulk carrier concentrations.

5.2 Metallic Ca:Bi₂Se₃ single crystal

It is well known (or noted) that both the first and second generation of 3D topological insulators, BiSb, Bi₂Te₃ and Bi₂Se₃, are all difficult to be studied on the helical surface states through electrical transport method due to the large bulk contribution issues. Therefore, doping a certain chemical into the above topological insulators during the growing procedure would be a way to tune the Fermi level and finally enhance the bulk resistivity while keeping the topological surface states. Here we doped Ca (~1%) into Bi₂Se₃ and fabricated the Ca: Bi₂Se₃ single crystals. The growth method of Ca: Bi₂Se₃ is similar to that of Bi₂Se₃ which is a bit different from reported [17]. Due to the equipment limitations, we cannot stir the evacuated quartz with containing melting sources inside during the growth period. Therefore, the Ca elements in the Bi₂Se₃ single crystals are not homogeneous which may affect the transport measurements. Due to the sedimentation effect, the Ca concentrations are different at different positions of the single crystal. However, such sedimentation effect has never been obtained in BSTS samples. The single crystals are also easily cleaved and revealed a flat shiny surface. Two rectangular pieces of sample with certain dimensions were cut from the different positions of the single crystal and standard four electrodes were prepared on them using silver paste. Figure 5.2 shows the temperature dependence of the resistivity curves of these two samples where sample 1 was cut from the top surface marked as TI 7.1 and sample 2 was cut from the middle region of the single crystal marked as TI 7.2.

Figure 5.2: Temperature dependence of longitudinal resistivity in the range of 2-300 K of Ca (1%) doped Bi_2Se_3 single crystal samples. Sample TI 7.1 and TI 7.2 are selected from upper surface and middle region of the same single crystal separately.

Although the Ca: Bi_2Se_3 sample shows a 10 times larger resistivity than Bi_2Se_3 in the whole temperature range, the R vs T curve still shows the metallic property as the resistivity decreases with decreasing of the temperature. Moreover, we found that TI 7.1 has a higher resistivity than TI 7.2 at all temperatures ranges. The difference of the resistivity between these two samples is due to the different stoichiometric of Ca in these two samples which is supported by EDS experiment. Based on the EDS results, TI 7.1 has a higher Ca concentration than TI 7.2 by an amount of 0.2% .

We also applied the low temperature magnetoresistance measurement (sample surface is set to perpendicular to magnetic field) to these two samples and the results are shown in Fig.5.3. Unlike the MR observed in pure Bi_2Se_3 sample, no obvious

Figure 5.3: (a) Magnetoresistances of TI 7.1 and TI 7.2 measured at 2 K with field perpendicular to sample surface. (b) For better visibility, $d\rho_{xx}/dB$ is plotted as the y axis and the suppressed SdH oscillations appear in TI 7.1 which was cut from up surface of single crystal.

Shubonikov-de Haas oscillations are difficult to be obtained in both of samples in the raw data as shown in Fig. 5.3(a). However, after the first derivation analysis, the oscillations are observed and $d\rho/dB$ vs magnetic field curve was plotted in Fig. 5.3(b). Interestingly, the weak and suppressed Shubonikov-de Haas oscillations appear in TI 7.1 which was selected from the top surface of the single crystal. However, no oscillations can be observed in sample 2. Based on the EDS results, the sample 1 contains less Ca elements than sample 2. Therefore, the MR results show that

Shubonikov-de Haas oscillations can only survive in the lightly doped Bi_2Se_3 samples and this result agrees with the conclusion reported in literature[17]. Comparing with the Shubonikov-de Haas oscillations obtained in doped and un-doped Bi_2Se_3 , the oscillations observed in TI 7.1 are strongly suppressed and it may be explained by the defects or disorders induced by Ca in the lattice structures.

