Topological Quantum Materials and the Quantum Anomalous Hall Effect Postprint

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Abstract

A brief overview of a novel class of material systems—"topological"quantum materials—that have emerged since around 2005, encompassing topological insulators, topological crystalline insulators, topological superconductors, and topological semimetals. The strong spin-orbit coupling in these materials leads to a rich variety of quantum phenomena, including the quantum anomalous Hall effect, which may provide significant impetus for the development of future technologies such as low-power electronics, topological quantum computing, and clean energy.

Full Text

Preamble

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Topological Quantum Materials and Quantum Anomalous Hall Effect

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ABSTRACT

We briefly review topological quantum materials, a new class of material systems developed since around 2005, including topological insulators, topological crystalline insulators, topological superconductors, and topological semimetals. The strong spin-orbit coupling in these materials leads to rich quantum phenomena such as the quantum anomalous Hall effect, which could significantly advance future technologies including low-energy-consumption electronics, topological quantum computation, and clean energy.

KEY WORDS review, topological insulator, quantum Hall effect, quantum anomalous Hall effect

1. Introduction

The development of materials science and condensed matter physics are mutually reinforcing. On one hand, the discovery of new materials and deeper understanding of existing ones compel physicists to revise and refine condensed matter theory. On the other hand, conceptual breakthroughs in condensed matter physics help materials scientists search for new material systems or discover novel properties in known materials. Over the past three decades, one major conceptual breakthrough in condensed matter physics has been the introduction of topological concepts from mathematics. Following the discovery of the quantum Hall effect in the early 1980s, researchers gradually realized that if a material' s electronic structure possesses unique topological properties, it could exhibit various quantum effects at macroscopic scales. This insight opened the door to a new world of materials—topological quantum materials. Around 2005, the discovery of topological insulators dramatically expanded the scope of topological quantum materials research. Building on topological insulators, theorists predicted numerous topological quantum states distinct from quantum Hall systems. More importantly, researchers found that many materials, including some long-familiar ones, belong to the topological insulator family and may display novel quantum effects at macroscopic scales. The realization of the quantum anomalous Hall effect in magnetically doped topological insulator thin films not only demonstrated how to obtain quantum effects through material engineering but also brought hope for practical applications of topological quantum materials. This review begins with the quantum Hall effect to recount the conceptual and experimental development of topological quantum materials, including topological insulators, topological crystalline insulators, topological superconductors, and topological semimetals, with particular emphasis on the research status and results concerning topological insulator materials and quantum effects, especially the theory and experiments of quantum anomalous Hall effect in magnetic topological insulators.

2. Quantum Hall Effect and Topological Properties of Electronic Structure

In the microscopic world, particle motion is governed by quantum mechanics, exhibiting phenomena fundamentally different from macroscopic physics. For instance, macroscopic motion always involves some energy dissipation, whereas microscopic motion does not. In an atom, electrons orbit the nucleus without energy loss—this dissipationless motion ensures the stability of atomic structure and the material world. The reason electrons can move without dissipation is that they occupy quantum states with definite energy that cannot be changed by small perturbations, a quintessential quantum mechanical effect. In fact, reflections on this problem at the beginning of the 20th century led to the birth of quantum mechanics [1].

How does electron motion change when large numbers of atoms aggregate into macroscopic materials? In insulators, electrons remain localized near individual atoms or chemical bonds between neighboring atoms, essentially no different from the single-atom case. These localized electrons retain their dissipationless quantum mechanical character but cannot conduct macroscopic current. In metals, electrons can travel much longer distances and conduct macroscopic current. However, during long-distance motion, electrons are inevitably scattered by impurities or lattice vibrations into different quantum states, leading to energy dissipation. The existence of electrical resistance and Joule heating in metals originates from such scattering.

Is it possible to find materials that conduct electricity at macroscopic scales like metals while maintaining the dissipationless quantum mechanical nature of electrons like insulators? More broadly, can we find materials that exhibit electronic quantum effects at macroscopic scales? Such quantum materials would possess properties completely different from conventional materials. If they could be applied at scale, they would revolutionize materials science and dramatically advance technology.

In fact, the first quantum material was discovered in the early 20th century: superconductors. When cooled below a critical temperature, a superconductor' s resistance drops to zero, enabling dissipationless current transmission. In the superconducting state, conduction electrons form Cooper pairs with bosonic properties. Large numbers of Cooper pairs can condense into a single quantum state that cannot be scattered into other states, thus eliminating energy dissipation [2]. Realizing room-temperature superconductivity has been a long-standing dream. After nearly a century of exploration, superconducting materials with critical temperatures exceeding 100 K have been obtained, but this remains far from room temperature. A more serious issue is that due to the complexity of the superconducting mechanism, there is no unified, clear understanding of whether superconductivity can really reach room temperature or how to raise the superconducting temperature to room temperature. Historically, most important superconducting materials, such as cuprate high-temperature superconductors and iron-based superconductors, were discovered largely by accident.

The discovery of the quantum Hall effect opened a completely different path for quantum materials. The Hall effect is one of nature' s most fundamental electromagnetic phenomena. When a current-carrying conductor is placed in a magnetic field perpendicular to the current direction, a voltage (Hall voltage) appears in the direction perpendicular to both the magnetic field and current. In ordinary non-magnetic conductors, Hall resistance (Hall voltage/current) is generally proportional to the magnetic field strength, with its sign and magnitude determined by the carrier polarity and concentration—this is the normal Hall effect [4].

In 1980, Klaus von Klitzing studied the transport properties of a two-dimensional electron gas (2DEG) at a semiconductor heterointerface under low temperature and strong magnetic fields. He discovered that the Hall resistance deviated from linear dependence on magnetic field, exhibiting a staircase shape (see Fig. 1a [Figure 1: see original paper]). Each plateau corresponded to a resistance value precisely equal to h/ne^2, where h is Planck' s constant, e is the electron charge, and n is an integer. Corresponding to each plateau, the longitudinal resistance dropped to zero, indicating dissipationless electron motion. These phenomena clearly demonstrated a quantum mechanical effect [3]. Notably, this effect could be observed in samples several millimeters in size, proving it is a macroscopic quantum phenomenon—later named the integer quantum Hall effect.

Two years later, Tsui, Störmer, and others discovered the fractional quantum Hall effect with n taking fractional values in higher-mobility 2DEG samples at III-V compound semiconductor interfaces [5]. The discoveries of integer and fractional quantum Hall effects earned Nobel Prizes in Physics in 1985 and 1998, respectively. Their importance lies in revealing an entirely new class of matter: topological quantum states.

Topology is a mathematical concept. For example, a donut has a hole that prevents its surface from being continuously and smoothly deformed into the surface of an orange (which has no holes). The number of holes distinguishes two-dimensional surfaces in three-dimensional space and is insensitive to details and continuous changes. A material' s properties are primarily determined by its electronic band structure. If similar topological features can be found in a material' s band structure, and the material' s physical quantities or quantum states depend on these topological features, one can obtain physical properties or quantum states that are insensitive to material defects, impurities, and other details.

In a metallic 2DEG under a strong perpendicular magnetic field, electrons undergo localized cyclotron motion. Correspondingly, the quasi-continuous energy bands transform into discrete Landau levels. When the Fermi level lies between Landau levels, the system becomes an insulator (Fig. 1a). Theorists discovered that the band structure of this magnetic-field-induced insulator possesses topological features different from ordinary insulators like vacuum, diamond, or Al_2O_3. It can be called a topologically non-trivial insulator, or simply a topological insulator [7]. Since its topological feature is defined by the first Chern invariant (named after mathematician Shiing-Shen Chern), it is also called a Chern insulator. Historically, this term emerged after the discovery of time-reversal-invariant topological insulators discussed later, so "topological insulator" usually refers specifically to time-reversal-invariant topological insulators, as it does in this review unless otherwise specified.

The topological feature of quantum Hall systems is determined by the number of filled Landau levels n, and the sample' s Hall resistance depends on this topological feature and a quantized constant: h/ne^2. Consequently, its value is insensitive to sample details. The contribution to the quantum Hall effect comes from n conductive channels at the sample edge, called edge states. The sample edge is also the boundary between the topologically non-trivial Landau-level insulator and topologically trivial vacuum (insulator). To realize the change in topological properties, Landau levels must cross the Fermi level near the boundary, inevitably creating metallic edge states. Quantum Hall edge states are chiral—under a fixed magnetic field, electrons can only move along the sample edge in one direction (clockwise or counterclockwise depending on the magnetic field direction) (Fig. 2a [Figure 2: see original paper]). This prevents backscattering to opposite-moving quantum states (backscattering is forbidden), which is the origin of zero longitudinal resistance in the quantum Hall effect.

