

Literature Review

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Interfacing topological insulators surface states with ferroics

Contents

1	Introduction	2
2	Discovery of topological insulators	3
2.1	History through Hall effects	3
2.1.1	Quantum Hall effect	3
2.1.2	Berry Phase	5
2.1.3	Quantum spin Hall effect	6
2.1.4	Topological insulators	6
2.2	Relevant theory and phenomena	7
2.2.1	Topological field theory	7
2.2.2	Dirac fermions	7
3	Topological insulator physics	8
3.1	Topological invariants	8
3.1.1	TKNN invariant	8
3.1.2	Z_2 invariant	8
3.1.3	Z_2 invariant in 3D	8
3.2	Spin-orbit coupling	8
3.2.1	Dresselhaus effect	9
3.2.2	Rashba effect	9
3.3	Hamiltonians of TI Materials	9
3.3.1	HeTe (Mercury Telluride)	10
3.3.2	Bi_2Te_3 (Bismuth Telluride)	10
4	Topological insulator materials	10
4.1	$\text{CdTe}/\text{HgTe}/\text{CdTe}$ quantum wells	10
4.2	$\text{Bi}_{1-x}\text{Sb}_x$	11
4.3	$\{\text{Bi},\text{Sb}\}_2\{\text{Se},\text{Te}\}_3$ family	12
4.3.1	Bi_2Te_3	13
4.3.2	Bi_2Se_3	14

5 Ferromagnetic Insulators	16
5.1 2D FIs	16
5.1.1 Chromium triiodide (CrI_3)	16
5.1.2 $\text{Cr}_2\text{Ge}_2\text{Te}_6$ (CGT)	16
5.1.3 Magnesium Titinate (MgTiO_3 , MTO)	18
5.1.4 Other 2D ferromagnetic materials	18
6 TI & FI heterostructures	18
6.1 Spin Torque Devices	19
6.1.1 Torque	20
6.2 Quantum Anomalous Hall Effect (QAHE)	20
6.3 Experiments	20
6.3.1 Conductive ferromagnetic films	20
6.3.2 Insulating ferromagnetic films	21
6.3.3 Not-quite TI-Ferromagnetic heterostructures	21
7 Experimental methods	21
7.1 Synthesis, Growth and Fabrication	21
7.1.1 MBE Growths	21
7.1.2 Doping	21
7.2 Spectroscopic Metrological Methods	22
7.2.1 ARPES	22
7.2.2 Spin polarized STS	23
7.3 Transport Metrological Methods	24
7.3.1 Quantum Oscillations	24
7.3.2 Weak localisation and anti-localisation	24
7.3.3 Magnetization	24
8 Quotes	25
9 Bibliography	25

1 Introduction

This review of existing literature is to support my Ph.D. thesis  experimentally probing the electronic transport of interfaces between topological insulators and ferroics. I will use this document to review the field of topological insulators, ferroics  (ferromagnetic insulators in particular) and resulting heterostructures.

In the last two decades there has been a vast discovery of new materials that exhibit phenomenal properties interesting for condensed matter physics research. Us  these materials are stumbled upon experimentally, however in the case of topological materials  the setting and expectation was purely theoretical.

Topological insulators (TIs) attract interest because of their unique properties. 2D and 3D TIs have edge and surface electronic states respectively, and these states exhibit useful properties such as momentum-spin locking (or localised  spin densities) and suppressed back-scattering, as well as exhibit Dirac fermions, similar to graphene. These properties may be used for applications  like directing currents that spin-polarised as well as robust to disorder, with low resistance.

This work ties into the research themes of FLEET (Future Low Energy Electronic Technologies) and the government funding goals to enable new, low energy computing technology. 

2 Discovery of topological insulators

2.1 History through Hall effects

To understand where topological insulators (TIs) have come from, it is useful to step back through the history of some significant physics phenomena.

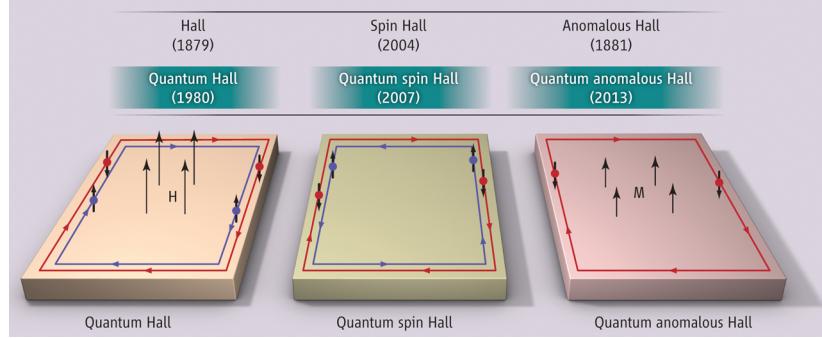


Figure 1: The three QH states discovered thus far. Source: Oh, Seongshik. [1], Science Vol. 340, 153 (2013)

2.1.1 Quantum Hall effect

The quantum hall effect (QHE) was first discovered in MOSFET¹ transistors 1980^[2], through a 2 dimensional electron gas (2DEG) found at the interface of a bulk semiconductor and the gate oxide.

When introducing a magnetic field to materials, electrons bands undergo Landau quantisation. A physical understanding for this behaviour can be drawn from the quantisation of cyclotron orbits for charged particles in magnetic fields². Landau quantisation has the effect of creating new bands called "Landau Levels", that each posses large numbers of orbitals. The degeneracy of the level goes as

$$\text{Degeneracy} = \frac{B \times A}{\phi_0} = \frac{B \times A}{h/e} \quad (1)$$

For some Fermi energy, the magnetic field can be chosen to an appropriate value to fill these Landau levels.

¹Metal-oxide semiconductor field effect transistors

²The reason for cyclotron orbits is due to the single valued electron wavefunction, ie $\oint \vec{P} \cdot d\vec{r} = 2\pi N$

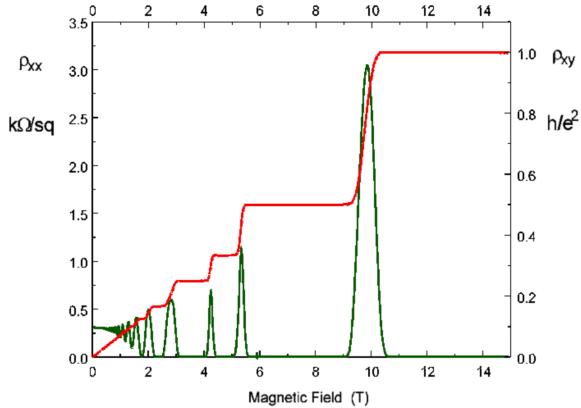


Figure 2: The quantum Hall effect. ρ_{xy} (red) is the Hall resistivity which obeys quantised values. ρ_{xx} (green) vanishes at each Landau level, before peaking when scattering to bulk states can occur.

Courtesy of D.R. Leadley, Warwick University 1997.

The QHE looks finely tuned. This is a result of electrons interacting at the edge, where there is a confinement of edge states^[3]. The edge states dominate transport when the Landau levels of the bulk are filled. Because they are dissipationless, the resistivity ρ_{xx} of the system vanishes, before reaching the next Landau level, where scattering between bulk states can occur again. This is the hallmark³ of topological states, where there exists some states between different phases of matter.

The Hall resistivity ρ_{xy} exhibits quantised levels however.

$$\sigma_{xy} = \nu \frac{e^2}{h} \quad (2)$$

Thouless (who later received the Nobel prize in 2016, before passing away in 2019), Kohmoto, Nightingale, and den Nijs (TKNN)^[4] were interested in gapped bulk systems but with conductive edges and a periodic lattice potential. These systems had been argued for explaining the QHE by Laughlin^[5] and the integer effects had earlier been demonstrated theoretically by Hofstadter's Butterfly^[6]. TKNN recognised that **K** space maps to a non-trivial Hilbert space for the QHE; the space has a topology. This topology can be specified by the integer ν which also corresponds to the Hall conductance above in Eq. 2. In particular, their calculations only depended on details of the wavefunctions of the bandstructure.

What makes this fascinating is that the QHE is a clear example of macroscopic quantum phenomena. Other such phenomena include Bose Einstein Condensation, where many atoms condense into the same uniform quantum state, and superconductivity where the pairing of electrons (Cooper pairs) also produce a new phase of macroscopic quantum behaviour.

For the QHE to occur, the Landau quantisation opens gaps in the band structure of the material, and the chemical potential is situated within this gap. In a classical picture, the boundaries of the material cannot sustain these cyclotron orbits, because of the phase transition to an ordinary insulator, rather than the QHE insulator, and so edge channels open in a **bulk-boundary correspondence**.

Note that the QHE does not preserve **time reversal (TR) symmetry**, as charge carriers experience different forces due to the magnetic field when their direction is inverted. The magnetic field breaks TR symmetry. This is explicit in the two degrees of freedom in the system; for an out-of-plane magnetic field, charge is separated into two lanes, and those channels move a particular direction, ie forward above for electrons, backward below for holes. Reversing the direction of carriers yields the same channels but switched, forward above for holes, backward below for electrons.

³Although unrelated to the Hall effect, get the pun? Haha.

2.1.2 Berry Phase

In 1983, Barry Simon^[25] recognised that the TKNN expression as an integral over the curvature associated with the Berry phase on the Brillouin zone. This is fundamentally important to understanding that the QHE is a quantum phase effect.

