Experimental techniques used in topological insulators and realization of quantum spin Hall effect

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Abstract. With the theoretical development and prediction of various topological insulators in two and three dimensions, experimental realization of those topological states and engineering of their characteristics have been hotly pursued, among which the quantum spin Hall effect exhibited in two-dimensional topological insulators have raised lots of concern. This paper aims to firstly give a brief introduction to experimental techniques used in growing and characterizing topological insulators and then we focus on a specific direction, the realization of quantum spin Hall effect, to elucidate one ramification of the experimental progress in the realm of topological insulator. The quantum spin Hall effect distinguishes itself due to its lack of non-magnetic scattering preserved by time reversal symmetry in its edge channels. Thus, materials showing such phenomenon are significantly promising for achieving dissipationless spintronics.

1. Introduction
Topological insulator (TI), distinct from conventional insulators, semiconductors and other condensed matter materials, has insulated bulk but metallic edges and surfaces depending on the dimension of it. Such exotic distribution of states is revealed as gapless edge/surface states in the energy band spectrum of TIs. The wave function of TI exhibits topologically nontrivial behavior characterized by appropriate topological invariants like the Chern number[1], $Z_2$ invariant[2], a set of $Z_2$ indices[3], etc. Here we focus on the experimental aspect of TIs where the intrinsic topology of TIs has led to intriguing electronic and spin transport behavior. Through extensive research, the realization of these states has been successful under more ambient conditions, which adumbrates the promising future for their application in constructing spintronics and quantum computation. In this paper, we first give some background introduction to experimental techniques and devices usually utilized in growing and characterizing TIs in section 2. Then in section 3, we cover the development of realization of quantum spin Hall effect (QSHE) and present the latest progress in this field. Finally, conclusions and outlook are given in section 4.

2. Experimental techniques used in cultivating and characterizing TIs

2.1. Technology for growing TI materials
Topological insulators like strained HgTe/CdTe, Bi$_2$Te$_3$, Bi$_2$Se$_3$ etc. are thin-film materials which have three growing types[4]. The first growth mode is Volmer-Weber mode where deposit atoms grow in island form. Atoms are more self-bounded compared with their attraction to the substrate under this circumstance so at the beginning, they nucleate on the substrate and gradually form into separate islands with empty channels among them. With crystal nucleus gaining sizes, new deposit material
atoms would adsorb on existing islands. Finally, separate islands would combine with each other and become continuous thin films. The second mode is Frank-Vanber Merwe (layer) mode where atoms grow layer by layer faithfully attracted to the substrate for surface adhesive force is stronger than adatom cohesive force. In the third intermediate mode, Stranski-Krastanov (SK) growth mode, layers come first and then the system switches to island growth. A schematic of the three growth modes is shown below in figure 1.

2.1.1. Basic introduction to molecular beam epitaxy (MBE)
Molecular beam epitaxy, having been developed for more than 40 years[5, 6], is a powerful method to grow thin film materials of oxides, semiconductors and metals[7], monitor and study the processes of crystal growth and manipulate parameters of materials at atomic or molecular level. MBE has advantages of precisely controlling smoothness and thickness and guaranteeing the ultimate purity of the growing materials, based on basic requirements of ultra-high vacuum (UHV) and specific temperature. As a result, ensuring the low pressure and modulating temperature become integral in constructing such a device. Usually, the air pressure in MBE system is maintained at approximately $10^{-10}$ mbar by UHV system. The UHV system combines a series of different pumps including mechanical pumps, turbo-molecular pumps, ion pumps, titanium sublimation pumps and cryo-pumps. Besides, an ion gauge is encompassed to measure the pressure and monitor the vacuum degree of the system. Among those pumps, mechanical pumps and turbo-molecular pumps are responsible for pumping gas out, but they function under different pressure ranges($10^{-5}$ mbar as the limit for mechanical pumps and $10^{-5}$ to $10^{-10}$ mbar for turbo-molecular pumps) so mechanical pumps work as backing pumps of turbo-molecular pumps[8]. However, outgassing from the cavity surfaces or the materials per se and permeation from the atmosphere due to enormous pressure difference are obstacles to achieving ultra-high vacuum. Thus, ion pumps and titanium sublimation pumps, which both use the adsorption of titanium film towards gas molecules, are mounted to get rid of gases beyond the reach of two previous pumps. Cryo-pumps are implemented to hold the temperature of the system under control. With complete UHV system, a schematic diagram of the modern version of an MBE machine is shown below.
Effusion cells (originated from Knudsen sources) are used for evaporating species involved in the deposition process. Together with shutters and baffles, they are designed to control the flux of the beam and the purity of the materials. The nitrogen panel around the whole cavity regulates the temperature, but that is far from sufficient because in the practice, temperature of each part like each effusion cell or the substrate (regulated by the stationary substrate heater) needs to be precisely controlled in order to keep the state of matter or adjust the growth rate, the diffusion uniformity, etc. In the whole process of growing certain material, the temperature is a dynamic parameter regarding to both space and time, changed according to different requirements of different modules and various phases of the experiment[10].