The dissipationless nature of quantum Hall edge states at macroscopic scales is very similar to superconductivity and could be used for electron transport. However, quantum Hall effects require several Tesla of strong magnetic field, making most applications impractical. Could there exist a quantum Hall effect without external magnetic fields? Since the quantum Hall effect arises from the non-trivial topological properties of a material' s electronic structure under magnetic fields, if a material' s electronic structure itself possesses similar non-trivial topological properties, one could obtain a quantum Hall effect without external magnetic fields. In 1988, Haldane proposed the first model of a quantum Hall system without external magnetic fields [8]. Based on a two-dimensional hexagonal honeycomb lattice of single-layer graphite—later known as graphene—this model introduced a hypothetical periodic magnetic field (with zero net macroscopic field) that opened a gap at the Dirac points of the band structure, transforming it into an insulator. This insulator has topological properties similar to an n=1 quantum Hall system and can exhibit the quantum Hall effect without external magnetic fields.

The Haldane model was far from reality at the time: single-layer graphene could not be experimentally realized, and the work did not propose how to introduce periodic magnetic fields in graphene. Nevertheless, it first made people realize that "natural" topological quantum materials independent of external magnetic fields might exist, laying the foundation for later theoretical developments in topological insulators and quantum anomalous Hall effects.

3. Topological Insulator Materials

3.2 Two-Dimensional Topological Insulator Materials

The first proposed two-dimensional topological insulator was graphene with spin-orbit coupling [10]. However, real graphene has extremely weak spin-orbit coupling, opening a gap at the Dirac point far smaller than 1 meV—too small to observe the quantum spin Hall effect experimentally. Therefore, graphene cannot be considered a true two-dimensional topological insulator material. The first realistic two-dimensional topological insulator material was predicted by Zhang et al.: (Hg,Cd)Te/HgTe/(Hg,Cd)Te quantum wells [16]. HgTe’ s bulk band structure features unique band inversion, making the quantum well a two-dimensional topological insulator when its thickness meets specific conditions, with a band gap up to 90 meV. Due to applications in infrared detectors, (Hg,Cd)Te/HgTe/(Hg,Cd)Te quantum wells had been studied long before their topological properties were recognized, and very high-quality samples could be obtained. Figure 3a [Figure 3: see original paper] shows the schematic structure of such a quantum well sample prepared by the Molenkamp group following Zhang’ s theory [17]. The actual structure is quite complex to optimize carrier mobility. They fabricated the quantum well into six-terminal Hall devices of different sizes using microfabrication techniques and finally observed the quantum spin Hall effect at 30 mK in a Hall device about 1 m in size with HgTe layer thickness exceeding 6.3 nm, measuring a longitudinal resistance plateau near $h/2e^2$ [18] (Fig. 3b). Later, they observed quantum spin Hall edge states through nonlocal transport experiments [19]. However, their experiments have not yet been independently reproduced by other groups.

Preparing high-quality (Hg,Cd)Te/HgTe/(Hg,Cd)Te quantum wells requires specialized molecular beam epitaxy (MBE) equipment and microfabrication techniques, available in only a few laboratories worldwide, slowing further progress. Moreover, these quantum wells have poor thermal stability and contain toxic elements, hindering large-scale production and application. Since graphene’ s main problem is weak spin-orbit coupling, a natural choice for finding two-dimensional topological insulators is materials with graphene-like structures but stronger spin-orbit coupling. Following this approach, theorists have predicted that single-element thin films with near-two-dimensional honeycomb structures—Bi [22,23], Si, Ge [24], Sn [25]—are two-dimensional topological insulators. These simple, single-element materials have large band gaps, especially Sn thin films with gaps up to hundreds of meV [25]. Experimentally, two-dimensional honeycomb Bi films have been epitaxially grown on the surface of three-dimensional topological insulator Bi_2Te_3, with angle-resolved photoemission spectroscopy (ARPES) observing band structures possibly indicating a two-dimensional topological insulator phase and scanning tunneling spectroscopy showing signatures of edge states [26,27]. However, the hallmark of two-dimensional topological insulators—the quantum spin Hall effect—has not been observed, mainly because the Bi_2Te_3 substrate itself is often conductive, making transport measurements unable to resolve the transport properties of the Bi film itself.

Hexagonal honeycomb Si films, also called silicene, have recently been prepared on Ag surfaces [28,29]. However, the conductive substrate problem similarly hinders transport studies. $ZrTe_5$ films have been predicted to be two-dimensional topological insulators with band gaps up to 400 meV, but no experimental reports have appeared [30].

In 2008, Zhang' s group predicted a two-dimensional topological insulator based on conventional III-V semiconductors: AlSb/InAs/GaSb/AlSb quantum wells [20]. AlSb is a wide-gap semiconductor barrier layer, while InAs and GaSb are narrow-gap semiconductors. In the InAs/GaSb heterojunction, GaSb' s valence band maximum lies above InAs' s conduction band minimum. In this structure, GaSb and InAs layers are confined by each other and AlSb layers, causing the highest-energy hole subband in GaSb to lie above the lowest-energy electron subband in InAs. This band inversion leads to topologically non-trivial properties. The Du group observed quantum spin Hall effect behavior in this system, confirming its two-dimensional topological insulator nature [21]. Recently, by introducing impurities and strain, they raised the observation temperature of the quantum spin Hall effect to around 30 K, obtaining much clearer quantum plateaus than the German group' s results in II-VI semiconductors. Currently, AlSb/InAs/GaSb/AlSb quantum wells appear to be promising two-dimensional topological insulator materials with great development potential.

3.3 Three-Dimensional Topological Insulator Materials

In 2007, Fu and Kane proposed a simple method to identify three-dimensional topological insulator materials, greatly simplifying the theoretical search process [13]. Using this method, they predicted that Bi_1 Sb alloys become three-dimensional topological insulators when x is between 0.07 and 0.22. Three-dimensional topological insulators can be identified by ARPES through counting the number of times surface states cross the Fermi level between two time-reversal-invariant points in the Brillou zone: odd crossings indicate a topological insulator, even crossings a normal insulator. The Hasan group studied the surface band structure of Bi_1 Sb alloy samples prepared by high-temperature sintering and found that samples with x between 0.07 and 0.22 showed five crossings between time-reversal-invariant points, providing the first experimental confirmation of three-dimensional topological insulators [31].

However, Bi_1 Sb alloys have a very small bulk gap (only 30 meV), chemical disorder, and complex surface state structure, making further in-depth studies extremely difficult. Soon, Zhang, Fang/Zhang, Hasan, and others identified a better class of three-dimensional topological insulator materials: the Bi_2Se_3 family, including Bi_2Se_3, Bi_2Te_3, and Sb_2Te_3 [32,33]. These materials have rhombohedral crystal structures, with five atomic layers forming a quintuple layer (QL) along the Z direction (see schematic in Fig. 4a [Figure 4: see original paper]). Within each QL, the five atomic layers have strong covalent interactions, while inter-QL forces are weak van der Waals interactions. Theoretical calculations show that among compounds composed of group V and VI elements, only Bi_2Te_3, Sb_2Te_3, and Bi_2Se_3 are topological insulators. Their bulk gaps can reach 0.3 eV (Bi_2Se_3), much larger than Bi_1 Sb , and their surface states contain only a single Dirac cone near the Γ point (theoretical band structure calculation for Bi_2Se_3 shown in Fig. 4b, ARPES measurement in Fig. 4c), much simpler than Bi_1 Sb . This greatly facilitates studies of three-dimensional topological insulator surface state properties. This family of materials has attracted intense interest and is currently the most studied topological insulator system [33,34].

Besides Bi_2Se_3 family materials, other material systems have been found to exhibit three-dimensional topological insulator characteristics under certain compositions or conditions (such as strain), including ternary chalcogenides (e.g., $TlBiVI_2$, $TlSbVI_2$ where VI represents chalcogen elements), pyrochlore-structure oxides (e.g., $A_2B_2O_7$ where A is rare earth elements like Sm, Eu and B is 5d elements like Ir), Heusler and half-Heusler intermetallic compounds, Ce-filled skutterudite-type compounds (e.g., $CeOs_4As_{12}$ and $CeOs_4Sb_{12}$), and chalcopyrite semiconductors with I-III-VI_2 or II-IV-V_2 compositions (e.g., Ag_2Te) [35]. Some have been confirmed as three-dimensional topological insulators by photoemission experiments. These material systems possess diverse properties: some have strong anisotropy, some strong electron correlation, and some can be easily transformed into magnetic or superconducting materials through doping. These rich properties provide great convenience for studying topological insulators.