The **Berry Connection** is the expectation value of a gradient operator of vector \vec{R} acting on a wavefunction over some path Γ with positions \vec{R} . Physically it is the mechanism from getting one geometric space point to another, i.e. the change of parameters.

$$\vec{\mathcal{A}}_n(\vec{R}) = i \langle \psi_n(\vec{R}) | \nabla_{\vec{R}} | \psi_n(\vec{R}) \rangle \quad (3)$$

This object has N parts, for each of the vector components. There's a different connection for every eigenstate, of every point in the space. It is a little more subtle than a vector - it transforms under gauge transformations. For an example wavefunction to include some additional phase factor (i.e. Charge transformation), the wavefunction transforms as $|\psi'_n(\vec{R})\rangle = e^{-i\beta(\vec{R})} |\psi_n(\vec{R})\rangle$. This results in a change for the Berry connection:

$$\vec{\mathcal{A}}'_n(\vec{R}) = \vec{\mathcal{A}}_n(\vec{R}) + \nabla_{\vec{R}} \beta(\vec{R}) \quad (4)$$

So the “connection” is that it transforms with the gradient of a function, like a vector potential.

Berry Curvature is the consequence or result of going from one set of starting parameters to the same set of parameters over some path (also called the Berry **connection**). The Schroedinger equation can be used to provide the Berry connection. It turns out that through the path evolution, the state can pick-up a phase factor, relative to the original starting state. This phase factor is also known as the Berry phase^[23], and has consequences for the quantum mechanical properties of the system.

The **Berry Phase** is the integral

$$\gamma_n(\Gamma) = \int_{\Gamma} \vec{\mathcal{A}}_n(\vec{R}) \cdot d\vec{R} \quad (5)$$

By changing the gauge of the Berry phase we can notice some properties that shift:

$$\gamma'_n(\Gamma) = \int_{\Gamma} \vec{\mathcal{A}}_n(\vec{R}) \cdot d\vec{R} + \int_{\Gamma} \nabla_{\vec{R}} \beta \cdot d\vec{R} \quad (6)$$

$$\implies \gamma'_n(\Gamma) = \gamma_n(\Gamma) + \beta(R_f) - \beta(R_i) \quad (7)$$

It is not gauge invariant, which makes it difficult to measure. However, if the path begins **AND** ends in the same configuration point (ie, a closed loop) then the value is Gauge invariant! This imposes a few restrictions on the system to achieve a result.

- Eigenstates have to be imaginary to achieve a non-zero Berry phase.
- If the path is 1D, then the integration cancels out to result in a zero Berry phase, again.

In 3D systems, the Berry phase over some path Γ enclosing a surface S , then Stokes theorem can specify the **Berry curvature** \vec{D}

$$\oint_{\Gamma} \vec{\mathcal{A}}_n \cdot d\vec{R} = \iint_S (\nabla \times \vec{\mathcal{A}}) \cdot d\vec{S} \quad (8)$$

$$\vec{D} = \nabla_{\vec{R}} \times \vec{\mathcal{A}}_n(\vec{R}) \quad (9)$$

The Berry phase is also useful for solving other phenomena such as the Aharonov-Bohm effect.

2.1.3 Quantum spin Hall effect

The spin  was observed in 2004 by Kato *et al.*^[7], where for particular semiconductors a spin density  was observed rather than a charge density, like in the regular hall effect. The origins for such an phenomenon are similar to that of the anomalous Hall effect in ferromagnets, sometimes being extrinsic  sometimes intrinsic. Intrinsically, the **Berry curvature** of the electronic valence-band Bloch wave functions result in spin-Hall effect.

The spin Hall insulator was proposed by Murakami *et al.*^[8], where the Berry phase is finite implying a finite spin Hall conductivity. While this idea did not “generate spin currents due to an absence of any electrons at the Fermi level”^[9], it allowed further exploration to find it quantised version, a quantum spin Hall (QSH) insulator^[10;11;12].

In a  1D system with spin, there are four degrees of freedom; Spin can move either direction, and can move forward and backward. This could be separated into two copies of the QH system - spin up moving forward with spin down moving backward at the top, and spin down moving forward with spin up moving backward on the bottom. This separation of states is referred to the quantum spin Hall effect (QSHE), however it requires a key ingredient to allow the separation of states to occur. In the QHE, it is the magnetic field that breaks time reversal symmetry. For QSH insulators, the essential ingredient is spin orbit coupling (SOC).

An important step made by Kane and Mele^[11] was finding a “topological invariant” to characterise the QSH insulator states using an index. This index is called the Z_2 index. Topological invariants have appeared before. In the QHE, the TKNN invariant μ , where the quantised conductance is proportional to ν . The consequence of the Z_2 index is it maps the parity of the number of times the 1D edge state crosses the Fermi level. An odd parity ensures the existence of an edge state and consequently the phase of a topological insulator. The result is significant in it shows how topological phases exist in band structures of insulators, and do not require external magnetic fields breaking TRS like in the QHE^[9].

Whilst Kane and Mele^[11] used their SOC model to investigate the bandgap opening of graphene, SOC is very difficult to experimentally detect in graphene due to the low coupling strength, compared to that of heavier species. Bernevig et al.^[10] (Zhang’s group) instead proposed a Z_2 model for the band structure of mercury telluride (HgTe). They predicted a CdTe/HgTe/CdTe quantum well would give rise to the QSH effect, when the HgTe layer reached a certain critical thickness. Experimental observation was confirmed soon thereafter by König^[13], who observed a quantised conductivity σ_{xx} of $2e^2/h$, due to two conducting edges. They also observed the thickness dependence.

2.1.4 Topological insulators

By this time theorists (Moore & Balents^[14], Fu, Kane and Mele^[15]) had already leapt forward and predicted 3D systems that would exhibit quantum spin hall effect^[16]. It was at this point that the term “topological insulator” was first referred to^[14]. Whilst these 3D systems cannot be called QSH insulators, they are an analogous 3D extension. The extension to 3D TIs results in no longer having one invariant to determine the topology of a system, but rather 4 separate invariants, for 16 classes of materials. Generally they could be separated into two groups - strong  and weak 3D TIs.

The first prediction for a 3D TI was by Fu and Kane^[16]. They predicted that the surface states of bismuth antimony ($\text{Bi}_{1-x}\text{Sb}_x$) could be observed by looking at angle resolved photo emission (ARPES, see 7.2.1). The signature for non-trivial topology was in observing surface states crossing the Fermi energy between two TR-invariant momenta^[16]. This was observed in the same system by Hsieh *et al.*^[17].

2.2 Relevant theory and phenomena

2.2.1 Topological field theory

In 2001 the QHE state was generalised to a 4D TR-invariant state by Zhang and Hu, and was generalised by to field theory by Bernevig *et al.*. It was also shown later by Zhang how the Z_2 topology could be described in this field theory, and be reduced to the 2D and 3D cases.

Practically this is useful for describing electromagnetic response of TIs and predicting magneto-electric effects, according to Ando's review^[9].

2.2.2 Dirac fermions

Within graphene, mentioned earlier for the Kane and Mele SOC model^[12], the 2D electron gas over the carbon honeycomb lattice creates interesting band structure features called "Dirac cones". The carriers occupying these band structure states behave according to a pseudo-relativistic Dirac equation, behaving differently to that of regular fermions in materials. Their physical behaviour mimics that of highly relativistic situations. 

Dirac fermion mass :

Sometimes the charge carriers in graphene are called "massless" Dirac fermions - they behave analogous to relativistic particles with zero rest mass conforming to the Dirac equation^[18]. This result comes from the observation of the fermions having a linear cyclotron mass with energy around the Dirac point's linear dispersion, effectively proving zero dependence on a "rest mass". Instead of their characteristic speed being the speed of light c , the Fermi velocity v_F describes their motion.

Generally in semiconductors and other insulators, the electronic bands either side of the bandgap become quite flat with curvature in momentum space. The important of such features is reflected in the effective mass of carriers as the Fermi energy locally fills states at the band edge. Historically, the effective mass of such carriers is calculated as^[19]

$$m^* = \left(\frac{\partial E^2}{\partial^2 k} \right)^{-1} \quad (10)$$

For parabolic, isotropic bands, a local approximation to the effective mass is:

$$E(\mathbf{k}) = E_0 + \frac{\hbar^2 \mathbf{k}^2}{2m^*} \quad (11)$$

(12)

which satisfies the above calculation. In graphene however, the dispersion equation near the Dirac cones (the K and K' points) are similar to the Dirac equation:

$$E(\mathbf{k}) \approx \hbar v_F \cdot \mathbf{k} \quad (13)$$

If we apply the same effective mass equation, we arrive at:

$$m^* = \left(\frac{\partial v_F \cdot \mathbf{k}}{\partial \mathbf{k}^2} \right)^{-1} \xrightarrow{\mathbf{k} \rightarrow \infty} \infty \quad (14)$$

If the carriers were really so heavy, conduction would be very restricted in Graphene. Instead, a different method of calculating the effective mass has to be used^[20] avoiding divergence. 

$$m^*(E, \mathbf{k}) = \frac{p}{v_g} = h^2 \mathbf{k} \left(\frac{\partial E}{\partial \mathbf{k}} \right)^{-1} \quad (15)$$

$$= \hbar k \frac{1}{v_F} = \hbar E \frac{1}{v_F^2} \quad (16)$$

This yields the mass proportional to the linear dispersion, just as $E = mc^2 \rightarrow m = E/c^2$, exhibiting relativistic behaviour.

Dirac particles in topological insulators :

As pointed out by Ando^[9], Spin-orbit interactions and 3D “massive” Dirac theory have been known for a long time in bismuth research, which has also been a playground for its diamagnetism (different to Pauli paramagnetism & diamagnetism). Wolff^[21] showed that a 2 band bismuth model can be transformed into a 4-part Dirac Hamiltonian.

It is interesting that both bismuth and another semi-metal antimony were used to produce the first 2D TI surface states in $\text{Bi}_{1-x}\text{Sb}_x$ where 2D surface “massless” Dirac systems exist. A distinguishing property of massless Dirac fermions is the Berry phase of π , discussed earlier, which provides an absence of backscattering^[22]. The TI surface states are consequently electronically protected from some scattering.

3 Topological insulator physics

3.1 Topological invariants

3.1.1 TKNN invariant

A detailed calculation for the TKNN invariant will be added in the future.