Reflection high energy electron diffraction (RHEED), in the middle of the MBE system in figure 2, is a fundamental tool to observe the growth and structural properties of materials and carry out in-situ characterization. The basic principle of RHEED is to employ the diffraction pattern of glancing incident monoenergetic electrons with energy ranging from 3 to 50 keV to explore surface topography. The ideal assumption on the elastic electronic scattering (satisfying two conditions: $k_{in} = k_{out}$ and $k_{out} - k_{in} = K_h$, where $K_h$ is the reciprocal lattice vector of the sample lattice) is that diffraction of electrons on reciprocal lattice lines gives a series of spots on a series of concentric circles on the screen. However, due to the roughness of the sample surface, the ideal reciprocal lattice lines are blurred and become reciprocal lattice rods[9], which would generate streaks on the diffraction pattern, as shown in figure 3. The intensities of these stripes are correlated with the roughness of the sample surface, so by recording the oscillation of RHEED intensities we could adumbrate the evolution of the growth and calculate relevant parameters[11]. For detailed information on RHEED oscillation, one can refer to the work of Joyce[12] in 1986. In MBE system, the rotating substrate holder is also of vital significance, which serves to keep the uniformity of beam flux arriving at the substrate and contributes
to a more germane temperature condition for atom diffusion.

2.1.2. Other methods for growing topological insulator materials

MBE is a sophisticated and systematic way to grow TI materials. Nevertheless, the cost of a complete MBE machine is fairly high. We also have alternative ways to obtain TI materials such as exfoliation[13, 14], sonochemical synthesis[15], metal-organic chemical vapour deposition (MOCVD)[16], chemical vapour deposition(CVD)[17-19], etc.

Among them, CVD is a very common method using chemical reaction of gaseous substances upon solid surfaces and their deposition tendency to obtain thin film materials. It is easier to get purer, more compact materials and is convenient to dope, but facilities of it are still quite complex, requirements for substrate temperature are strict and the output is not productive.

2.2. Technology for observing and characterizing TI materials

RHEED, as a system to proceed in-citu characterization of materials, inside MBE chamber, has been introduced in previous part, while tools like x-ray diffraction (XRD), scanning tunneling microscopy (STM), atomic force microscopy (AFM), transmission electron microscopy (TEM), angle-resolved photoemission spectroscopy (ARPES) and so on are mighty methods to study properties of TI specimens and probe electronic features of them. In this paragraph, introduction in a nutshell is given on topics listed above. Besides, lots of nontrivial methods in condensed matter experiment are applied in the realm of studying TI materials, which are too many contents to be mentioned for this section.

2.2.1. X-ray diffraction and transmission electron microscopy

The use of X-ray on investigating lattice structure arises from the principle of Laue equations[20] which reduce to Bragg’s law, a more compact form, as expressed below.

\[ 2d \sin \theta = n \lambda \]  

\( \theta \) and \( \lambda \) are the incident angle and the wavelength of X-rays, respectively. \( d \) is the space between different planes of sample lattice and \( n \) is an integer. After scattered by arrays of atoms, the scattering waves would interfere with incident waves and diffraction pattern that reveals the distribution function of arrays of atoms can be obtained and analyzed.

Working under vacuum condition, TEM makes use of high-energy electrons to obtain the image of sample or the diffraction pattern. Emitted from an electron gun, electrons would be properly adjusted by electrostatic or electromagnetic lenses and apertures before transmitting through a specimen. After the transmission, electrons carrying information of the sample would be focused by objective lens and be magnified by electron lenses system. Finally, they would be projected onto a phosphor screen or a camera. The resolution of a TEM is of the same order of magnitude with the wavelength of a relativistic electron. For high-resolution TEM (HRTEM, image mode of TEM), the point resolution could reach 0.05nm[21].