From a materials science perspective, there is no essential difference between three-dimensional and two-dimensional topological insulators. Making three-dimensional topological insulator materials into quantum thin films several nanometers thick may yield two-dimensional topological insulator phases [36], while stacking two-dimensional topological insulators layer by layer into three-dimensional systems can also produce three-dimensional topological insulator phases under certain conditions (e.g., strain), undoubtedly expanding the range of topological material choices [37,38].

The main experimental challenge for three-dimensional topological insulators is the contribution from bulk carriers. Most interesting properties and quantum effects of three-dimensional topological insulators originate from the Dirac surface states within the bulk gap. Since topological insulators are narrow-gap semiconductors, vacancies and antisite defects easily form during preparation, making the bulk material metallic. Solving this problem requires: (1) improving the quality of three-dimensional topological insulator materials, (2) achieving effective control over the material' s electronic structure and chemical potential, and (3) maximizing the surface state contribution.

Many groups have attempted to use chemical doping to tune carrier concentration in topological insulators. For example, doping Bi_2Se_3 with Ca or Mg can achieve charge-neutral or even p-type samples [39-41]. Since Bi_2Te_3 is typically n-type while Sb_2Te_3 is p-type, mixing them to form ternary topological insulator compounds $(Bi,Sb)_2Te_3$ enables free tuning from n-type to p-type [42,43]. Substituting Te with Se in Bi_2Te_3 to form ordered ternary Bi_2Te_2Se can greatly reduce bulk carrier concentration. In transport studies of these materials, contributions from Dirac surface states can be easily observed, including Shubnikov-de Haas (SdH) oscillations from surface states [44-46].

All three problems can be solved through molecular beam epitaxy (MBE) growth of high-quality thin films. The electronic structure and chemical potential of MBE-grown films are easily controlled through growth conditions, layer thickness, surface/interface chemical environment, and gate voltage. Thin films have much larger surface-to-volume ratios than bulk materials, making surface state contributions more prominent in electrical transport and other measurements.

The Xue group at Tsinghua University first established the growth kinetics for Bi_2Se_3 family three-dimensional topological insulator thin films, achieving layer-by-layer growth and obtaining macroscopically uniform films. By controlling MBE growth kinetics, they dramatically reduced defect density and defect-induced electron/hole doping [47-50]. Figure 6 [Figure 6: see original paper] shows ARPES evolution of MBE-grown Bi_2Se_3 films from 1 QL to 6 QL thickness [49]. Clear quantum well states demonstrate macroscopic thickness uniformity and high quality. For thicknesses below 6 QL, the gap in Dirac surface states due to hybridization between top and bottom surface states is clearly visible. Moreover, through chemical composition control and gate voltage, they achieved chemical potential tuning in Bi_2Se_3 family epitaxial films. The realization of high-quality, controllable three-dimensional topological insulator thin films laid a solid foundation for studying various quantum effects.

3.4 Quantum Effects in Topological Insulators

The importance of topological insulator materials lies in the novel quantum phenomena associated with Dirac edge or surface states. The quantum spin Hall effect in two-dimensional topological insulators has been experimentally observed, as described earlier. For three-dimensional topological insulators, one of the first observed properties is the absence of backscattering—the inability of surface state electrons to be scattered backward by impurities. Researchers obtained quasiparticle interference patterns of three-dimensional topological insulator surface states using scanning tunneling spectroscopy and confirmed the absence of backscattering in Bi_1 Sb alloys and Bi_2Te_3 by analyzing these patterns [51,52].

Another characteristic of three-dimensional topological insulator surface states is their Landau quantization behavior, which differs from ordinary two-dimensional electron gases. First, surface states near the Dirac point form zero-order Landau levels that do not change with magnetic field. Second, different Landau level indices are not equally spaced. Using scanning tunneling spectroscopy under strong magnetic fields, researchers have observed unique Landau quantization of Dirac surface states in Bi_2Se_3, Bi_2Te_3, and Sb_2Te_3

[53-56].

Observation of Landau levels suggests the possibility of observing quantum Hall effects in surface states, which underlies many quantum effects in three-dimensional topological insulators. Experimental observation requires samples with high surface electron mobility and low bulk carrier concentration. When HgTe films grown on CdTe substrates are thinner than several tens of nanometers, strain makes the HgTe film a three-dimensional topological insulator [37]. Since very high-quality HgTe can be obtained by MBE, the Molenkamp group observed the quantum Hall effect from Dirac surface states [38]. Recently, several groups have also observed quantum Hall effects in Bi_2Se_3 family three-dimensional topological insulators.

Exciton condensation is another interesting quantum effect in three-dimensional topological insulator thin films. Excitons are quasiparticles composed of electrons and holes bound by Coulomb interaction in semiconductors. As bosons, excitons can undergo Bose-Einstein condensation at low temperatures. Similar to superfluidity and superconductivity, exciton condensation is also a macroscopic quantum effect. Previous predictions of exciton condensation focused on double-layer quantum Hall systems separated by insulating layers, where sample preparation was a major challenge. Since high-quality three-dimensional topological insulator thin films are naturally double-layer quantum Hall systems, observing this effect becomes easier [57].

Beyond these quantum effects, combining topological insulators with magnetic or superconducting materials produces fascinating quantum phenomena. Introducing magnetism into topological insulators leads to quantum anomalous Hall effect, topological magnetoelectric effect, magnetic monopole effects, etc., while introducing superconductivity generates Majorana states. The quantum anomalous Hall effect has been realized in magnetic topological insulators, representing another important quantum effect observed in topological insulators after the quantum spin Hall effect and currently the only confirmed important effect.

4. Magnetic Topological Insulators and Quantum Anomalous Hall Effect

In the quantum spin Hall effect, the two edge states with opposite spin and propagation directions occupy the same physical location. Only when scattering between them is completely absent can dissipationless transport be achieved. Time-reversal symmetry guarantees that elastic scattering between the two edge states does not occur, but cannot prevent inelastic scattering processes that break time-reversal symmetry. Consequently, the quantum spin Hall effect can only be observed when the sample size is smaller than the inelastic scattering mean free path, greatly increasing application difficulty. In the quantum Hall effect, edge states with opposite propagation directions are located at opposite sides of the sample, and scattering between them is prohibited as long as the sample is not too small. Therefore, the quantum Hall effect is a more applicable topological quantum effect. Realizing Haldane' s vision of a zero-magnetic-field quantum Hall effect is crucial for future applications.

The Hall effect can appear without magnetic fields—this is the anomalous Hall effect in ferromagnetic materials [58,59]. Soon after the normal Hall effect was discovered, Hall observed a nonlinear dependence of Hall resistance on magnetic field strength in ferromagnetic materials, with large slopes at low fields. This strong low-field Hall effect reflects the magnetization of ferromagnetic materials and was later called the anomalous Hall effect [58]. When a ferromagnetic thin film has an easy magnetization axis perpendicular to the film plane, it can maintain spontaneous magnetization perpendicular to the film even at zero external field, making the anomalous Hall effect an important field-free phenomenon.

The anomalous Hall effect is common in magnetic materials and has been known for over a century, but its mechanism remains debated. Some believe it is mainly determined by band structure properties (intrinsic mechanism), while others attribute it primarily to impurity scattering (extrinsic mechanism) [59]. After the quantum Hall effect discovery, theorists found that the intrinsic mechanism of anomalous Hall effect has a similar expression to the quantum Hall effect and can be viewed as a non-quantized version of quantum Hall effect in ferromagnetic metals. Conversely, if a two-dimensional ferromagnetic insulator with topologically non-trivial electronic structure could be realized, its anomalous Hall resistance might become quantized, achieving zero-magnetic-field quantum Hall effect. This magnetic-field-free quantum Hall effect caused by spontaneous magnetization in ferromagnetic materials is called the quantum anomalous Hall effect (QAHE) (Fig. 6).

Theorists have proposed several ferromagnetic material systems that might realize QAHE [59,60], but experimental progress has been limited. The discovery of topological insulators brought new hope. Introducing ferromagnetism to break time-reversal symmetry in either two-dimensional or three-dimensional topological insulators could lead to QAHE [61-65]. In two-dimensional topological insulators, ferromagnetism destroys one of the spin-momentum-locked edge state pairs, transforming helical edge states into chiral edge states and converting quantum spin Hall effect into QAHE [62,64]. For three-dimensional topological insulator thin films (where side surface contributions can be neglected), introducing ferromagnetism with easy axis perpendicular to the film opens gaps at the Dirac points in both top and bottom surface states. These gapped Dirac surface states are topologically non-trivial insulators with different topological properties on opposite surfaces. Chiral edge states then appear at the film side surfaces where the two different topological phases meet, and QAHE can be observed when the Fermi level lies within both surface gaps [63,65].