Simon first showed^[25] that the Berry phase is significantly related to the TKNN topological invariant.

3.1.2 Z_2 invariant

A detailed calculation for this invariant will be added in the future.

3.1.3 Z_2 invariant in 3D

A detailed calculation for this invariant will be added in the future.

- (1;000) - Dirac cone is centred on the $\bar{\Gamma}$ point.

3.2 Spin-orbit coupling

Spin-orbit coupling (SOC) is coupling of both spin and orbital angular momentum properties of particles. Spin provides a magnetic moment, and the motion of the charged electron through its orbital angular momentum also provides a magnetic moment. From the electrons point of view, it can be considered to be the interaction of the electron’s spin with the orbital motion^{of the nucleus, or through a crystal with a periodic potential.}

Llewellyn Thomas derived the spin-orbit energy splitting for the hydrogen atom in 1926^[26;27]. You can derive the SOC from the Dirac equation for a spin $\frac{1}{2}$ particle in the presence of an electromagnetic field (ie, proton for hydrogen atom).

$$(E - mc^2) \psi = \left(\frac{p^2}{2m} - \frac{p^4}{8m^3c} + V - \frac{\hbar}{4m^2c^2} \frac{\partial V}{\partial r} \frac{\partial}{\partial r} + \frac{1}{2m^2c} \frac{1}{r} \frac{\partial V}{\partial r} \mathbf{S} \cdot \mathbf{L} \right) \psi \quad (17)$$

The first and third terms are the same as the non-relativistic Schrodinger equation, the second term is the relativistic correction, and the last term is the spin-orbit coupling.



You may also see the SOC represented by Hamiltonian terms such as

$$H_{SO} \propto \boldsymbol{\sigma} \cdot (\vec{\mathbf{E}} \times \vec{\mathbf{p}}) \quad (18)$$

where $\boldsymbol{\sigma}$ are the Pauli matrices, $\vec{\mathbf{E}}, \vec{\mathbf{p}}$ are the external electric field and momentum operator respectively. You can think of the momentum of the electron moving through the electric field as inducing a magnetic field interaction with the spin.

This coupling causes splitting, similar to Zeeman splitting, of electronic energy levels. For different atoms, the SOC is different, with the trend that heavier atoms have a larger SOC^[28].

Within materials, SOC doesn't break time-reversal symmetry like magnetic field does for the QHE, but it can lead to the QSHE, where electrons differentiated by their spin move in opposite directions. Physical effects of SOC in most solid state materials typically result in the Rashba effect or the Dresselhaus effect^[29]

3.2.1 Dresselhaus effect

The SOC for Bloch electrons (crystalline solid state) was first noted by Elliot and Dresselhaus^[30]. Dresselhaus in particular demonstrated the SOC splitting of atomic orbitals could alter cyclotron resonance spectra in semiconductors, and went onto study type III-V and II-VI semiconductor structures in the early 1950's. The Dresselhaus effect occurs in crystals with bulk inversion asymmetry.

Inversion symmetry is when the crystal is symmetric under inversion, from some lattice point from the origin). The breaking of inversion symmetry (inversion asymmetry) is usually important in allowing non-linear effects to take place in materials, otherwise even harmonics of wave functions become suppressed. This includes materials that allow high harmonic generation, such as in ultra-fast femtosecond pulses^[31].

Practically, the fact that net electric fields exist within crystals for particular crystal directions gives rise to spin-orbit interactions.

A typical Dresselhaus Hamiltonian contribution would look like:

$$\mathcal{H}_D \propto p_x(p_y^2 - p_z^2)\sigma_x + p_y(p_z^2 - p_x^2)\sigma_y + p_z(p_x^2 - p_y^2)\sigma_z \quad (19)$$

3.2.2 Rashba effect

Rashba studied a different type of crystal to Dresselhaus, known as a "wurtzite structure" in the late 1950's. His paper with Sheka demonstrated linear dispersion relationships near the Γ point. This paper (hard to come by online due to lack of translation and availability in the Soviet Union) is translated to English and included in Bihlmayer *et al.*^[30]. The Rashba effect occurs in crystals also with net electric field; this one is due to structural inversion asymmetry, rather than bulk asymmetry. In particular systems such as those with uniaxial symmetry (in one axis only) such as the hexagonal crystals of cadmium-sulphur and cadmium-selenide where it was originally found by Rashba and Sheka.

The simple Rashba Hamiltonian contribution is

$$H_R = \alpha (\boldsymbol{\sigma} \times \mathbf{p}) \cdot \hat{z} \quad (20)$$

3.3 Hamiltonians of TI Materials

The following models from Qi & Zhang^[32] are formed for QSHE by a Hamiltonian that is a Taylor expansion in the wavevector \mathbf{k} of interactions between highest and lowest conduction bands.

3.3.1 HeTe (Mercury Telluride)

$$H(\mathbf{k}) = \epsilon(k) \mathbb{1} + \begin{pmatrix} M(k) & A(k_x + ik_y) & 0 & 0 \\ A(k_x + ik_y) & -M(k) & 0 & 0 \\ 0 & 0 & M(k) & -A(k_x + ik_y) \\ 0 & 0 & -A(k_x + ik_y) & -M(k) \end{pmatrix} \quad (21)$$

with

$$\epsilon(k) = C + Dk^2, M(k) = M - Bk^2 \quad (22)$$

Here the upper 2x2 block describes the spin-up electrons in the s-like E1 conduction and p-like H1 valence bands. The lower 2x2 block describes the spin-down electrons in those same bands.

ϵ is the unimportant bending of all bands, where $\mathbb{1}$ is the identity matrix. The energy gap between the bands is $2M$, and B describes the curvature of the bands. A incorporates the inter-band coupling at lowest order. $M/B < 0$ has eigenstates of a trivial insulator. $M/B > 0, M$ becomes negative, and solution yields edge states of QSHE. You can also practise doing this with a 2D TI honeycomb lattice to gain explicit understanding like Kane and Mele^[12;11].

3.3.2 Bi₂Te₃ (Bismuth Telluride)

$$H(\mathbf{k}) = \epsilon(k) \mathbb{1} + \begin{pmatrix} M(\mathbf{k}) & A(k_x + ik_y) & 0 & A_1 k_z \\ A(k_x + ik_y) & -M(\mathbf{k}) & A_1 k_z & 0 \\ 0 & A_1 k_z & M(\mathbf{k}) & -A(k_x + ik_y) \\ A_1 k_z & 0 & -A(k_x + ik_y) & -M(\mathbf{k}) \end{pmatrix} \quad (23)$$

with

$$\epsilon(k) = C + D_1 k_z^2 + D_2 k_{\perp}^2, M(k) = M - B_1 k_z^2 - B_2 k_{\perp}^2 \quad (24)$$

Bi₂Te₃ follows a similar model, in the context of bonding and anti-bonding p_z orbitals with both spins. B_1 and B_2 have the same sign, and as before, depending on the sign of M, the bands undergo inversion.

It is essential to be in 3D to be able to construct a single Dirac cone for a 2D surface - the degeneracy of graphene with two Dirac cones makes it clear. The 2D HgTe quantum well at the crossover point d , also has two Dirac cones.

4 Topological insulator materials

4.1 CdTe/HgTe/CdTe quantum wells

The {Hg,Cd}Te quantum well (QW) was the first successful experiment of a QSH insulator (a 2D TI), with thin layer of HgTe sandwiched between two layers of CdTe (Fig. 3). This system becomes 2D due to quantum confinement from quenching of the out-of-plane direction. Bulk HgTe exhibits a band inversion necessary to observe a QSH effect, whereas CdTe does not. However, HgTe is also a zero-gap semiconductor due to a symmetry protected Γ point. By sandwiching HgTe in CdTe, the slightly larger lattice parameter of CdTe applies a strain on the crystal structure of the thin HgTe when grown by MBE, such that the cubic lattice symmetry is broken and leads to a bandgap opening, becoming a genuine insulator. BHZ predicted that above some critical thickness, the band inversion would be retained and become a TI^[10]

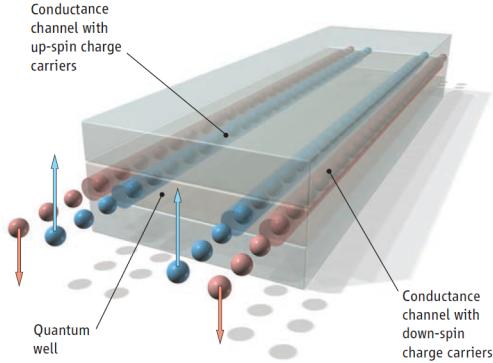


Figure 3: Schematic of CdTe/HgTe QW structures. Devices were patterned into hall bar geometries.

Source: König *et al.*^[13], Science 318

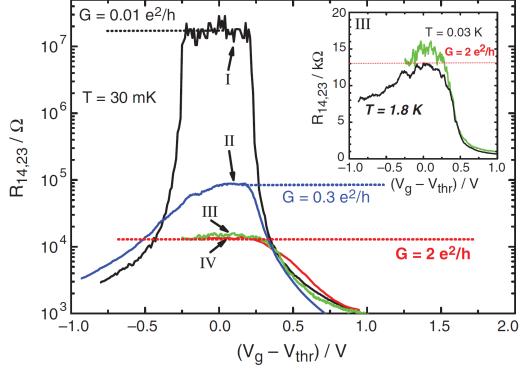


Figure 4: The longitudinal four-probe resistance of various CdTe/HgTe QW structures. HgTe thickness in I is 5.5nm, II,III and IV are 7.3nm. All devices measured at $B=0\text{T}$, $T=30\text{mK}$.

Source: König *et al.*^[13], Science 318

These characteristics were confirmed later by König *et al.*^[13] from the Würzburg group, as shown in Fig. 4. The detected thickness at which the band inversion occurred was at 6.3nm (almost exactly at the prediction of 6.4nm^[10]), where the observation of a quantised conductance became present at low temperatures, previously having diverging resistance. This material only had a bandgap of 10meV resulting from the strain, and so could only be properly measured at sufficiently low temperatures. A quick back of the envelope calculation using $k_B T = \Delta E$ gives a temperature of 116.04 K.