Based on both $R(T)$ and magnetoresistance results, we conclude the study of Ca: Bi_2Se_3 in the following three points: Firstly, the resistance of doped Bi_2Se_3 is still on the level of milliohms although the Ca dopants enhance the resistivity to 10 times larger than Bi_2Se_3 . The resistance is so small that it is impossible to discriminate the surface transport contributions from the total signals. If we increase the driven current, other effects may be induced during the measurement and the nonexpectable results will be observed. Secondly, the quantum effect of SdH oscillations is strongly suppressed by the doping level of Ca. The effect becomes weaker and weaker as the concentration of Ca increase. Thus, low Ca concentration is required for the SdH oscillations study. However, higher resistivity of the sample requires the higher concentration of Ca according to Fig.5.2. Such a contradiction makes it quite difficult to study the topological properties of the sample. Thirdly, the equipment limitations make it impossible to fabricate the homogeneous single crystals resulting that the samples and experiments are not repeatable.

In conclusion, the Ca: Bi₂Se₃ single crystal fabricated in our experiment is not a good platform to the further study in topological insulators.

5.3 Superconductor-like behavior in Bi₂Se₃

Since the topological band theory and superconductivity have the direct analogy [19-21], some concepts of topological insulator are also applicable to superconductor. Seeking for the Majorana fermions is one of the most interesting fields in the topological insulator study. This new particle is predicted to exist at the interface between superconductor and topological insulator [22-23]. Therefore, the interactions between topological insulator/superconductor and the transition from topological insulator to superconductor attract a lot of attentions.

There are a few pioneering works on it and the superconductivity in topological insulator has been achieved by doping Cu or Pd into Bi₂Se₃ or Bi₂Te₃ [24-27]. In their report, the dopant of Cu are intercalated into the Se layers where bond by the van der Waals forces [24]. The resistivity and AC magnetizations have been well studied [26]. However, the mechanism behind the transition from topological insulator to superconductor hasn't been figured out yet.

Before our modifying the single crystal furnace, one batch of Bi₂Se₃ single crystals was fabricated using the old method. In such samples, all of them show the different low-temperature transport properties with the ones we report in chapter 1 and 5.1. The preliminary research has been applied on such samples. In this section, we are going

to report the interesting superconducting behavior, low-temperature resistance and anomalous magnetoresistance observed in Bi_2Se_3 . Moreover, angular dependent magnetoresistance measurements were also studied on it. A series of measurements have been taken, yet the mechanism behind such attractive phenomena is still unclear. To figure out the physics behind them, further work will be needed. The experimental procedures and results are discussed below.

Two regular size samples were selected and cut from bulk Bi_2Se_3 and silver epoxy was used as the electrodes in the study of transport property. Standard four probe method was used for all the measurements. The RT curve shows the same behavior as the normal BS single crystals in the range from 10 K and 300 K. While at temperature below 4 K an abrupt decrease of resistance was obtained. We prepared two BS samples from the same position on the same single crystals. A series of $R(T)$ curves were obtained with the magnetic fields varying from 0 Oe to 2100 Oe and the results are shown in Figure 5.4. For sample 1, an abrupt decrease appears at around 3.7 K while the transition appears at 4 K for sample 2 (Fig 5.4(b)). The low temperature resistance behaves like a superconductor but nonzero resistance is achieved. As the field applied, the resistance below transition temperature (T_T) continuously increases with the increase of the magnetic field. It should be noted that such transition behavior disappears for both samples as the magnetic field is large than 2.5 kOe. Below the T_T , sample 1's resistance decreases with temperature in all the magnetic field range as shown in Fig. 5.4(a). However, sample 2 shows a different

R vs T behavior below the TT. When the temperature is below the first TT (4 K) and the applied field is a very low (<100 Oe), a second transition appears. The resistance starts to increase from the decreasing background when the temperature is below the second TT. Interestingly, it varies with magnetic field and increases with the strength of field. The second TTs obtained from the experiment are 3.2 K, 2.8 K and 2.5 K which respond to the field of 0, 50 and 100 Oe separately. However, no more second TTs can be observed if the magnetic field is larger than 100 Oe.

The abrupt decrease of resistance at low temperature observed in BS samples is very similar to the superconductor of $\text{Cu}_x\text{Bi}_2\text{Se}_3$ [25]. However, we didn't observe the zero resistance below T_c as reported. Such phenomenon is also very different from the reported results in superconductor/TI structure. In their report, the abrupt upturn of resistance is observed below the T_c of the superconductor electrodes and moreover the resistance enhancement is strongly suppressed by the magnetic field [21].