Realizing QAHE requires introducing ferromagnetism into topological insulators, fabricating thin film structures, and eliminating contributions from carriers other than edge states. The greatest challenge is introducing ferromagnetism into topological insulator materials. Introducing ferromagnetism into semiconductors or insulators has been a central problem in spintronics over the past two decades, with few successful examples. For QAHE, the problem is more complex: ferromagnetism must be achieved while maintaining complete insulation, otherwise the quantum Hall effect from edge states will be overwhelmed by other conductive channels. Most ferromagnetic materials in nature are metals; ferromagnetic insulators are rare. In dilute magnetic semiconductors, bulk free carriers are essential mediators for ferromagnetism, making it impossible to maintain ferromagnetism when carriers are depleted.

Two approaches can introduce ferromagnetism into topological insulators: (1) ferromagnet/topological insulator heterointerfaces, and (2) magnetic impurity doping of topological insulators. In ferromagnet (FM)/three-dimensional topological insulator (TI)/ferromagnet (FM) sandwich structures, the top and bottom ferromagnetic layers open gaps in the topological insulator' s surface states, leading to QAHE [63]. For two-dimensional topological insulators only one or two atomic layers thick, perhaps a single ferromagnetic layer suffices to generate QAHE. The ferromagnetic layer material must be a ferromagnetic insulator to avoid creating conductive channels. In recent years, scientists have searched for suitable ferromagnetic insulator materials to realize heterostructures with topological insulators. For example, EuS is a ferromagnetic insulator with Curie temperature of 16.6 K, and Bi_2Se_3/EuS heterostructures have been prepared with induced ferromagnetism observed in Bi_2Se_3 [66]. However, the measured anomalous Hall effect is far from quantization, mainly because the electronic structure coupling between ferromagnetic and topological insulator layers is too weak to induce sufficiently strong ferromagnetism. Since ferromagnetic insulators with Curie temperatures above room temperature exist (e.g., yttrium iron garnet $Y_3Fe_5O_{12}$ with Tc = 550 K), realizing room-temperature QAHE in ferromagnetic insulator/topological insulator structures is a promising research direction.

The other approach is magnetic impurity doping, commonly used in magnetic semiconductors and insulators [67,68]. The key is finding suitable long-range ferromagnetic coupling mechanisms because direct atomic ferromagnetic coupling only acts over distances of a few tenths of nanometers, while magnetic impurity spacing in doped semiconductors or insulators is much larger. In typical Mn-doped dilute magnetic III-V semiconductors, ferromagnetism arises from RKKY interactions mediated by bulk carriers [67]. This mechanism cannot realize QAHE because ferromagnetism disappears when bulk carriers are depleted. Theories propose that Dirac surface states in three-dimensional topological insulators can also mediate RKKY-type long-range ferromagnetic exchange [69]. This Dirac surface state-induced RKKY ferromagnetism does not require bulk carriers and becomes stronger when the Fermi level approaches the Dirac point (lowest surface carrier concentration), theoretically enabling QAHE. Theoretical work by Fang, Zhang, and others showed that the inverted band structure of Bi_2Se_3 family topological insulators gives their valence electrons enormous van Vleck susceptibility even in the insulating state. Dopant magnetic moments can couple ferromagnetically through this huge van Vleck susceptibility, achieving ferromagnetism without carriers and bringing hope for QAHE in Bi_2Se_3 family materials [64].

Experimentally, since quantum spin Hall effect was realized in (Hg,Cd)Te/HgTe/(Hg,Cd)Te quantum wells, magnetic doping of this system was first attempted. In Mn-doped HgTe layers, quantized Hall effect was observed at low magnetic fields below 1 T. However, since ferromagnetic order cannot be achieved in this material, zero-field quantum Hall effect is impossible. Before the concept of three-dimensional topological insulators, magnetic element doping (V, Cr, etc.) in Sb_2Te_3 had been studied, showing good ferromagnetism [70]. Hor et al. achieved ferromagnetism with Curie temperature of 12 K and easy axis perpendicular to the cleavage plane in Mn-doped Bi_2Te_3 prepared by high-temperature sintering [71]. In Fe- or Mn-doped Bi_2Se_3, ARPES observed opened gaps at the Dirac point, but no clear evidence of long-range ferromagnetic order [72,73]. In Mn-doped $Bi_2(Se,Te)_3$, ferromagnetism was observed to strengthen with decreasing carrier concentration, suspected to be Dirac surface state-mediated RKKY ferromagnetism [74]. However, due to material quality issues, these experimental results remain far from realizing QAHE.

Using MBE technology to obtain high-quality, controllable thin films, and leveraging its non-equilibrium growth to achieve uniform, high-concentration magnetic doping in semiconductors/insulators, the Xue group at Tsinghua University systematically attempted magnetic doping of Bi_2Se_3 family topological insulators. They found that Cr doping causes minimal lattice damage and mainly substitutes for host atoms, achieving long-range ferromagnetism in both Bi_2Te_3 and Sb_2Te_3 [75]. No ferromagnetism was observed in Cr-doped Bi_2Se_3; later studies showed this absence is due to two factors: first, Cr atoms distribute non-uniformly in Bi_2Se_3, forming superparamagnetic clusters with short-range but not long-range ferromagnetic order [76]; second, at high Cr doping concentrations, Cr substitution significantly reduces the material' s spin-orbit coupling, changing the bulk band structure from inverted to normal. This transforms the system into a topologically trivial insulator phase, destroying the van Vleck magnetic coupling mechanism that depends on band inversion [77].

Cr-doped Bi_2Te_3 and Sb_2Te_3 exhibit good long-range ferromagnetic order with easy axis perpendicular to the film plane, establishing the foundation for QAHE. To finally observe QAHE, bulk carrier contributions must be eliminated. Cr doping of Bi_2Te_3 is typically electron-type doping, while Sb_2Te_3 is hole-type. Mixing them into Cr-doped $(Bi\ Sb_1\ )_2Te_3$ ternary topological insulator compounds allows effective carrier concentration control by tuning the Bi/Sb ratio. Experiments show that varying the Bi/Sb ratio can tune the carrier type from hole-type to electron-type. Regardless of carrier concentration and type, Hall resistance shows good hysteresis loops, indicating carrier-independent long-range ferromagnetic order with Curie temperature nearly independent of carrier concentration and type (Figs. 7a-f). This confirms the existence of a van Vleck mechanism-induced ferromagnetic insulator phase [75]. Since the magnetically induced gap is small (a few meV), it is difficult to position the Fermi level within the gap by chemical composition control alone. This can be achieved through field effect—applying an electric field to the film via a dielectric layer gate to control Fermi level position. Strontium titanate has a large dielectric constant at low temperatures (up to 20,000 at 2 K), making it ideal. In experiments, they directly used a 0.5 mm thick strontium titanate substrate as the gate dielectric, avoiding dielectric deposition and microfabrication processes that could damage the topological insulator thin film [78] (Figs. 7g, h).

In a 5 QL-thick Cr-doped $(\mathrm{Bi,Sb})_2\mathrm{Te}_3$ film on a strontium titanate substrate, the Xue group first observed QAHE [79]. Figure 8a [Figure 8: see original paper] shows the anomalous Hall resistance versus magnetic field at different gate voltages at 30 mK. The anomalous Hall resistance changes significantly with gate voltage, reaching a maximum of h/e^2 at -1.5 V. At this gate voltage, the Hall resistance remains unchanged from zero to high field, staying on a quantized resistance plateau. Figure 8b shows zero-field Hall resistance and longitudinal resistance versus gate voltage. The Hall resistance shows a plateau of height 1 h/e^2 near -1.5 V, while longitudinal resistance drops significantly to a minimum of 0.1 h/e^2, indicating dramatically reduced energy dissipation—a hallmark of quantum Hall states.