Q: Why was there a lack of band-inversion prior to 6.3nm? As the crystal gets thicker, the less the epitaxial stretching affects the lattice parameters and consequently allows room for band inversion, whilst maintaining a bandgap? If it gets too thick, do I just return to HgTe behaviour which has a closed gap?

4.2 $\text{Bi}_{1-x}\text{Sb}_x$

Bismuth Antimony ($\text{Bi}_{1-x}\text{Sb}_x$) is the first 3D topological insulator discovered^[17], after its prediction well grounded prediction by Fu and Kane^[16]. ARPES was used to confirm the material, as shown in Fig. 6. The theoretical concentrations for which the alloy $\text{Bi}_{1-x}\text{Sb}_x$ becomes a TI are shown in Fig. 5.

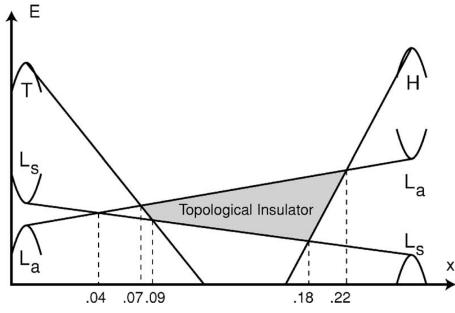


Figure 5: The band energy evolution of $\text{Bi}_{1-x}\text{Sb}_x$ as function of the antimony percentage x .

Source: Fu and Kane^[16], Phys. Rev. B 76, 045302

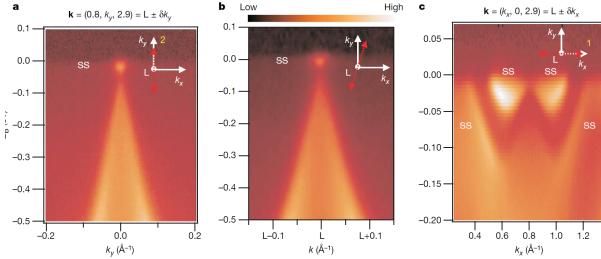


Figure 6: Dirac-like dispersions near the L-point in the bulk BZ in $\text{Bi}_{0.9}\text{Sb}_{0.1}$. **a**) Cut along k_y , **b**) Cut 10° from k_y and **c**) the k_x directions.

Source: Hsieh *et al.*^[17], Nature Vol. 452, 970 (2008)

It is worth noting that $\text{Bi}_{1-x}\text{Sb}_x$ is topologically classed by the Z_2 3D invariant as (1;111). $\text{Bi}_{1-x}\text{Sb}_x$ boasts a high mobility of $\sim 10^4 \text{ cm}^2/\text{Vs}$, whilst the bulk carrier density can be around $\sim 10^{17} \text{ cm}^{-3}$ ^[33]. This sort of mobility makes it easy to study 2D quantum transport according to Ando^[9].

High mobility devices useful for studying because?

Unfortunately, the material's surface states also inherit the strong Rashba-split surface states that are not topological coming from Bismuth; there are 2-4 Fermi-level crossings owing to the Rashba effect, whilst only one topological band (depending on chemical potential), making it complicated to clearly analyse the topological physics present, as discussed in Sec. 7.2.1.

4.3 $\{\text{Bi},\text{Sb}\}_2\{\text{Se},\text{Te}\}_3$ family

Bi_2Te_3 was originally suggested as a candidate without band calculations by Fu and Kane^[16]. However, Zhang *et al.*^[34] filled in the calculations gap, and additionally predicted a whole family of related structures, generalised by the form $A_2B_2B'_1$

These materials form in a rhombohedral crystal structure, containing 26 A atoms, 26 B atoms, and 20 B' atoms, seen in Fig 7a). However, they're named after their stacked structure of **quintuple layers**, where there are 10 A atoms, 10 B atoms and 5 B' atoms (on average), making a ratio of (2,2,1), or if $B \equiv B'$, (2,3). In the latter case, they are also known as tetradymites.

This class of materials covalently bond within their quintuple layers, but only weakly interact through van der Waals forces, which allows the cleaving between such layers.

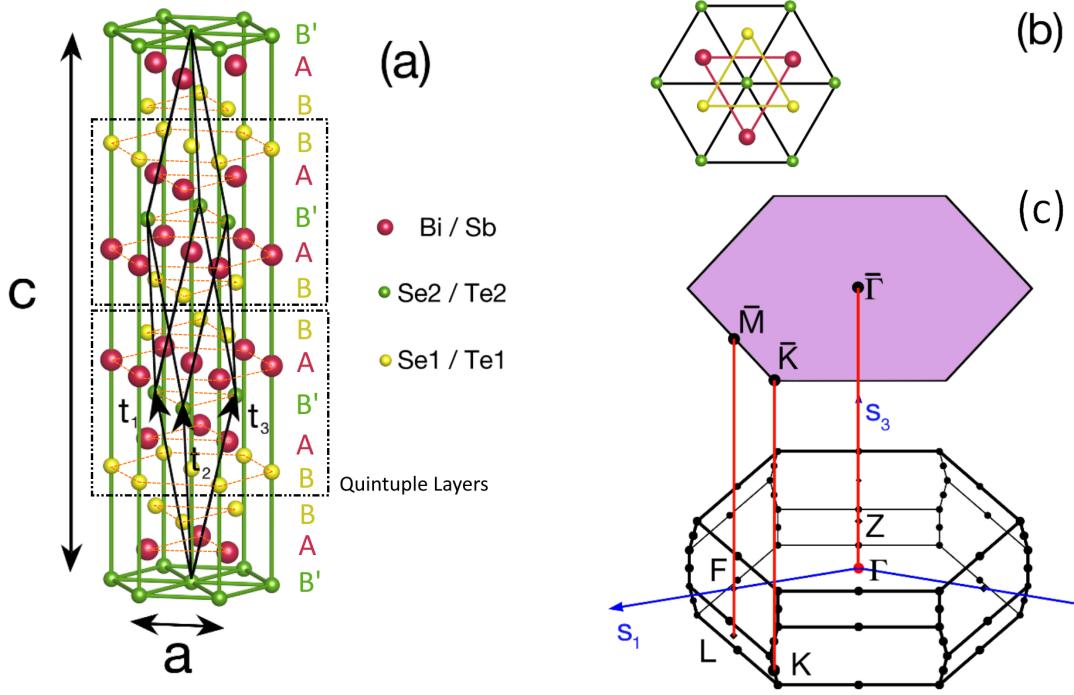


Figure 7: $A_2B_2B'_1$ family. **a)** Rhombohedral unit cell ($c \sim 3$ nm). **b)** Z-axis view of unit cell. **c)** Bulk Brillouin zone projected along the [111] direction (purple shaded area).
Adapted from: Aramberri and Muñoz^[35], Phys. Rev. B 95, 205422 (2017)

The tetradymite members of this family include Bi_2Te_3 , Bi_2Se_3 , and Sb_2Te_3 . Interestingly Zhang's calculations determined Sb_2Se_3 to not be a topological insulator.

Other materials that are tetradymite chalcogenides include Bi_2Te_2Se .

4.3.1 Bi_2Te_3

Bismuth telluride has a much smaller bandgap (0.18eV) than that of Bi_2Se_3 (0.35eV), and consequently is difficult to experimentally isolate the topological surface state^[36].

In a landmark study, Hsieh *et al.* observed the first significant evidence of helical Dirac fermions. They used SR-ARPES (see Sec. 7.2.1), with a double Mott detector to systematically measure all three components of the spin as a function of energy and momentum. Because of the Bi_2Se_3 degenerate bulk conduction band close to the Γ point, there is an extra contribution to the density of states, making it clearer to observe momentum-spin locking (the helical nature) in Bi_2Te_3 . These results are shown in Fig. 8.

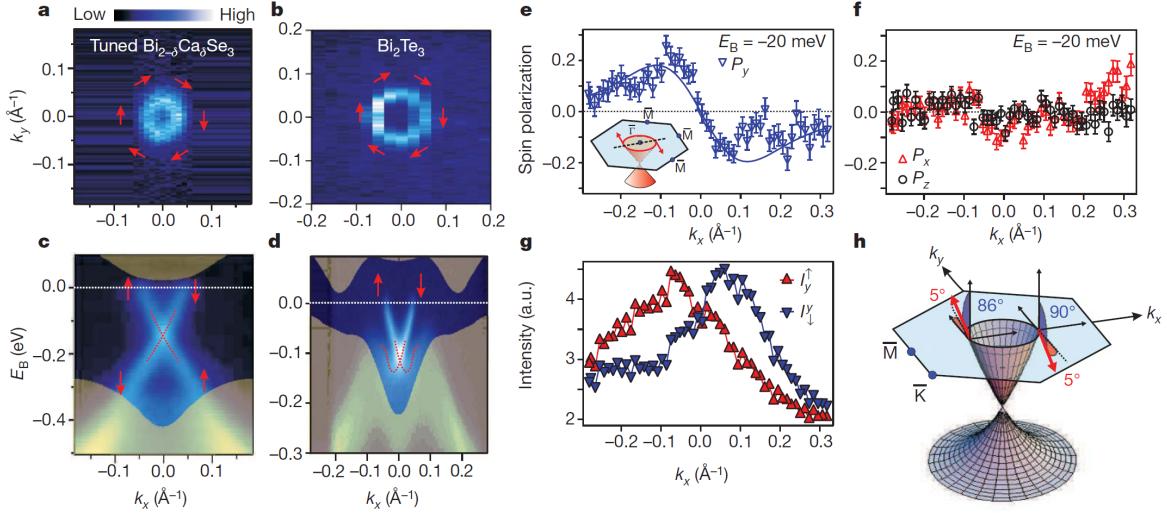


Figure 8: **a,b)** ARPES intensity map of E_F on (111) surface of **a** $\text{Bi}_{2-\delta}\text{Ca}_\delta\text{Se}_3$ and **b** Bi_2Te_3 . **c,d)** Respective ARPES dispersions along the k_x cut. Yellow shaded regions are projections of bulk bands of pure Bi_2Se_3 and Bi_2Te_3 . **e,f)** y component and $\{x,z\}$ components (triangles, circles) of spin polarisation along the $\Gamma - M$ direction, respectively in Bi_2Te_3 . **g)** Spin resolved spectra - counts of different spins at different k_x . Errors attributed to time evolution of the surface band dispersion in Bi_2Te_3 . **h)** Fitted values of spin polarisation vector \mathbf{P} (S_x, S_y, S_z). Angles are shown, contributing to unit vectors $P_{k_x} = (-0.087, -0.996, 0)$ and $P_{-k_x} = (0.087, 0.994, 0.070)$. Errors are of order ~ 10 deg for angles, or ± 0.15 for magnitude.