Temperature dependent magnetoresistance results of sample 1 are shown in Fig. 5.6. During this measurement, the magnetic field was always set to perpendicular to the sample surface on the rotator sample holder. Figure 5.6(a) shows the magnetic field dependence of resistance with various temperature of sample 1. It is easy to observe that an obvious negative MR dip appears near the zero field only when the temperature is lower than 3.7 K. The amplitude of the MR dip increases with decrease

Figure 5.4: Temperature dependent resistance behavior with various field in bulk Bi_2Se_3 . (a) Resistance versus temperature of sample 1 under a magnetic field range from 0 to 2100 Oe. (b) Resistance versus temperature of sample 2 under a magnetic field range from 0 to 2100 Oe.

Figure 5.5: The schematic of scanning method for sample 1.

temperature and the maximum dip is only takes 1.3% of the MR background. Figure 5.6(b) is the zoom in of (a) near the zero field.

For further study, the angular dependent magnetoresistance measurements are performed on both samples. For sample 1, the magnetic field was scanned in the plane formed by axis **a** and **c** as shown in Fig. 5.5 while the scanning plane formed by **b** and **c** is used for sample 2. The measurements for sample 1 and 2 were performed simultaneously on the rotator puck and the current directions of two samples were perpendicular to each other. Temperature dependent magnetoresistance results of sample 1 are shown in Fig. 5.7.

Figure 5.6: Temperature dependence of resistance in bulk BS sample 1 contacted by silver epoxy. (a) Resistance versus Magnetic field of sample 1 under a series of temperature range from 2 to 4 K. (b) Zoom in of (a) near the zero field.

Figure 5.7: Angular dependence of resistance in bulk BS sample 1 at 2 K contacted by silver epoxy. (a) Resistance versus magnetic field of sample 1 under a series of angle range from 0° to 86°. (b) Zoom out of (a) near the zero field.

Figure 5.8: Temperature dependence of resistance in bulk BS sample 2 contacted by silver epoxy. (a) Resistance versus Magnetic field of sample 2 under a series of temperature range from 2 to 4 K. (b) Zoom out of (a) near the zero field.

Figure 5.9: Angular dependence of resistance in bulk BS sample 2 contacted by silver epoxy. (a) Resistance versus magnetic field of sample 2 under a series of angle range from 0° to 86°. (b) Zoom out of (a) near the zero field.

A series of magnetoresistance with angles from 0° to 86° were systematically studied where 0° means the sample surface is parallel to magnetic field while 90° means the sample surface perpendicular to field. It is obvious that the MR dips appeared at all angles in the range of (-200 Oe, 200 Oe) (Figure 5.7(b)) at 2 K which is below T_c . As the increase of the angle between BS surface and field, the magnetic field range of the dip in MR also increases and it becomes the largest when they are perpendicular to each other. Similarly, we performed the same measurement procedure for sample 2 and the results are shown in Figure 5.8 and Figure 5.9.

To understand the origin of such results, it is necessary to look back to the study of $\text{Cu}_x\text{Bi}_2\text{Se}_3$. As reported, Bi_2Se_3 sample changes from normal topological insulator to superconductor due to the intercalations of Cu. In their Scanning Tunneling Microscopy (STM) study, Cu clusters were formed by the surface diffusion on the cleaved surface while the symmetry defined shape of the intercalated Cu was obtained in the subsurface. They also concluded that Cu is not uniquely donor or acceptor in such system [26]. Thus, the origin of the superconductivity observed in $\text{Cu}_x\text{Bi}_2\text{Se}_3$ may be from the subsurface. Other donor or acceptor may play the role in the subsurface to produce the superconductivity. The intercalated Cu may just enhance the concentration of such donor or acceptor. Since no other impurities were introduced into our sample, the unclear donor and acceptor should be from the defects in the sample.

In our experiment, no superconductor or other impurities were introduced into the sample; the observed phenomena cannot be explained by the doping or proximity effect. One of the possible ways to understand such results is the defects in the crystal structure. Such defects should be formed like the unclear donor or acceptor in the subsurface of $\text{Cu}_x\text{Bi}_2\text{Se}_3$. The old single crystal growth method must produce some unexpected crystallizations resulting in the defects in a certain step.