The non-zero longitudinal resistance arises because besides edge states, other conductive channels still exist. In conventional quantum Hall effects, magnetic fields serve two purposes: (1) creating topologically non-trivial Landau level structures, and (2) localizing electrons outside edge states so they don' t contribute to conductance. In QAHE systems, although topologically non-trivial electronic structure doesn' t require external magnetic fields, without magnetic field assistance, quantum well states and surface states still contribute to conductance, causing non-zero longitudinal resistance. Applying an external magnetic field to localize these electrons can achieve true zero resistance, which they confirmed experimentally. Figure 9 [Figure 9: see original paper] shows Hall and longitudinal resistance versus magnetic field. Except for peaks near the coercive field, longitudinal resistance gradually decreases with increasing magnetic field, dropping to zero above 10 Tesla. Meanwhile, Hall resistance remains on the h/e^2 quantum plateau, indicating the system stays in a quantum Hall state throughout. These experimental results provide definitive proof of QAHE realization.

The experimental observation of QAHE ends a 25-year pursuit of magnetic-field-free quantum Hall effects, lays the foundation for studying other quantum phenomena in topological insulators, and makes the dissipationless edge states of quantum Hall effects promising for electron transport applications.

5. Applications of Quantum Spin Hall and Quantum Anomalous Hall Effects in Low-Energy Electronics

The quantum spin Hall effect in two-dimensional topological insulators and the quantum anomalous Hall effect in magnetic topological insulators both feature edge states that can conduct electrons without dissipation. Their successive experimental realizations allow serious consideration of their potential applications in electronics. Below we briefly discuss the issues that must be resolved for these effects to become applicable, which depend on further improvements in topological insulator material properties—a major future direction for the field.

5.1 Temperature

The biggest challenge for applying quantum spin Hall and quantum anomalous Hall effects is that they still require very low temperatures. Both effects were first observed below 100 mK. Recent reports of quantum spin Hall effect at 30 K remain far from practical temperatures like liquid nitrogen temperature, and far below the highest superconducting transition temperatures. For these topological quantum effects to become practical, they must be realized at higher temperatures, ideally room temperature.

Low temperatures primarily serve to suppress conductive channels other than dissipationless edge states. In quantum spin Hall or anomalous Hall thin films, besides edge states, there exist quantum well states (quantized bulk bands due to finite size effects) and surface states that dissipate energy when conducting electrons. Edge states lie energetically within the gaps of these quantum well or surface states. At absolute zero, when the Fermi level is in the gap and only crosses edge states, only edge states conduct. At non-zero temperatures, carriers in quantum well or surface states are thermally excited, and edge state electrons can also be excited into these states. When contributions from quantum well and surface states approach or exceed those from edge states, the system' s energy dissipation becomes non-negligible, masking quantum spin Hall or anomalous Hall signals.

The key solution is increasing the gap size. For quantum spin Hall effect, the gap is the energy difference between the bottom of conduction band quantum well states and the top of valence band quantum well states. This gap size is determined by the material' s spin-orbit coupling strength and specific material properties. As mentioned earlier, theoretical calculations have predicted several two-dimensional topological insulators with larger gaps up to hundreds of meV. Experimentally, Du et al. increased the gap in AlSb/InAs/GaSb/AlSb quantum wells through strain tuning, observing clear quantum spin Hall plateaus at 30 K. This significantly raises the temperature and brings hope for applications.

For QAHE systems, gap size depends on two factors: the bulk gap of the topological insulator material (determined by spin-orbit coupling strength) and the Curie temperature of ferromagnetism in the material. Many topological insulators have bulk gaps up to hundreds of meV, so the key to raising QAHE temperature lies in increasing the ferromagnetic Curie temperature. Currently, QAHE has only been observed in magnetic-doped Bi_2Se_3 family thin films. Early theoretical and experimental work suggested Curie temperatures could exceed liquid nitrogen temperature but would be difficult to surpass room temperature. Room-temperature QAHE is more likely to be realized in heterostructures of topological insulators and ferromagnetic insulators, since ferromagnetic insulators with Curie temperatures above room temperature exist (e.g., yttrium iron garnet with Tc = 550 K). This brings hope for room-temperature QAHE, though experimental research in this direction remains preliminary.

Note that QAHE in Cr-doped $(Bi,Sb)_2Te_3$ was observed below 100 mK, far below the sample' s ferromagnetic Curie temperature (15 K). The reason is not fully understood. One possibility is that in magnetic-doped semiconductors/insulators, ferromagnetism is not as uniform as in conventional ferromagnets, so the effective gap temperature is much lower than the Curie temperature.

Besides increasing the gap, another possible approach to raise observation temperature is increasing film disorder. According to condensed matter theory, when metallic disorder becomes sufficiently strong, electron motion becomes localized, transforming the metal into an insulator (Anderson insulator). Edge states in quantum spin/anomalous Hall films cannot backscatter, so their conductive properties are unaffected by impurities and disorder. Quantum well and surface state electrons are affected by disorder, so introducing disorder may allow observation of quantum anomalous Hall effect at higher temperatures. Theoretical work suggests that in magnetically doped three-dimensional topological insulators, surface states become localized by impurities, and QAHE should appear as long as the Fermi level is in the bulk gap. Du et al. recently found that doping AlSb/InAs/GaSb/AlSb quantum wells with Si impurities can facilitate the emergence of quantum spin Hall effect. Disorder, dimensionality, and localization are fundamental theoretical issues in condensed matter physics. Systematic study of the relationship between quantum spin/anomalous Hall effects and disorder, dimensionality, and localization is crucial for applications and will advance condensed matter theory.

For quantum spin Hall effect, another factor is the electron inelastic scattering mean free path. The prerequisite for quantum spin Hall effect is that electrons do not scatter between edge states with opposite spin and propagation directions. Time-reversal invariance of two-dimensional topological insulators prevents such transitions through elastic scattering, but inelastic scattering processes, which break time-reversal symmetry, may cause inter-edge-state transitions and destroy the quantum spin Hall effect. Inelastic scattering is mainly caused by lattice vibrations (phonons). To raise temperature, materials with weak electron-phonon coupling should be selected, and sample sizes must be smaller than the inelastic scattering mean free path. For QAHE, edge states with opposite propagation directions are at different sample edges, so inelastic scattering is not a concern.

5.2 Magnetic Field

Both quantum spin Hall and quantum anomalous Hall effects 原则上 do not require external magnetic fields. However, experimental results for QAHE in magnetically doped topological insulator thin films still require a magnetic field to achieve completely dissipationless transport. Possible reasons are: (1) quantum well and surface states still contribute to conductance at zero field, and magnetic fields help localize these topologically trivial electronic states; (2) magnetic fields help suppress magnetic disorder and increase the effective gap. Thus, reducing the required magnetic field and raising QAHE temperature share the same approach: increasing material insulation and ferromagnetism.

5.3 Contact Between Edge States and Electrodes

Although edge states in quantum spin Hall or quantum anomalous Hall effects are themselves resistanceless, contact resistance at their connection to electrodes cannot be eliminated. Even for perfect contacts, the contact resistance between each edge state and electrodes will not be lower than h/e^2. Therefore, using quantum spin/anomalous Hall edge states as wires cannot achieve completely dissipationless electron transport. This contact resistance has two characteristics: first, it exists only at contact points, so unlike ordinary resistance, it does not scale with sample length or cross-sectional area—even for very long quantum spin/anomalous Hall samples, resistance does not increase; second, each edge state' s contact resistance is h/e^2 (for perfect contacts). By paralleling as many edge states as possible, total contact resistance can be effectively reduced, enabling low-energy electron transport. Three approaches exist:

(1) Higher-order quantum anomalous Hall effect
In conventional quantum Hall effects, higher-order quantum Hall effects can be obtained by changing magnetic fields, increasing edge state number. Current QAHE systems contain only one edge state. Higher-order QAHE can be achieved by tuning material electronic structure. J. Wang et al. predicted that doping Cr-doped $(Bi,Sb)_2Te_3$ thin films with Se could reduce spin-orbit coupling, potentially realizing multi-edge-state QAHE systems under appropriate thickness [80].

(2) Superlattice structures
If suitable ordinary insulator materials can be found, quantum spin/anomalous Hall thin films can be made into multi-period superlattice structures with ordinary insulator thin films, achieving parallel structures of multiple quantum spin/anomalous Hall systems. Recently, theorists predicted that some materials naturally form such superlattice structures: Dirac semimetals and Weyl semimetals. Quantum thin films of these materials are respectively superlattices of quantum spin Hall films and quantum anomalous Hall films with ordinary insulator films [81-84].

(3) Wire array structures
Since contact resistance between electrodes and edge states is independent of film geometry, quantum spin/anomalous Hall thin films can be etched into parallel fine wire structures, creating large numbers of paralleled edge states. Such wire arrays can be fabricated very densely—thin films centimeters wide can be processed into tens or hundreds of thousands of parallel wires, greatly reducing

contact resistance effects. Future quantum spin/anomalous Hall effect-based wires will likely be such dense wire array structures.

