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Jinsong Zhang

Transport Studies  
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Topological Insulator  
Thin Films



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Jinsong Zhang

# Transport Studies of the Electrical, Magnetic and Thermoelectric Properties of Topological Insulator Thin Films

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# Supervisor's Foreword

This thesis reports systematic transport studies on the electrical, magnetic, and thermoelectric properties of topological insulator (TI)—thin films grown by molecular beam epitaxy. TI is one of the most important frontiers in condensed matter physics in recent years. Due to the existence of strong spin-orbital coupling (SOC) in TIs, the electronic band structure shows a non-trivial topology, which promotes the formation of massless Dirac fermions in the TI surfaces. These unique properties not only generate novel physical phenomena in the fundamental researches, but may also have unique applications in spintronics and quantum computation. The experimental results presented in this thesis offer useful information for understanding the peculiar properties in TIs and demonstrate the feasibility of application in future devices.

The main conclusions and contributions of this thesis are as follows:

- (1) One of the most challenging problems in TI is the elimination of bulk carriers, so that one can investigate the intrinsic properties of surface states. By using isostructural isovalent mixtures of two prototypical TIs,  $\text{Bi}_2\text{Te}_3$ , and  $\text{Sb}_2\text{Te}_3$  both of which are V-VI compounds and have opposite bulk carriers, we successfully fabricated ideal TIs with truly insulating bulk and tunable charge carriers (electron- or hole-like) in the surface states (see Chap. 3). Thermoelectric measurements reveal that when the Fermi level lies around the Dirac point, the magnetoelectric properties are dominated by the surface-state electrons, whereas the thermoelectric effect at room temperature is predominately controlled by bulk states (see Chap. 6). These results demonstrate new routes for investigating the novel quantum transport properties of the topological surface states and designing high-performance TI devices.
- (2) With increasing Se concentration in Cr-doped  $\text{Bi}_2(\text{Se}_x\text{Te}_{1-x})_3$  TI thin films, we observe a magnetic quantum phase transition from ferromagnetism to paramagnetism accompanied by a sign reversal of the anomalous Hall effect. Across the critical point, a topological quantum phase transition of the band structures is confirmed by angle-resolved photoemission measurements and density functional theory calculations. Finally, the effective model calculations

show that the bulk band topology is the fundamental driving force for the magnetic quantum phase transition. More specifically, the topologically non-trivial band structure prefers ferromagnetic ordering at low temperatures, while the topologically trivial band structure tends to form paramagnetic ordering (see Chap. 4). These findings significantly increase the understanding and controlling of the topological and magnetic properties, providing an ideal platform for realizing the exotic topological quantum phenomena induced by breaking the time reversal symmetry in magnetic TIs.

- (3) By fine tuning the chemical composition, film thickness and substrate morphology, the quantum anomalous Hall effect (QAHE) is experimentally observed in Cr-doped  $(\text{Bi, Sb})_2\text{Te}_3$  TI thin films (see Chap. 5). At zero magnetic field and ultralow temperature (30 mK), the anomalous Hall resistance reaches the quantized value of  $h/e^2$  when the Fermi level is tuned into the sub-band-gap. Here  $h$  is the Planck constant and  $e$  is the elementary charge. Meanwhile, the longitudinal resistance shows a considerable drop that is consistent with the dissipationless edge state transport. Under strong magnetic fields, the longitudinal resistance totally vanishes, whereas the Hall resistance remains quantized. Moreover, the exact quantization is achieved on a macroscopic scale sample with relatively low mobility. Such a robust QAHE implies that it may be used in future low-energy-consumption electronics and quantum computation.

Beijing  
February 2016

Yayu Wang

# Abstract

The non-trivial bulk band topology and massless surface Dirac fermions are the most peculiar features of topological insulators, which are generated by strong spin-orbit coupling and protected by the time reversal symmetry. In this thesis, we present transport studies of the electrical, magnetic and thermoelectric properties of topological insulator thin films grown by molecular beam epitaxy. The transport measurements are performed on the isostructural isovalent mixtures of  $\text{Bi}_2\text{Se}_3$  family compounds as well as the Cr-doped alloys in temperature range from 0.03 to 300 K and magnetic field up to 18 T. The experimental results reported here provide useful information for understanding and utilizing the unique properties of topological insulators.

The existence of significant bulk conduction is a challenging problem for the experimental observation of novel quantum phenomena in topological insulators. To eliminate the bulk-carrier contribution, we employ the band structure engineering method in  $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$  ternary compounds. Electrical transport and angle-resolved photoemission spectroscopy (ARPES) measurements show that the Fermi energy can be tuned systematically across the Dirac point and the electrical properties are consistent with that of a single spin-polarized Dirac cone. Most remarkably, through the band engineering we have achieved ideal topological insulators with truly insulating bulk and tunable surface states. Further thermoelectric effect measurements on the same system displays a sign disparity between the Hall and Seebeck coefficients at certain Sb concentrations and temperatures, where the Hall effect has a negative sign but the thermopower is positive. Theoretical calculations and analyses reveal that this anomalous effect is produced by the high-mobility surface Dirac fermions and large bulk Seebeck effect when the Fermi level is in the vicinity of valence band maximum. Around the charge neutral point, the surface Dirac fermions always dominate electrical transport up to room temperature but the thermoelectric effect is predominately controlled by bulk electrons at high temperatures.

Breaking the time reversal symmetry is generally detrimental to the gapless surface states, but it may lead to exotic topological quantum effects. In Cr-doped  $\text{Bi}_2(\text{Se}_x\text{Te}_{1-x})_3$  topological insulator films, we observed a magnetic quantum phase



transition accompanied by the sign reversal of the anomalous Hall effect. Across the same critical point, a topological quantum phase transition is revealed by both ARPES measurements and density functional theory calculations. We present strong evidence that the bulk band topology is the fundamental driving force for the magnetic quantum phase transition, where the ferromagnetic order is strongly promoted in the inverted band structures.

Based on the above experimental achievements, we have been capable of fabricating ferromagnetic insulators through band structure engineering method. In the resulting Cr-doped  $(\text{Bi,Sb})_2\text{Te}_3$  thin films, the predicted quantum anomalous Hall effect is experimentally realized: when the Fermi level is tuned into the sub-band gap, the anomalous Hall resistance shows a quantized value of  $h/e^2$  in the absence of external magnetic field, accompanied by a considerable drop in the longitudinal resistance. Intriguingly, the exact quantization is observed on a macroscopic sample with relatively low mobility and non-zero bulk conduction. Such a robust quantum state not only reflects the topological nature of the quantum transport but also provides an ideal platform for the realization of potential application in future devices.

**Keywords** Topological insulator · Transport measurements · Magnetism · Quantum anomalous Hall effect · Thermoelectric effect

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