From the experiment results, we can observe from the $R(T)$ curve that under zero field resistance cannot achieve zero for both samples. It can be understood by that the defects only appear in the isolated islands which are surrounded by Bi_2Se_3 in the microscopic scale in the sample. When the current is driven through the sample, the paths in the islands contribute to the superconductivity while the paths out of the islands contribute to the normal conductivity. The observed T_c s of sample 1 and 2 are 3.7 K and 4 K separately and the difference of the T_c s may be due to the inhomogeneous concentration of the defects in the two samples although they were from the same position. It further indicates that such defects formation during the growth is very weak and sensitive. The study of the defects will become very necessary and important for understanding the transition from topological insulator to superconductor.

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Chapter 6

Summary and Future work

6.1 Summary

A series of high quality topological insulator single crystals samples were fabricated in the experiment: Bi_2Se_3 , $\text{Bi}_2\text{Ca}_x\text{Se}_3$ and $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$. Within these samples, only a certain kind of single crystals attract our attentions for further study due to the high bulk resistivity and repeatability. A few pioneer studies have been performed on the above samples, such as ferromagnetic/topological insulator interactions, transition from topological insulator to superconductor, thickness dependent resistivity, weak anti-localization (WAL) effect and universal conductance fluctuations (UCF). In these experiments, we observed the interesting phenomena such as: the spin valve effect obtained in the $\text{CoFe}(5\text{ nm})/\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ single crystal which indicated the existence of spin coupling effect, thickness dependent resistivity obtained in the bulk $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ and nanodevices which shows that the observed resistivity follow the two channel model, and the WAL results obtained from the nanodevice samples whose fitting results show that such effects should origin from the two dimensional topological surface states. All such results have directly or indirectly demonstrated the existence of spin polarized topological surface states. Especially for the $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ nanodevice, based on the two conducting mode and angular

dependent WAL effects, we demonstrated that nearly 97% conduction comes from the surface states. Moreover, the defects induced transition from topological insulator to superconductor obtained in Bi_2Se_3 single crystal is also important and meaningful.

6.2 Future Work

As discussed in Chapter 5, the mechanism of transition from topological insulator to superconductor-like behavior is proposed by defects. To observe and study the defects in the subsurface of Bi_2Se_3 will be very necessary in the future. Moreover, the study on the mechanism of transition from topological insulator to superconductor induced by defects will be more urgent. The further transport study on it will be necessary which needs a lower temperature cryostat system (mKelvin). The conductance verse voltage bias experiment will also be important in the study.

Although we discovered the spin valve effect in $\text{CoFe}/\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ structure, it is still not the direct evidence of existence of spin polarized surface. To observe the spin polarized current in the topological insulators, we need to fabricate the sensitive and precise spintronic devices. Here we propose such a method that can be used to directly detect the spin polarized electrons on the surface of topological insulator, Tunneling magnetoresistance (TMR) junctions. TMR junctions are commonly used in spintronics research. TMR is made of three layers with topological insulator/tunneling barrier/ferromagnetic layer structure. The basic idea of this junction device is that when a current driven through the three layers the spin

polarized electrons will jump through (tunneling) the insulating barrier with a certain possibility and be scattered with the spin polarized electrons in the ferromagnetic layer which results in a high or low resistance. When the electron spins on topological insulator surface is aligned to the spin orientation in ferromagnetic layer, a lower resistance will be observed. While when the electron spins on topological insulator surface is opposite to the spin orientation in ferromagnetic layer, the higher resistance will be observed. The possibility of tunneling depends on the thickness, area and crystal quality of the tunneling barrier which is commonly used as Al_2O_3 or MgO . The typical thickness of the tunneling barrier is around a few nanometers. Based on this junction, the spin polarization of the topological insulator surface can be calculated directly. Thus, it will be the main part of the future work.

List of Publications

- (1) B. Xia, P. Ren, A. Sulaev, P. Liu, S. Q. Shen, and L. Wang, “Indications of surface-dominated transport in single crystalline nanoflake devices of topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ ” *Phys. Rev. B* **87**, 085442 (2013)
- (2) A. Sulaev, P. Ren, B. Xia, Q. H. Lin, T. Yu, C. Y. Qiu, Z. P. Li, W. G. Zhu, M. Y. Han, S. Q. Shen, L. Wang “beta- Ag_2Te : A topological insulator with strong anisotropy” *AIP Advances* **3**, 032123 (2013)
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