Source: Hsieh *et al.* Nature 1101, 460 (2009)

4.3.2 Bi_2Se_3

The first observation of the Dirac cone in bismuth selenide (Bi_2Se_3) was reported by Xia *et al.*^[37] Unlike many other topological materials, Bi_2Se_3 is close to the Dirac cone undoped, as shown in Fig. 9a. Normally Bi_2Se_3 is n-type doped due to vacancies or excess selenium^[37] that accumulate through sample growth.

The demonstration of helical Dirac fermions by Hsieh *et al.* mentioned earlier also demonstrated the doped tuning of Bi_2Se_3 to a (relatively) insulating state by using a stoichiometric $\text{Bi}_{2-\delta}\text{Ca}_\delta\text{Se}_3$ ^[36], where Ca acts as a hole donor. Remarkably they are able to demonstrate ARPES data clearly retaining the topological Dirac point at room temperature.

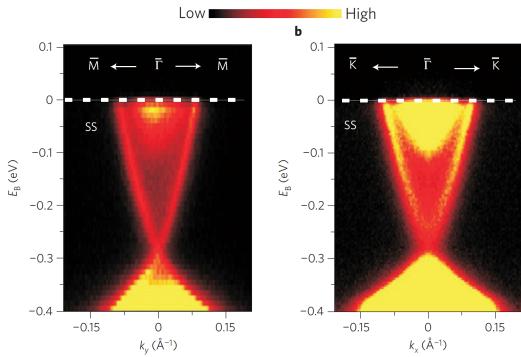
The first significant transport experiment in Bi_2Se_3 by Kim *et al.*^[38] demonstrated a predicted^[39] zero hall coefficient (diverging hall carrier density) at the Dirac point - rather than a zero hall carrier density as one might expect when changing carrier type. This is shown in Fig. 9b. A zero Hall coefficient gives rise to either zero electric field generated or a diverging current density, which is very similar to the behaviour in graphene.

$$R_H = \frac{E_y}{j_x B_z} \quad (25)$$

Kim measured the mobilities of Bi_2Se_3 to be 320-1,500 cm^2/Vs , with the exfoliated flakes possessing n-doped charge densities of $\gtrsim 10^{13} \text{ cm}^{-2}$. This is much greater than the bulk charge density of $\approx 10^{17} \text{ cm}^{-3}$.

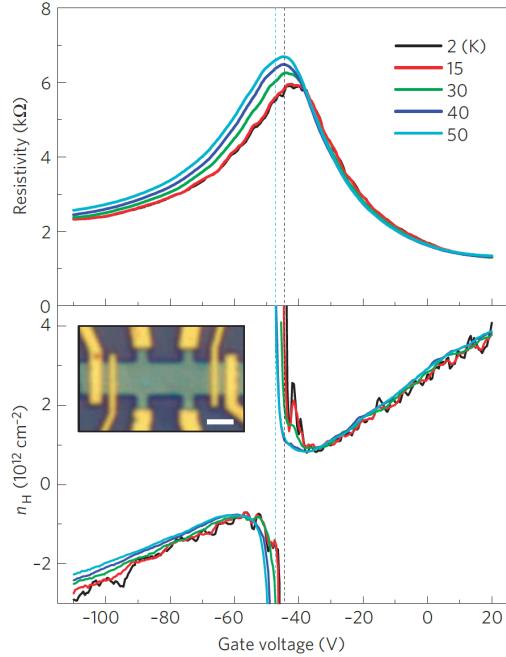
Bi_2Se_3 is classed by the 3D Z_2 invariant as (1;000). Bi_2Se_3 has a bulk bandgap of ~ 0.35 eV. This is a sizeable gap, however unfortunately, even at low temperature (where thermal effects are diminished), the bulk conductivity is quite high due to the small bandgap.

(a) ARPES of Bi_2Se_3 . **a)** From $\Gamma \rightarrow M$ **b)** From $\Gamma \rightarrow K$
 Source: Xia *et al.* Nature Physics 398, Vol 5 (2009)



(b) Bi_2Se_3 longitudinal resistivity ρ_{xx} (top) and sheet carrier density $n = 1/(eR_H)$ (bottom) where R_H is the Hall coefficient and e is the elementary charge.

Source: Kim *et al.* Nature Physics 459, Vol 8 (2012)



It has also been reported that some of these materials (Bi_2Se_3 in particular) undergo surface oxidisation when exposed to ambient (light & air) conditions, degrading their crystal quality and topological properties. The results of Kong *et al.* are shown in Fig. 10^[40]. This has also been reported with other similar materials, such as in the transition metal dichalcogenides (TMDs)^[41].

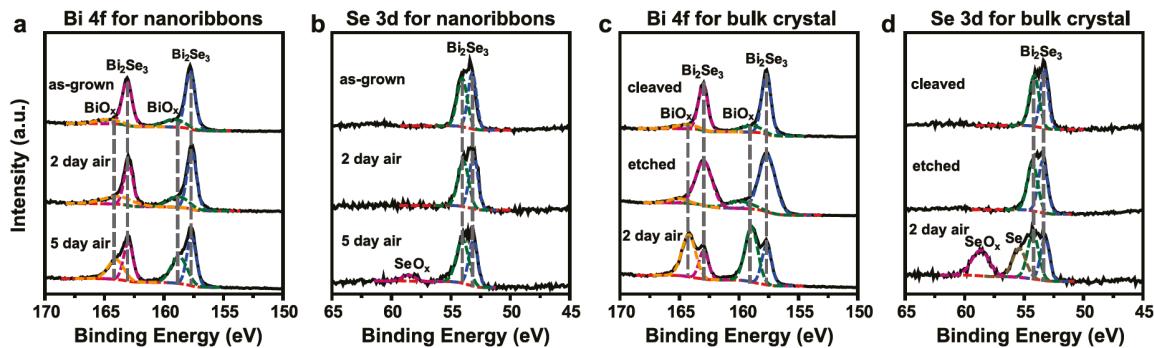


Figure 10: XPS studies on Bi_2Se_3 showing oxidation after exposure to air.
 Source: Kong *et al.*, ACS Nano Vol. 5, No. 6, 4698 (2011)

5 Ferromagnetic Insulators

In this section, I will discuss various ferromagnetic insulator (FI) materials that might be used to ~~speak~~ with Bi_2Se_3 . Properties that will matter ~~will~~ include the cleanliness of the interface, the lattice parameters, any resulting strain that might occur, and magnetic properties such as magnetisation amplitude. I will cover new few-layer 2D ferromagnetic insulators that have potential to be used for spin-torque experiments.

FIs are rare, because most magnetic materials that incorporate iron, cobalt or nickel are often conductive. That being said, some 3D FIs have been known for a while, and recently some new FIs that can be exfoliated down to thin flakes have been discovered.

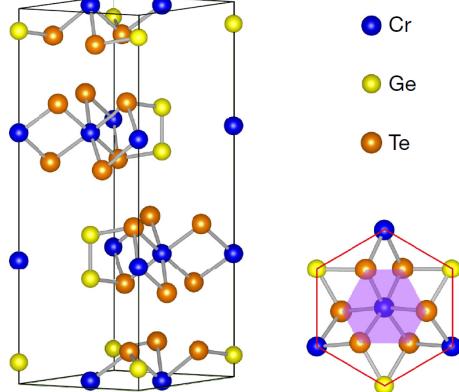


Figure 11: Crystal structure of $\text{Cr}_2\text{Ge}_2\text{Te}_6$.
Source: Gong *et al.*, Nature Vol. 546, 265 (2017)

5.1 2D FIs

5.1.1 Chromium triiodide (CrI_3)

CrI_3 was first reported on around 2014^[42]. The crystal undergoes a phase change below 220 K from monoclinic to rhombohedral. It has a Curie temperature at 61K in the bulk, and has a resistivity of 900 Ωcm .

Recent reports have demonstrated CrI_3 's ferromagnetic properties persist down to few-layers^[43;44]. Huang *et al.* reports a curie temperature of 45K for monolayer CrI_3 , data shown in Fig. 12. The followup paper shows electrostatic gate control of CrI_3 between ferromagnetic and anti-ferromagnetic states with some spin-locking^[45].

5.1.2 $\text{Cr}_2\text{Ge}_2\text{Te}_6$ (CGT)

$\text{Cr}_2\text{Ge}_2\text{Te}_6$ is also a new thin ferromagnetic insulator. It consists of quintuple layers, similar to Bi_2Te_3 or Bi_2Se_3 , and is also terminated with hexagonal Te planes (Fig. 11), making it a van der Waals material.

Crystals have been synthesised in bulk before through a flux method, such as demonstrated by Zhang *et al.*^[46], with clear insulating and magnetic properties.