In summary, many issues must be resolved before quantum spin/anomalous Hall edge states can be applied in low-energy power transmission, representing an important future research direction.

6. Other Topological Quantum Materials

Beyond topological insulators and magnetic topological insulators, several other topological quantum materials have been proposed, all having important research or application value and currently being hot topics in the field.

6.1 Topological Superconductors

Topological superconductors are among the hottest research directions in condensed matter physics because Majorana bound states that may appear in them can be used for error-tolerant quantum computation [85]. Quantum computing shows amazing computational power for many tasks, but the main obstacle is errors caused by quantum state decoherence. Majorana bound states' unique properties can solve this problem. Majorana states might be realized in 5/2 fractional quantum Hall states, p-wave superconductors, and B-phase He-3 superfluid, but sample preparation and measurement are very difficult and results remain controversial. Theorists predicted that superconducting proximity effects in heterojunctions of ordinary s-wave superconductors and topological insulator thin films can turn topological insulators into topological superconductors [86]. Subsequently, several similar structures were proposed, such as heterojunctions of s-wave superconductors and semiconductor thin films with strong spin-orbit coupling. These theoretical advances greatly promoted topological superconductor research [85].

Several experiments have reported superconducting proximity effects in topological insulators or strong spin-orbit coupling semiconductors. Jia et al. observed proximity-induced superconducting gaps and vortices under magnetic fields in topological insulator Bi_2Se_3 thin films on superconductor $NdSe_2$ using scanning tunneling spectroscopy [87]. In heterojunctions of high-temperature superconductor BSCCO and topological insulator Bi_2Se_3, the proximity-induced superconductivity in Bi_2Se_3 was found to have s-wave symmetry, suggesting high-temperature superconductors could also be used to obtain topological superconductors, potentially raising the realization temperature significantly [88]. Several groups observed zero-energy conductance peaks in proximity-induced superconducting gaps and attributed them to signatures of Majorana bound states, including results from Kouwenhoven's group in InSb nanowires [89]. Since many physical processes can produce zero-energy conductance peaks, whether they are truly related to Majorana states remains controversial.

6.2 Topological Crystalline Insulators

Three-dimensional topological insulators' topological non-trivial properties are protected by time-reversal symmetry. L. Fu et al. predicted a class of topological insulators protected by crystal symmetry, called topological crystalline insulators [90]. The gapless surface states of topological crystalline insulators only appear on surfaces with specific symmetries. Since crystal symmetry is more easily altered by electric fields and strain, topological crystalline insulators are readily applicable to field-effect and pressure-sensing devices. They can even appear in materials with weak spin-orbit coupling, where gapless surface states show parabolic rather than Dirac-like linear dispersion. Both theoretically and experimentally, Pb_1 Sn Se(Te) has been identified as a topological crystalline insulator at specific compositions and low temperatures [91-93]. Introducing ferromagnetism into topological crystalline insulators may produce higher-order QAHE [94].

6.3 Strongly Correlated Topological Insulators and Fractional Quantum Anomalous Hall Effect

Most topological insulator theories are based on band theory without considering electron-electron correlation. Known topological insulator materials have weak electron-electron correlation, and their electronic structures basically match band theory predictions. Strong electron correlation brings rich physical phenomena, problems, and properties that are difficult to predict theoretically. For copper-based high-temperature superconductors, many researchers believe strong electron correlation plays an important role in the high superconducting transition temperature, making the effect of electron correlation on topological insulator electronic properties a very interesting question.

Compounds containing rare earth elements often have strong electron-electron interactions due to f-electron properties. This is a typical strongly correlated system in condensed matter physics, called heavy fermion materials (metals) or Kondo insulators (insulators). Theorists predicted that some Kondo insulators have topologically non-trivial electronic structures and gapless surface states, called topological Kondo insulators [95]. Topological Kondo insulators provide a platform to study the interplay between topological electronic states and electron correlation. Recent theoretical and experimental work indicates that SmB_6 and YB_6 are topological Kondo insulators [96,97]. Since rare earth compounds often exhibit novel superconducting and magnetic properties, research on topological Kondo insulators is expected to yield very rich results.

Fractional quantum Hall effects are caused by electron correlation. If strong electron correlation can be introduced into magnetic topological insulators, fractional quantum anomalous Hall effect—zero-field fractional quantum Hall effect —might be obtained. According to recent theoretical work, if a QAHE system has a very flat band whose energy width is smaller than electron correlation energy, electron correlation will lead to fractional QAHE [98-100]. Such topologically non-trivial flat band structures might be realized in organic thin films [101].

6.4 Dirac Semimetals and Weyl Semimetals

If the spin-orbit coupling strength of a three-dimensional topological insulator material is gradually reduced, the bulk gap first decreases to zero then increases again. This is a topological phase transition from three-dimensional topological insulator to ordinary insulator. At the transition point, the conduction band minimum and valence band maximum intersect at a point in momentum space, forming a gapless three-dimensional Dirac cone—generalizing graphene' s electronic structure to three dimensions—called a Dirac semimetal with many interesting properties [82,83]. Thin films of Dirac semimetals naturally form superlattice structures of quantum spin Hall films and ordinary topological insulators [82]. Experimentally, topological phase transitions can be achieved by doping topological insulators with lighter elements, though precisely tuning to the zero-gap transition is challenging. Some materials have crystal symmetry-protected three-dimensional Dirac points robust against parameter changes; Na_3Bi and Cd_3As_2 belong to this class of symmetry-protected Dirac semimetals [84,102,103].

Introducing ferromagnetism into a Dirac semimetal transforms it into another topological material: a Weyl semimetal [81,82]. This is an even more interesting state of matter, previously predicted in A-phase He-3 superfluid but never experimentally confirmed. Weyl semimetal band structures contain pairs of singularities in momentum space called Weyl points—essentially magnetic monopoles in momentum space. Some surfaces of Weyl semimetals exhibit "Fermi arc" surface states. Like Dirac semimetals, Weyl semimetal quantum thin films form superlattice structures of QAHE films and topological insulator films. Recent calculations by Fang' s group show that $HgCr_2Se_4$ is a Weyl semimetal [104], while pyrochlore oxide $RE_2Ir_2O_7$ (RE = rare earth) is a strongly correlated Weyl semimetal [81]. Dirac and Weyl semimetals are also hot topics in current topological quantum materials research.

7. Conclusion

Research progress on topological insulators over the past decade has greatly enriched the field of topological quantum materials that began with quantum Hall effects. Researchers from physics, materials science, chemistry, electronics, and other disciplines have begun paying attention to this field. Although topological quantum materials are still far from practical application, their relatively clear physical understanding and rich material choices offer hope for rapid future development. Materials science research is particularly important for further development because novel quantum phenomena in topological quantum materials are mainly based on band theory (except for strongly correlated topological Kondo insulators and fractional QAHE). Therefore, the key to experimentally realizing and applying these properties and quantum phenomena lies in obtaining high-quality, tunable materials. Conversely, realizing applicable topological quantum materials will also greatly advance materials science.

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References

1. Dirac P. A. M., *The Principles of Quantum Mechanics* (Oxford University Press, USA)
2. Feng D., Jin G. J., *Condensed Matter Physics* (Beijing, Higher Education Press, 2013)
3. Klitzing K. V., Dorda G., Peper M., New method for high-accuracy determination of the fine-structure constant based on quantized Hall resistance, *Physical Review Letters* **45**, 494 (1980)
4. Hall E. H., On a new action of the magnet on electric currents, *American Journal of Mathematics* **2**, 287 (1879)
5. Tsui D. C., Stormer H. L., Gossard A. C., Two-dimensional magnetotransport in the extreme quantum limit, *Physical Review Letters* **48**, 1559 (1982)
6. von Klitzing K., The quantized Hall effect, *Physics Today* **56**(7), 38 (2003)
7. Thouless D. J., Kohmoto M., Nightingale M. P., den Nijs M., Quantized Hall conductance in a two-dimensional periodic potential, *Physical Review Letters* **49**, 405 (1982)
8. Haldane F. D. M., Model for a quantum Hall effect without Landau levels: condensed-matter realization of the "parity anomaly" , *Physical Review Letters* **61**, 2015 (1988)
9. Novoselov K. S., Geim A. K., Morozov S. V., et al., Electric field effect in atomically thin carbon films, *Science* **306**(5696), 666 (2004)
10. Kane C. L., Mele E. J., Quantum spin Hall effect in graphene, *Physical Review Letters* **95**, 226801 (2005)
11. Bernevig B. A., Zhang S. C., Quantum spin Hall effect, *Physical Review Letters* **96**, 106802 (2006)
12. Fu L., Kane C. L., Mele E. J., Topological insulators in three dimensions, *Physical Review Letters* **98**, 106803 (2007)
13. Fu L., Kane C. L., Topological insulators with inversion symmetry, *Physical Review B* **76**, 045302 (2007)
14. Hasan M. Z., Kane C. L., Colloquium: Topological insulators, *Reviews of Modern Physics* **82**(4), 3045 (2010)