2.2.2. Scanning tunneling microscopy and atomic force microscopy

The STM was invented by Binning and Rohrer in 1982[22, 23] and in 1986, one half of the Nobel Prize was awarded to Binning and Rohrer for their design of STM. Basically, the measuring object of STM is the local density of states (LDOS) of electrons of the sample surface, while the scope of samples is restricted to conducting samples[24]. A metal tip of atomic size is used to approach the sample surface at a distance of 5 to 10 Å, where the wave function of electrons in the tip and in the sample could overlap and tunneling effect would occur if a bias voltage is exerted. The principle of the tunneling process in STM can be illustrated by simple tunneling effect model in quantum mechanics[25].

The tunneling current can be expressed as
\[ I \propto \sum_{E_n = E_F - eV}^{E_F} |\psi_n(0)|^2 \exp(-2\kappa Z) \]  

(2)

where \( E_F \) is the Fermi energy, \( Z \) is the distance between sample surface and tip and \( \kappa \) is a constant determined by the energy required to remove an electron from metal bulk to vacuum level. Defining LDOS for small energy unit \( \epsilon \) as

\[ \rho_S(z, E) = \frac{1}{\epsilon} \sum_{E_n = E - \epsilon}^{E} |\psi_n(z)|^2 \]  

(3)

we can express the tunneling current in terms of LDOS.

\[ I \propto V \rho_S(0, E_F) \exp(-2\kappa Z) \]  

(4)

So by holding \( I \) constant (This is one of the two working modes of STM. Tip regulated by feedback system moves in \( z \)-direction according to surface variation in order to keep \( I \) unchanged), we can measure the LDOS and get the image \( z(x,y) \). Alternatively, we can keep \( z \) constant and trace the change of tunneling current and therefore gather information about sample surface. A schematic diagram of how STM works is drawn below. Piezoelectric translators control the three-dimensional movements of the tip. The dashed-line arrow in figure 4 represents the situation with inverse bias voltage where electrons in the sample tunnel their way to the tip.

Figure 4. Brief illustration of the working process of STM.

A whole STM system is, of course, far more sophisticated than the picture above. Usually, it contains a complete set of controlling system connected to computer that coarsely or rigorously controls piezoelectric scanners and tip, offers bias voltage and provides feedback information, magnifies and analyzes the circuitual signal, a mechanic system that exerts force on devices, transfers the sample, provides cushion and so on and a UHV system and a temperature control system like MBE. Due to the sensitive function relation between tunneling current and tip-surface distance and an atomic level tip (usually obtained through electrochemical corrosion), STM could achieve a vertical resolution of \( 0.1 \text{Å} \) and a lateral resolution of \( 1 \text{Å} \).

Following the inspiration of STM, Binning et al. invented AFM which is used to probe non-metallic materials in 1986[26]. Tip in AFM is an mechanic one rather than a tunneling one in STM[25], which senses all kinds of forces from the sample. Interaction between the tip and the sample surface would deflect the tip, thus deflecting the pliable cantilever on which the tip is fixated. The deflection would be detected by the detecting system and translated into electronic signal for analysis and feedback. Lots of techniques are implemented in the detecting system to perceive the tiny deflection of the cantilever, including imposing an STM above the cantilever[26], optical interferometer[27], monitoring reflected optical beam deflection[28], piezoelectric detection[29], etc.
In different operation modes, different sorts of forces dominate. In contact mode where tip touches the surface and operates raster scan, the force is basically repulsive contact force. In non-contact mode, forces include Van der Waals forces, electrostatic or magnetic forces, Casimir forces, etc. In sum, through those techniques and interaction, AFM could measure the mechanical characteristics of sample, image the sample surface and manipulate sample at atomic level.

STM and AFM both belong to the family of scanning probe microscopy (SPM) which is broadly applied in the study of condensed matter physics, nano-chemistry, biochemistry, etc.

2.2.3. Angle-resolved photoemission spectroscopy
ARPES, a refinement of traditional photoelectron spectroscopy, is state-of-the-art technology to directly observe the electronic structure, band dispersion and the Fermi surface of the samples. The fundamental principle of ARPES originates from the photoelectric effect observed by Hertz in 1887[30] and explained by Einstein in 1905[31]. In principle, an ARPES emits a beam of low-energy(<100eV) X-ray, usually by means of synchrotron radiation[32]. An electron in valence band of the sample absorbs a photon, escapes from the constraint and then is captured by an electron analyzer that can be a assigned to specific polar angle $\theta$ and azimuthal angle $\phi$. Within the non-interacting electron picture[33], the kinetic energy of the photoelectron can be expressed as

$$E_k = h\nu - |E_b| - \phi$$

(5)

where $h\nu$ is the energy of incident light, $E_b$ is the binding energy of the photoelectron and $\phi$ is the sample work function and the photoelectron quasi-momentum parallel to the sample surface(noting that $\mathbf{P}_\perp$ does not conserve due to lack of translational symmetry) is

$$\mathbf{P}_\parallel = h\mathbf{k}_\parallel = \sqrt{2m_eE_k}\sin\theta$$

(6)