The discovery paper of 2017^[47] shows that bilayers are stable for exfoliation, and magnetic order is found using a scanning Kerr microscope. It was noted that monolayers were thought to be invisible. Whilst the bulk material has a Curie temperature of 61K, the 2D material is less stable, being well defined below 40K. This is very similar to CrI_3 . While this is quite novel for investigating low dimensional physics and magnetic-spin effects, this material lacks the high temperature range to be practical around ambient temperatures.

More recent transport data was published by using Pt electrodes, showing the conductivity that appears above 60 K, compared to the massive resistance below^[48]. There are also promising predictions to increase the Curie Temperature of CGT towards liquid nitrogen temperatures, through the use of absorption of gas molecules (NO , NO_2)^[49].

The perpendicular magnetic anisotropy (PMA) is a measure of how strong the magnetic interactions are in the perpendicular direction. This is particularly important for memory or torque devices. For CGT, a PMA energy of 10^5 erg/cm³ is reported^[50].

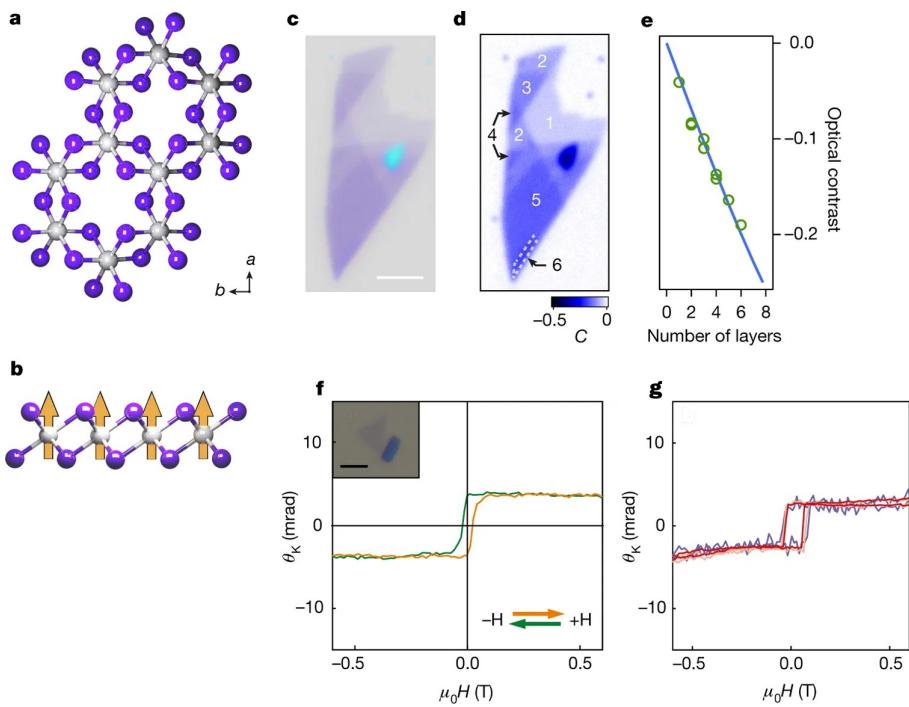


Figure 12: **a)** CrI_3 monolayer atomic lattice. **b)** Out-of-plane view, depicting Ising spin. **c)** Optical image, scalebar $3\mu\text{m}$. **d)** Calculated contrast map. **e)** Linearity of contrast to thickness. **f)** Kerr signal of monolayer. **g)** Power dependence of Kerr signal taken at powers of 3 μW (blue), 10 μW (pink), 30 μW (red)

Adapted from: Huang *et al.*, Nature Vol. 546, 270 (2017)

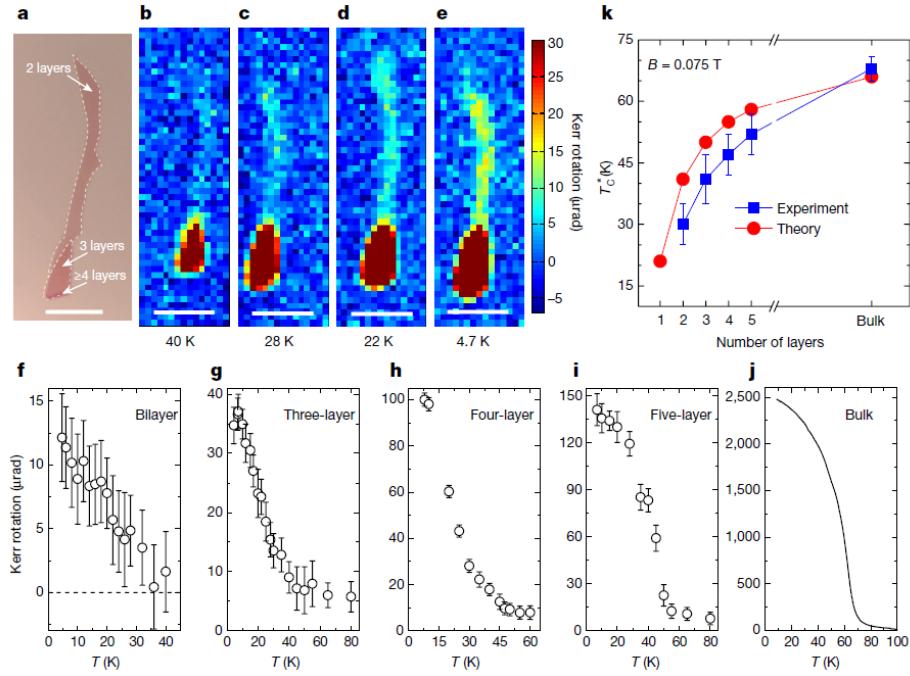


Figure 13: **a)** Microscopy of $\text{Cr}_2\text{Ge}_2\text{Te}_6$. **b-e)** Appearance of Kerr rotation signal **f-j)** Temperature dependence of Kerr signal in different thicknesses. **k)** Calculations of transition temperatures.

Source: Gong *et al.*, Nature Vol. 546, 265 (2017)

5.1.3 Magnesium Titinate (MgTiO_3 , MTO)

MTO is another recent addition to thin film ferromagnetic insulator films. Frantti *et al.* reported the stoichiometric material in 2019^[51].

MTO has a rhombohedral ilmenite structure, and in this study was grown via a pulsed laser deposition technique to deposit films of thicknesses around 50-60 nm. Across a variety of samples, Frantti saw magnetization swings of up to $\Delta M = 80 \text{ emu cm}^{-3} = 8 \times 10^4 \text{ Am}^{-1}$ (see Fig. 14). A few resistivity measurements of samples were reported, which were 0.07 and 265 $\text{M}\Omega \text{ cm}$, the latter reported to be very typical of most samples.

5.1.4 Other 2D ferromagnetic materials

While an insulator is preferable due to the clear picture expected when interfacing with topological insulator surface states, it's worth noting other 2D ferromagnetic developments. As of 2018, Fe_3GeTe_2 has also been recognised as a 2D van der Waals material possessing ferromagnetism to a much higher Curie temperature ($\sim 230 \text{ K}$)^[52]. This material also has a much larger PMA than CGT, boasting $\sim 10^7 \text{ erg/cm}^3$.

6 TI & FI heterostructures

There has been much interest in creating spintronic devices for modern applications. One focus in particular is a heterostructure (two or more layers) that creates a spin-torque device. That is, some ferromagnetic layer that is switchable by running an electric current through another layer. Researchers have been trying to make these devices a reality using a heterostructure of a topological

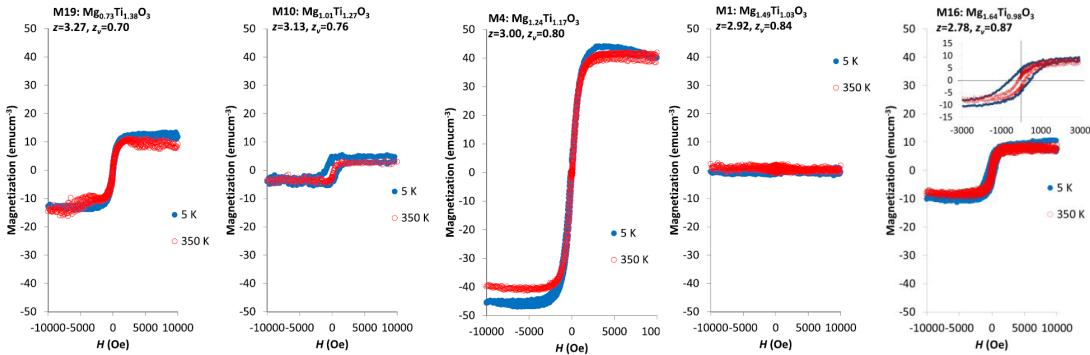


Figure 14: Magnetisation measurements for various mixtures of Mg and Ti in MGO, for two different temperatures. Average valence z and octahedral filling fractions z_v are shown.

Source: Frantti *et al.*, J. Phys. Chem. C 123, 19970 (2019)

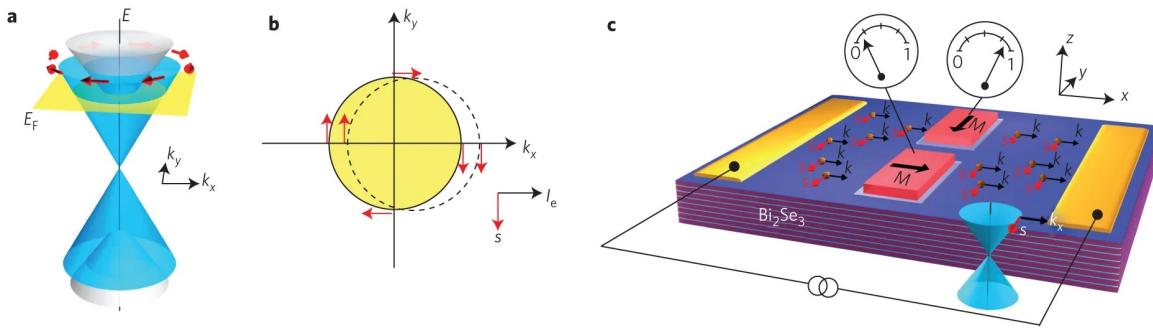


Figure 15: **a)** Momentum-spin locking - a Dirac cone with spin pointing at right angles to momentum. **b)** Top view of a slice of k_x - k_y plane. An applied voltage produces a net momentum, and a net spin-current. **c)** The transport system, where spin polarised currents drive transitions in local ferromagnets.