15. Qi X. L., Zhang S. C., Topological insulators and superconductors, *Reviews of Modern Physics* **83**, 1057 (2011)
16. Bernevig B. A., Hughes T. L., Zhang S. C., Quantum spin Hall effect and topological phase transition in HgTe quantum wells, *Science* **314**(5806), 1757 (2006)
17. Hinz J., Buhmann H., Schäfer M., et al., Gate control of the giant Rashba effect in HgTe quantum wells, *Semiconductor Science and Technology* **21**(4), 501 (2006)
18. König M., Wiedmann S., Brüne C., et al., Quantum spin Hall insulator state in HgTe quantum wells, *Science* **318**(5851), 766 (2007)
19. Roth A., Brüne C., Buhmann H., et al., Nonlocal transport in the quantum spin Hall state, *Science* **325**(5938), 294 (2009)
20. Liu C., Hughes T. L., Qi X. L., et al., Quantum spin Hall effect in inverted type-II semiconductors, *Physical Review Letters* **100**, 236601 (2008)
21. Knez I., Du R. R., Sullivan G., Evidence for helical edge modes in inverted InAs/GaSb quantum wells, *Physical Review Letters* **107**, 136603 (2011)
22. Murakami S., Quantum spin Hall effect and enhanced magnetic response by spin-orbit coupling, *Physical Review Letters* **97**, 236805 (2006)
23. Liu Z., Liu C. X., Wu Y. S., et al., Stable nontrivial Z_2 topology in ultrathin Bi(111) films: A first-principles study, *Physical Review Letters* **107**, 136805 (2011)
24. Liu C. C., Feng W., Yao Y., Quantum spin Hall effect in silicene and two-dimensional germanium, *Physical Review Letters* **107**, 076802 (2011)
25. Xu Y., Yan B., Zhang H. J., et al., Large-gap quantum spin Hall insulators in tin films, *Physical Review Letters* **111**, 136804 (2013)
26. Hirahara T., Bihlmayer G., Sakamoto Y., et al., Interfacing 2D and 3D topological insulators: Bi(111) bilayer on Bi_2Te_3, *Physical Review Letters* **107**, 166801 (2011)
27. Yang F., Miao L., Wang Z. F., et al., Spatial and energy distribution of topological edge states in single Bi(111) bilayer, *Physical Review Letters* **109**, 016801 (2012)
28. Vogt P., Padova P. D., Quaresima C., et al., Silicene: compelling experimental evidence for graphenelike two-dimensional silicon, *Physical Review Letters* **108**, 155501 (2012)
29. Chen L., Liu C. C., Feng B., et al., Evidence for Dirac fermions in a honeycomb lattice based on silicon, *Physical Review Letters* **109**, 056804 (2012)
30. Weng H., Dai X., Fang Z., Transition-metal pentatelluride $ZrTe_5$ and $HfTe_5$: A paradigm for large-gap quantum spin Hall insulators, *Physical Review X* **4**, 011002 (2014)
31. Hsieh D., Qian D., Wray L., et al., A topological Dirac insulator in a quantum spin Hall phase, *Nature* **452**(7190), 970 (2008)
32. Zhang H. J., Liu C. X., Qi X. L., et al., Topological insulators in Bi_2Se_3, Bi_2Te_3 and Sb_2Te_3 with a single Dirac cone on the surface, *Nature Physics* **5**(6), 438 (2009)
33. Xia Y., Qian D., Hsieh D., et al., Observation of a large-gap topological-

insulator class with a single Dirac cone on the surface, *Nature Physics* **5**(6), 398 (2009)

34. Chen Y. L., Analytis J. G., Chu J. H., et al., Experimental realization of a three-dimensional topological insulator, Bi_2Te_3, *Science* **325**(5937), 178 (2009)
35. Yan B., Zhang S. C., Topological materials, *Reports on Progress in Physics* **75**(9), 096501 (2012)
36. Liu C. X., Zhang H. J., Yan B., et al., Oscillatory crossover from two-dimensional to three-dimensional topological insulators, *Physical Review B* **81**, 041307(R) (2010)
37. Dai X., Hughes T. L., Qi X. L., et al., Helical edge and surface states in HgTe quantum wells and bulk insulators, *Physical Review B* **77**, 125319 (2008)
38. Brüne C., Liu C. X., Novik E. G., et al., Quantum Hall effect from the topological surface states of strained bulk HgTe, *Physical Review Letters* **106**, 126803 (2011)
39. Hor Y. S., Richardella A., Roushan P., et al., p-type Bi_2Te_3 for topological insulator and low-temperature thermoelectric applications, *Physical Review B* **79**, 195208 (2009)
40. Hsieh D., Xia Y., Qian D., et al., A tunable topological insulator in the spin helical Dirac transport regime, *Nature* **460**(7259), 1101 (2009)
41. Kuroda K., Arita M., Miyamoto K., et al., Hexagonally deformed Fermi surface of the 3D topological insulator Bi_2Se_3, *Physical Review Letters* **105**, 076802 (2010)
42. Zhang J., Chang C. Z., Zhang Z., et al., Band structure engineering in $(Bi_1\ Sb\ )_2Te_3$ ternary topological insulators, *Nature Communications* **2**(12), 574 (2011)
43. Kong D., Chen Y., Cha J. J., et al., Ambipolar field effect in the ternary topological insulator $(Bi\,Sb_1\ )_2Te_3$ by composition tuning, *Nature Nanotechnology* **6**(11), 705 (2011)
44. Ren Z., Taskin A. A., Sasaki S., et al., Large bulk resistivity and surface quantum oscillations in the topological insulator Bi_2Te_2Se, *Physical Review B* **82**, 241306 (2010)
45. Xiong J., Petersen A. C., Qu D., et al., Quantum oscillations in a topological insulator Bi_2Te_2Se with large bulk resistivity ($6\ \Omega \cdot cm$), *Physica E* **44**(5), 917 (2012)
46. Arakane T., Sato T., Souma S., et al., Tunable Dirac cone in the topological insulator $Bi_2\ Sb\,Te_3\ Se$, *Nature Communications* **3**(1), 636 (2011)
47. Li Y. Y., Wang G., Zhu X. G., et al., Intrinsic topological insulator Bi_2Te_3 thin films on Si and their thickness limit, *Advanced Materials* **22**(36), 4002 (2010)
48. Song C. L., Wang Y. L., Jiang Y. P., et al., Topological insulator Bi_2Se_3 thin films grown on double-layer graphene by molecular beam epitaxy, *Applied Physics Letters* **97**(14), 143118 (2010)
49. Zhang Y., He K., Chang C. Z., et al., Crossover of the three-dimensional topological insulator Bi_2Se_3 to the two-dimensional limit, *Nature Physics*

6(8), 584 (2010)