To get the real space momentum of the electron, we only need to add an arbitrary reciprocal lattice vector to equation (6), so $\mathbf{p} = h(\mathbf{k} + \mathbf{K}_b)$ where $\mathbf{k} = \mathbf{k}_\parallel + \mathbf{k}_\perp$. This is only a primitive description of the kinetics of photoelectron and for detailed methods of mapping electronic structure from the kinetic behavior and band dispersion, one can refer to Ref[33-36]. In order to epitomize the underlying process of ARPES experiment, it is of vital importance to introduce the three-step model[37] and the one-step model[38]. Within three-step model, the photoemission is artificially separated into three steps but involved calculation is simplified. The first step is optical excitation of the electron into a bulk final state, the second is travel of the electron to the surface and the final step is transmission through the surface into vacuum. The photoemission intensity is then given by the product of three probabilities in three steps. Besides sudden approximation that assumes electron emission and system potential change are both abrupt is applied in the calculation of one-particle spectral function and photoemission intensity[33]. On the contrary, within one-step model, photon absorption, electron emission and detection are considered to be an integral and continuous process. As a result, Hamiltonian of such a system has to take into account surface contribution to the interaction Hamiltonian, which is fairly complex, so three-step model is more pragmatic in understanding the result of ARPES.

As a summary of section 3, it is necessary to mention that in an experiment on the growth and characterization of topological insulators, a joint system isolated from external environment, which combines all the techniques and devices in need, is essential so as to ensure purity of samples, preclude disturbance from outside, and enhance efficiency and continuity of experiments. A typical exemplar is the combination of MBE-low temperature STM (LTSTM) -ARPES where systematic characterization and band property investigation could be operated on high-quality topological insulators.

3. Experimental realization of QSHE
HgTe/CdTe quantum wells is a two-dimensional (2D) topological insulator possessing helical edge
states, a hallmark of quantum spin Hall systems, preserved by time reversal symmetry (TRS). The helical edge state is a pair of counter-propagating electron currents where the orientation of the electron momentum is firmly locked to its spin direction. As a low dimensional system, QSHE in 2D TI system is provided with the advantage of lacking backscattering and having robustness against many-body interaction due to protection of TRS. And that is partly why experimental realization has been ardently pursued. The theoretical prediction was realized in 2007 by König et al.\[39]\ In their experiment, they constructed a four-terminal Hall bar to measure non-local transport characteristics and demonstrated the existence of quantum spin Hall states (QSHS) in the quantum wells. The longitudinal four-terminal resistance $R_{14,23}=V_{23}/I_{14}$ was measured as a function of gate voltage for $B=0T$ and $T=30mK$. When the size of the device was sufficiently small, a $2e^2/h$ conductance plateau emerged as manifestation of QSHS. Part of the experimental measurement result is shown below in figure 5.

![Figure 5. $R_{14,23}$ versus gate voltage measured for $B=0T$ at $T=30mK$ from Ref[39]. Device I is $(20.0\times13.3)\mu m^2$ with a normal structure ($d=5.5nm$), while devices II, III, IV are $(20.0\times13.3)\mu m^2$, $(1.0\times1.0)\mu m^2$, $(1.0\times0.5)\mu m^2$, respectively, with inverted QW structures ($d=7.3nm$). Saturation near $2e^2/h$ conductance is evident in devices III and IV.](image)