Source: Li *et al.*^[56], Nature Nano. 9, 218 (2014)

insulator and ferromagnetic materials for about 10 years, since the first suggestions^[53;54] of such applications of TIs.

Here I will outline the basic mechanics (what makes the wheels spin) of a spin-torque device, as well as the measurements and figures of merit by which to judge their success. I will show some significant results for conductive and insulating ferromagnetic materials, and identify gaps in experiments.

6.1 Spin Torque Devices

There have been some theoretical predictions for spin-torque devices for a while, considering metallic ferromagnet - topological insulator heterostructures as spin torque devices^[54;55].

The basic mechanism is that a current that runs through the surface state of a topological insulator is momentum-spin locked - this means the a net spin polarisation for carriers moving in a particular direction. The idea is that this spin density is strong enough to influence the ferromagnetic material that is within proximity. This scenario is shown in Fig. 15, from Li *et al.*, in which small metallic ferromagnetic pads produced μV responses to mA currents^[56].

In addition to the above effects, the proximity effect of the ferromagnetic material's field to

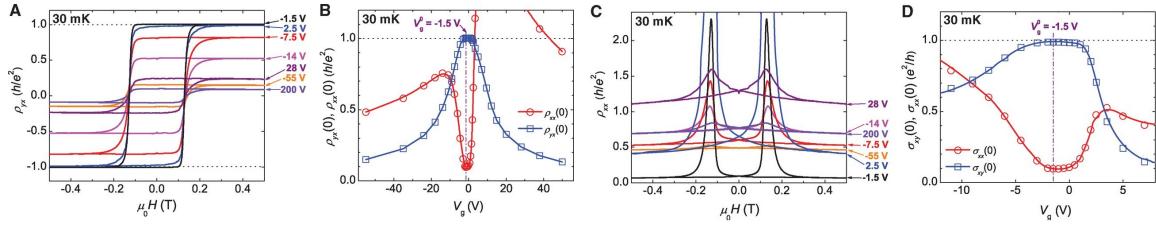


Figure 16: **a)** Magnetic field dependence of ρ_{xy} for different V_g **b)** Dependence of ρ_{xy} (blue) and ρ_{xx} (red) on V_g **c)** Magnetic field dependence of ρ_{xx} for different V_g **d)** Dependence of σ_{xy} (blue) and σ_{xx} (red) on V_g

Source: Chang *et al.*^[57], Science Vol. 340, 167 (2013)

topological insulator surface state can be used to break the time-reversal symmetry that protects the surface state, possibly acting as a transistor switch.

6.1.1 Torque

Total angular moment L.

$$\frac{d\vec{L}}{dt} = \vec{T} \quad (26)$$

A spin orbit torque (SOT) is described as

$$\tau_{SO} = -\gamma \mathbf{M} \times \mathbf{B}_{SO} \quad (27)$$

where γ is gyromagnetic ratio for some effective B_{SO} .

6.2 Quantum Anomalous Hall Effect (QAHE)

The anomalous hall effect been observed in ferromagnetic materials for a long time. It's anomalous because it appears without the need of an external magnetic field. The origins for the effect can be both intrinsic, or extrinsic (system dependent disorder)

It turns out that just like the Hall effect and the spin Hall effect have quantised versions, there also exists a quantum anomalous Hall effect (QAHE). The first observation of this was made by Chang *et al.*^[57] in the material $\text{Cr}_{0.15}(\text{Bi}_{0.1}\text{Sb}_{0.9})_{1.85}\text{Te}_3$, shown in Fig. 16. The signature of the QAHE is that the zero-field Hall resistance still exhibits a distinct plateau with the quantized value h/e^2 around $V_g = -1.5$ in sub-figure B. The particular sample used has a Curie temperature of ~ 15 K, and a mobility of $760 \text{ cm}^2/\text{Vs}$.

6.3 Experiments

6.3.1 Conductive ferromagnetic films

Mellnik demonstrated the use of Bi_2Se_3 in conjunction with the ferroic permalloy $\text{Ni}_{81}\text{Fe}_{19}$. They also reported

- $\sigma_{S,\parallel} = 2.0 \pm 0.4 \times 10^5 \hbar/2e \Omega^{-1}\text{m}^{-1}$

- $\sigma_{S,\perp} = 1.6 \pm 0.2$

More recently, Wang *et al.* reported a spin-conversion efficiency of 1-1.75 in thin Bi_2Se_3 / NiFe heterostructures, with a switching current of $6 \times 10^5 \text{ A cm}^{-2}$ ^[59]. This is roughly an order of magnitude smaller than with heavy metals.

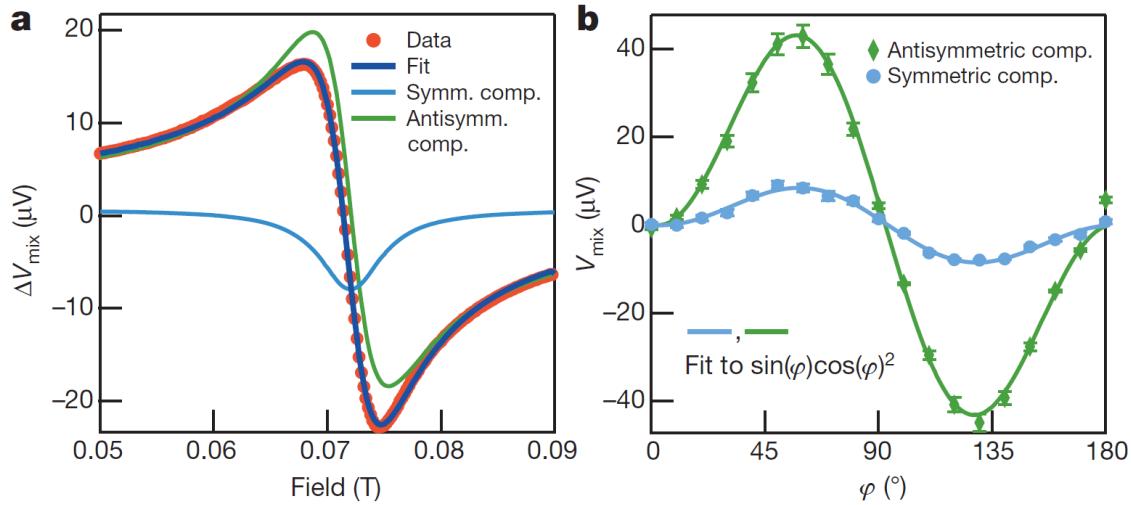


Figure 17: **a)** Spin Torque Ferromagnetic Resonance of current induced torque, with fits. **b)** Dependence on magnetif field angle ϕ .

Source: Mellnik *et al.*^[58], Nature Vol. 511, 449 (2014)

6.3.2 Insulating ferromagnetic films

6.3.3 Not-quite TI-Ferromagnetic heterostructures

In 2014, Fan *et al.*^[60] reported the observation of magnetised switching in a chromium-doped $(\text{Bi}_{0.5}\text{Sb}_{0.5})_2\text{Te}_3$ bilayer heterostructure at a critical current density of $8.9 \times 10^4 \text{ A cm}^{-2}$ at 1.9 K. However, this system is not only 3D spin-hall effects, but also Rashba interfacial interactions, and so the torque is not directly understood in terms of the topological surface state. The switching of the AHE resistance is shown in Fig. 18.

7 Experimental methods

7.1 Synthesis, Growth and Fabrication

TODO: fill in with summary of previous growth methods and experimental methods used for heterostructures.

7.1.1 MBE Growths

7.1.2 Doping

Kim *et al.*^[38] managed to heavily p-dope a sample of Bi_2Se_3 through the use of F4TCNQ molecules, as well as the use of an electrochemical doping with a polymer electrolyte top gate, in order to p-type dope the sample (see Fig. 19). The latter method works by a capacitance between sample surface to induce negatively charged ions. This was necessary to be able to gate the sample near the Dirac point.

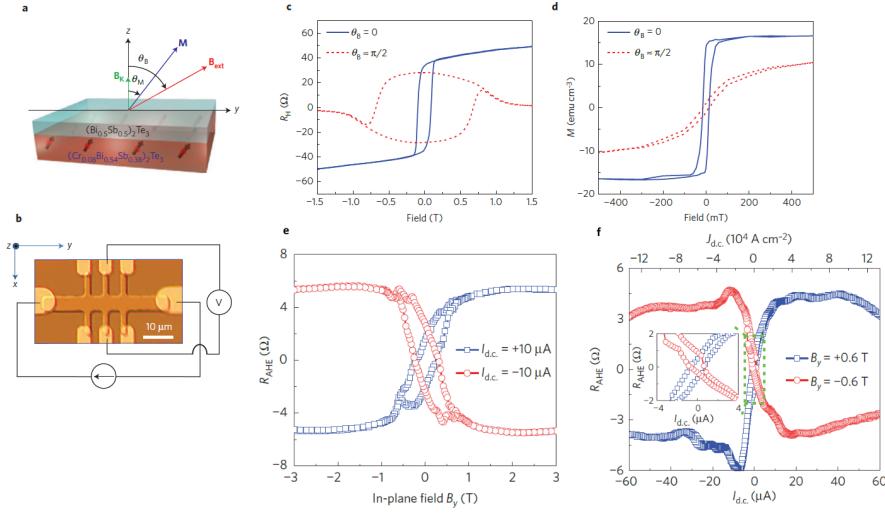


Figure 18: **a)** Bilayer heterostructure. Top (blue) 3 quintuple layers, bottom (red) 6 quintuple layers. **b)** Microscopy **c,d)** Hall resistance and magnetisation of sample. **e,f)** Current induced and field induced AH resistance.