50. Zhang G., Qin H. J., Chen J., et al., Growth of topological insulator Bi_2Se_3 thin films on $SrTiO_3$ with large tunability in chemical potential, *Advanced Functional Materials* **21**(13), 2351 (2011)
51. Roushan P., Seo J., Parker C. V., et al., Topological surface states protected from backscattering by chiral spin texture, *Nature* **460**(7259), 1106 (2009)
52. Zhang T., Cheng P., Chen X., et al., Experimental demonstration of topological surface states protected by time-reversal symmetry, *Physical Review Letters* **103**, 266803 (2009)
53. Cheng P., Song C. L., Zhang T., et al., Landau quantization of topological surface states in Bi_2Se_3, *Physical Review Letters* **105**, 076801 (2010)
54. Hanaguri T., Igarashi K., Kawamura M., et al., Momentum-resolved Landau-level spectroscopy of Dirac surface state in Bi_2Se_3, *Physical Review B* **82**, 081305(R) (2010)
55. Jiang Y., Wang Y. L., Chen M., et al., Landau quantization and the thickness limit of topological insulator thin films of Sb_2Te_3, *Physical Review Letters* **108**, 016401 (2012)
56. Schlenk T., Böttner M., Kern K., et al., Landau levels of topological surface states in Bi_2Te_3, *Physical Review Letters* **110**, 216801 (2013)
57. Seradjeh B., Moore J. E., Franz M., Exciton condensation and charge fractionalization in a topological insulator film, *Physical Review Letters* **103**, 066402 (2009)
58. Hall E. H., On the new action of magnetism on electric currents, *Philosophical Magazine* **12**, 157 (1881)
59. Nagaosa N., Sinova J., Onoda S., et al., Anomalous Hall effect, *Reviews of Modern Physics* **82**(2), 1539 (2010)
60. Onoda M., Nagaosa N., Quantized anomalous Hall effect in two-dimensional ferromagnets: Quantum Hall effect in metals, *Physical Review Letters* **90**, 206601 (2003)
61. Qi X. L., Wu Y. S., Zhang S. C., Topological quantization of the spin Hall effect in two-dimensional paramagnetic semiconductors, *Physical Review B* **74**, 085308 (2006)
62. Liu C. X., Qi X. L., Dai X., et al., Quantum anomalous Hall effect in Hg_1 Mn Te quantum wells, *Physical Review Letters* **101**, 146802 (2008)
63. Qi X. L., Hughes T. L., Zhang S. C., Topological field theory of time-reversal invariant insulators, *Physical Review B* **78**, 195424 (2008)
64. Yu R., Zhang W., Zhang H. J., et al., Quantized anomalous Hall effect in magnetic topological insulators, *Science* **329**(5987), 61 (2010)
65. Wang J., Lian B., Zhang S. C., Universal scaling of the quantum anomalous Hall plateau transition, *Physical Review B* **89**, 085106 (2014)
66. Wei P., Katmis F., Assaf B. A., et al., Exchange-coupling-induced symmetry breaking in topological insulator/ferromagnet heterostructures, *Physical Review Letters* **110**, 186807 (2013)
67. Ohno H., Chiba D., Matsukura F., et al., Electric-field control of ferromagnetism in (Ga,Mn)As, *Nature* **408**(6815), 944 (2000)

68. Dietl T., Ohno H., Matsukura F., et al., Zener model description of ferromagnetism in zinc-blende magnetic semiconductors, *Science* **287**(5455), 1019 (2000)
69. Liu Q., Liu C. X., Xu C., et al., Magnetic impurities on the surface of a topological insulator, *Physical Review Letters* **102**, 156603 (2009)
70. Chien Y. J., *Transition Metal-Doped Sb_2Te_3 and Bi_2Te_3 Diluted Magnetic Semiconductors*, Thesis of the University of Michigan (2007)
71. Hor Y. S., Roushan P., Beidenkopf H., et al., Development of ferromagnetism in the doped topological insulator Bi_2 Mn Te_3, *Physical Review B* **81**, 195203 (2010)
72. Chen Y. L., Chu J. H., Analytis J. G., et al., Massive Dirac fermion on the surface of a magnetically doped topological insulator, *Science* **329**(5992), 659 (2010)
73. Xu S. Y., Neupane M., Liu C., et al., Hedgehog spin texture and Berry' s phase tuning in a magnetic topological insulator, *Nature Physics* **8**(8), 616 (2012)
74. Checkelsky J. G., Ye J., Onose Y., et al., Dirac-fermion-mediated ferromagnetism in a topological insulator, *Nature Physics* **8**(10), 729 (2012)
75. Chang C. Z., Zhang J., Liu M., et al., Thin films of magnetically doped topological insulator with carrier-independent long-range ferromagnetic order, *Advanced Materials* **25**(7), 1065 (2013)
76. Chang C. Z., Tang P., Wang Y. L., et al., Chemical-potential-dependent gap opening at the Dirac surface states of Bi_2Se_3 induced by aggregated substitutional Cr atoms, *Physical Review Letters* **112**, 056801 (2014)
77. Zhang J., Chang C. Z., Tang P., et al., Topology-driven magnetic quantum phase transition in topological insulators, *Science* **339**(6127), 1582 (2013)
78. Chen J., Qin H. J., Yang F., et al., Gate-voltage control of chemical potential and weak antilocalization in Bi_2Se_3, *Physical Review Letters* **105**, 176602 (2010)
79. Chang C. Z., Zhang J., Feng X., et al., Experimental observation of the quantum anomalous Hall effect in a magnetic topological insulator, *Science* **340**(6129), 167 (2013)
80. Wang J., Lian B., Zhang S. C., Universal scaling of the quantum anomalous Hall plateau transition, *Physical Review B* **89**, 085106 (2014)
81. Wan X., Turner A. M., Vishwanath A., et al., Topological semimetal and Fermi-arc surface states in the electronic structure of pyrochlore iridates, *Physical Review B* **83**, 205101 (2011)
82. Burkov A. A., Hook M. D., Balents L., Topological nodal semimetals, *Physical Review B* **84**, 235126 (2011)
83. Young S. M., Zaheer S., Teo J. C. Y., et al., Dirac semimetal in three dimensions, *Physical Review Letters* **108**, 140405 (2012)
84. Wang Z., Sun Y., Chen X. Q., et al., Dirac semimetal and topological phase transitions in A_3Bi (A = Na, K, Rb), *Physical Review B* **85**, 195320 (2012)
85. Nayak C., Simon S. H., Stern A., et al., Non-Abelian anyons and topological quantum computation, *Reviews of Modern Physics* **80**(3), 1083 (2008)

86. Fu L., Kane C. L., Superconducting proximity effect and Majorana fermions at the surface of a topological insulator, *Physical Review Letters* **100**, 096407 (2008)
87. Wang M. X., Liu C., Xu J. P., et al., The coexistence of superconductivity and topological order in the Bi_2Se_3 thin films, *Science* **336**(6077), 52 (2012)
88. Wang E., Ding H., Fedorov A. V., et al., Fully gapped topological surface states in Bi_2Se_3 films induced by a d-wave high-temperature superconductor, *Nature Physics* **9**(10), 621 (2013)
89. Mourik V., Zuo K., Frolov S. M., et al., Signatures of Majorana fermions in hybrid superconductor-semiconductor nanowire devices, *Science* **336**(6084), 1003 (2012)
90. Fu L., Topological crystalline insulators, *Physical Review Letters* **106**, 106802 (2011)
91. Hsieh T. H., Lin H., Liu J., et al., Topological crystalline insulators in the SnTe material class, *Nature Communications* **3**(7), 982 (2012)
92. Tanaka Y., Ren Z., Sato T., et al., Experimental realization of a topological crystalline insulator in SnTe, *Nature Physics* **8**(11), 800 (2012)
93. Dziawa P., Kowalski B. J., Dybko K., et al., Topological crystalline insulator states in Pb_1 Sn Se, *Nature Materials* **11**(12), 1023 (2012)
94. Fang C., Gilbert M. J., Bernevig B. A., Large-Chern-number quantum anomalous Hall effect in thin-film topological crystalline insulators, *Physical Review Letters* **112**, 046801 (2014)
95. Dzero M., Sun K., Galitski V., et al., Topological Kondo insulators, *Physical Review Letters* **104**, 106408 (2010)
96. Lu F., Zhao J., Weng H., et al., Correlated topological insulators with mixed valence, *Physical Review Letters* **110**, 096401 (2013)
97. Weng H., Zhao J., Wang Z., et al., Topological crystalline Kondo insulator in mixed valence ytterbium borides, *Physical Review Letters* **112**, 016403 (2014)
98. Neupert T., Santos L., Chamon C., et al., Fractional quantum Hall states at zero magnetic field, *Physical Review Letters* **106**, 236804 (2011)
99. Tang E., Mei J. W., Wen X. G., High-temperature fractional quantum Hall states, *Physical Review Letters* **106**, 236802 (2011)
100. Sheng D. N., Gu Z. C., Sun K., et al., Fractional quantum Hall effect in the absence of Landau levels, *Nature Communications* **2**, 389 (2011)
101. Liu Z., Wang Z. F., Mei J. W., et al., Flat Chern band in a two-dimensional organometallic framework, *Physical Review Letters* **110**, 106804 (2013)
102. Liu Z. K., Zhou B., Zhang Y., et al., Discovery of a three-dimensional topological Dirac semimetal, Na_3Bi, *Science* **343**(6173), 864 (2014)
103. Neupane M., Xu S. Y., Sankar R., et al., Observation of a three-dimensional topological Dirac semimetal phase in high-mobility Cd_3As_2, *Nature Communications* **5**, 3786 (2014)
104. Xu G., Weng H., Wang Z., et al., Chern semimetal and the quantized anomalous Hall effect in $HgCr_2Se_4$, *Physical Review Letters* **107**, 186806 (2011)