An introduction of a small magnetic field perpendicular to the material plane, which broke TRS, opened a gap between two helical edge states and destroyed edge conduction, was also discussed in the paper. Then in 2009, a more detailed investigation using Landauer-Büttiker formalism[40] to coincide with the experimental transport properties was performed and dissipationless spin states transport was discussed, compared with chiral edge states[41]. Very interestingly, unconventional transport properties of HgTe quantum wells under broken TRS was reported in 2015[42] where the edge conduction(contribution of conductivity was confirmed to come from edge states) of 7.5-nm quantum well surprisingly survived at a 9T (expected field is about 3.8T) magnetic field. The discovery implies that principles beyond traditional QSH theory, including consideration of SOC of this specific material, surface effects, many-body interactions and so on, need further exploring. High-quality preparation and precise tuning of HgTe quantum wells are quite challenging, so easier ways of realizing QSHS have been pursued. On the one hand, methods of preparing high-quality HgTe structures have been developed. Recently, a chemical etching method for fabricating Hg/CdTe microstructures[43] was proposed and demonstrated to be a more effective way to cultivate this structure with better mobility and mean free path length. On the other hand, in 2008, InAs/GaSb/AlSb quantum wells which possess similar band structure to HgTe/CdTe quantum wells but are easier manipulated were predicted to have QSH phase[44] and soon in 2011, it was performed experimentally[45]. The material distinguishes itself through independence of edge modes from mesoscopic size of conductive bulk and magnetic field[45], as well as persistence of both conductance plateau and its inverse proportional property to sample size in a wide range of temperature[46].
However, QSHE in room temperature is still a major obstacle for application of QSHE. Bismuth bilayer, a 2D topological insulator, has been studied on its topological edge states experimentally in order to pursue deeper understanding of its physical nature and practical application at room temperature[47-51]. Recently, a method that utilized bismuth honeycomb lattice on top of SiC(0001) substrate to open a large energy gap up to 0.8eV for dissipationless spin current transport in room temperature was proposed by F. Reis, G. Li, et al.[52] This method whose viability has also been demonstrated experimentally in the work guides our attention to the crucial role of substrate in engineering large energy gap QSH insulators. Another promising candidate for achieving high temperature QSH phase is a monolayer crystal WTe$_2$. Since two-dimensional transition metal dichalcogenides (TMD) MX$_2$ with M=(Mo, W) and X=(S, Se, Te) was reported to be large gap QSH insulators[53], extensive work has been focusing on 1T’ monolayer WTe$_2$[54-59]. Recently, stable QSHE in monolayer WTe$_2$ up to 100 Kelvin was demonstrated by S. Wu, V. Fatemi et al.[60] They verified the nature of QSHE of the material from three aspects conclusively. First, they ruled out the contribution of trivial diffusive edge modes and examined the quantized edge conductance $e^2/h$ for each helical edge transport channel. Second, in the short edge limit, they confirmed the existence of edge conductance plateau. Third, they applied magnetic fields (both in-plane and out-of-plane magnetic fields) to break TRS of the system and observed Zeeman gap and suppression of quantized conductance as expected. What’s more, the robust helical edge modes survives up to a 100K high temperature, which steps closer to the realization of QSHE in ambient temperature for constructing dissipationless spin states transport devices. Experimental results of the temperature dependence of edges conductance are given in figure 6.

Figure 6. From Ref[60], the figure shows the behavior of edges conductance with varying temperature. In (a), where the sample is a 100nm channel, the breakdown of QSHE after 100K is shown to arise from the contribution of bulk conductance. (b) on the right reveals the temperature dependence of the full-length sample.

After that, further, the local conductivity of monolayer WTe$_2$ was imaged using microwave impedance microscopy and extra conducting lines and rings were observed in the experiment, indicating more researches on the properties of devices incorporating WTe$_2$ or other air-sensitive 2D materials[61]. In addition, another member among TMD, 1T’ monolayer WSe$_2$, growing on bilayer graphene substrate, was reported to be a large-gap (up to 129meV) 2D TI with tunable band gap[62]. So far, it has been unquestionable that in order to fabricate materials exhibiting QSHE at room temperature, limited sizes and large energy gap of materials have been fundamental factors. So materials located in bismuth and particularly in TMD are highly promising for dissipationless spintronics.
4. Conclusions and outlook

In summary, experimental study of QSHE in 2D TIs has enormously boosted the advancement in the study of topological materials. Those quantum spin Hall insulators, taking advantage of more sophisticated modernized experimental techniques in condensed matter physics, have been demonstrated to be promising and applicable in manipulating electronic and spin behavior for constructing new generation of spintronics. Besides 2D TIs and QSHE, lots of materials beyond topological insulators and other effects have been intensively researched since the emergence of topological insulator. For example, quantum anomalous Hall effect (QAHE) in both 2D and 3D cases where ‘helical’ is reduced to ‘chiral’ due to ferromagnetic suppression, topological crystalline insulators with additional point group symmetry, topological superconductors supporting unique gapless Majorana modes at their boundaries and the heterostructure of different kinds of topological materials based on the principle of proximity effect, the research of which has greatly reinforced our understanding of topology into condensed matter systems and provided us with an unprecedentedly broad platform to find their application in spintronics and quantum computation. For practical use of topological insulators and their relating materials, we still face challenges like room temperature realization, interaction with external fields, problems in tuning materials, impurities and so on. However, there is no doubt that these realms are highly promising and they are potential solutions to energy dissipation and quantum computation.

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