Source: Fan *et al.*^[60], Nature Mat. Vol. 13, 699 (2014)

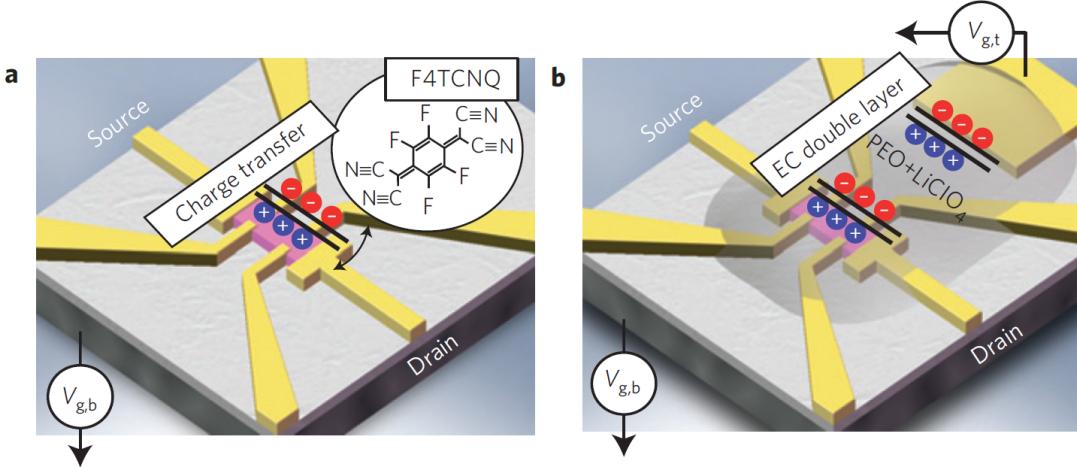


Figure 19: **a)** P-type doping and gate configuration for charge transfer doping with F4TCNQ molecules **b)** and polymer electrolyte (PEO + LiClO₄) top gating.

Source: Kim *et al.*^[38], Nature Physics 459, 8 (2012)

Alternatively, Hsieh *et al.*^[36] managed to p-type dope Bi₂Se₃ through the use of an extrinsic molecule nitrogen dioxide (NO₂) in UHV.

7.2 Spectroscopic Metrological Methods

7.2.1 ARPES

For material characterisation, angle resolved photo-emission spectroscopy (ARPES) is a method of resolving the ejection of electrons from a material by high energy photons. Analysis of the momentum

of the incident and emission resolve the electronic band structure.

There now exist a variety of different ARPES methods that can be employed in understanding the electronic structure of materials; these include “soft-X-ray ARPES, time-resolved ARPES, spin-resolved ARPES and spatially resolved ARPES”^[61]. The progress  V and soft-X-ray synchrotron light sources make it possible to distinguish between bulk and surface states for TI materials. Spin detectors determine the spin state of emitted electrons, allowing insight into spin-textures of surface states. Time-resolved ARPES (using femtosecond pulses) allows observation of “ultrafast electronic dynamics and states above the chemical potential”^[61]. Finally, spatially resolved ARPES can be used to pinpoint sub-micron scale features, particularly if you want to distinguish between phases, or across material gradients (such as gradient MBE growths).

Spin Resolved ARPES The first spin resolved (SR) ARPES that yielded the direct observation of helical spin-polarization was done by Hsieh *et al.*^[24] at the end of 2008 (see Fig. 20), again in $\text{Bi}_{1-x}\text{Sb}_x$, followed with more detailed results by Nishide *et al.*^[62].

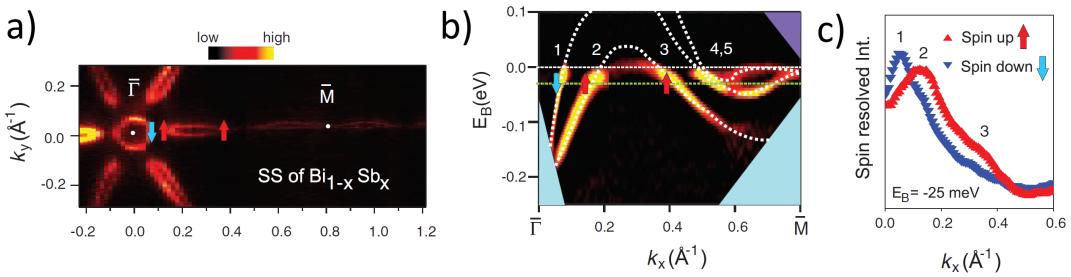


Figure 20: ARPES observation of spin polarised surface states in $\text{Bi}_{1-x}\text{Sb}_x$. **a)** Spin integrated intensity map of $\text{Bi}_{0.91}\text{Sb}_{0.09}$ at E_F . **b)** Surface band-dispersion along the $\bar{\Gamma} \rightarrow \bar{M}$ direction showing 5 Fermi-level crossings. **c)** Spin-resolved momentum distribution curves at binding energy $E_B = -25$ meV, showing band degeneracy.

Source: Hsieh *et al.*^[24], Science Vol. 323, 920 (2009)

In these experiments, topological details could be found by studying the surface band-dispersion and their respective spin polarizations. For example, the fact that there are 5 bands implies that an odd number of Fermi-surfaces enclose the spin-degenerate K points. This gives rise to determining the topological quantum number $\nu_0 = 1$ (0 if even).

However Ando argued that these conclusions were questionable, due to purely relying on states already existing in Bismuth, which did not agree well with theoretical calculations^[9].

A study completed by April 2009 and published in July also by Hsieh *et al.* made the first conclusive observation of helical Dirac fermions in Bi_2Se_3 ^[36]. Here the detection of the spin angular momentum components were measured by the use of two 40-kV Mott detectors.

7.2.2 Spin polarized STS

The first scanning tunnelling spectroscopy (STS) study on $\text{Bi}_{1-x}\text{Sb}_x$ was published in 2009 by Roushan *et al.*^[63] who detected the absence of backscattering even with strong system disorder. By taking fourier transforms of the surface state data they take, they can infer scattering information and lattice information. Combining this with spin-resolved ARPES they could determine the spin dependent scattering probability, and match their STS measurements to 95%. Doing the same with spin-independent scattering yields a match of only 80%.

7.3 Transport Metrological Methods

7.3.1 Quantum Oscillations

Taskin & Ando^[64] used quantum behaviour reported in Bi and $\text{Bi}_{1-x}\text{Sb}_x$ to differentiate between coherent electronic transport (edge states) and incoherent transport within an impurity band. The measurements (along with magnetic & resistivity measurements) were implemented using the “de Haas-van Alphen (dHvA) effect. To do this, you measure the magnetisation M across magnetic field strengths B. They also measured this quantity for different orientations of the sample, however it is also present in the bare resistivity of the sample.

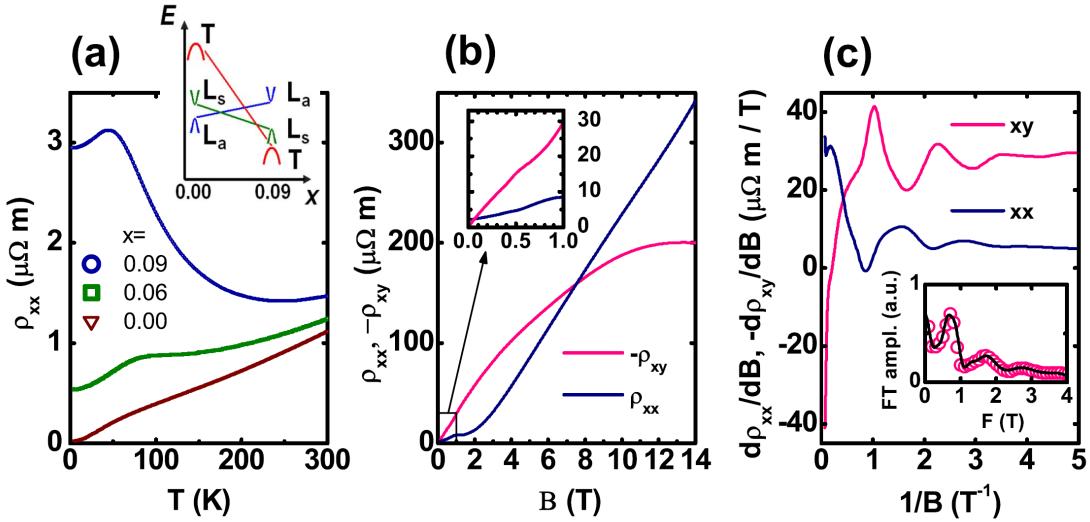


Figure 21: Observation of the quantum oscillations in $\text{Bi}_{1-x}\text{Sb}_x$.

- a) Temperature dependent ρ_{xx} for different x . b) Magnetic field dependent ρ_{xx} and ρ_{xy} for $x=0.09$ @ 1.4K c) Derivatives of b), against $1/B$. Inset shows FT spectrum $-\rho_{xy}$.

Source: Taskin and Ando^[64] Phys. Rev. B 80, 085303 (2009)

The derivative of the both ρ_{xx} and ρ_{xy} with respect to B, plotted against $1/B$ show Shubnikov-de Haas (SdH) oscillations below 2T field strength, as shown in Fig 21c. This is a quantum effect, and consequently is argued that it comes from a well-defined Fermi surface, not some impurity band which wouldn't be stable. The previous measurements of the resistivity with different doping proportions confirmed the insulating nature of the samples.

7.3.2 Weak localisation and anti-localisation

TODO.

7.3.3 Magnetization

Taskin & Ando^[64] measured DC magnetization using a SQUID magnetometer.

8 Quotes

Finally we introduce the parameter of which all the fuss is about.

R.F. Hofstadter,^[6]

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