## 1 Title of the Project

Investigation of charge and spin transport in ultrathin films and nanoribbons based on bismuth selenide ($Bi_2Se_3$) and bismuth telluride ($Bi_2Te_3$)

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## 4 Abstract

Helical Dirac fermions (HDF)—charge carriers that behave as massless spin polarized Dirac fermions—are important for future dissipationless spintronic and computing technologies [1-4]. Requirements for spin polarized surfaces states are satisfied by the presence of odd number of Dirac cones in the surface Brillouine zone of topological insulators (TI). TIs are new class of materials with bulk band gap but with topologically protected metallic HDF surface (in 3D) or edge states (in 2D) that are robust to impurity scattering even at room temperature [5-7].
It has recently been shown that $Bi_2Se_3$ and $Bi_2Te_3$ based TIs possess tunable topological order at room temperature which can be controlled by surface doping or gate control paving way for studies of field effect TI [8] devices analogous to recent research on graphene.

To the best of our knowledge, no room temperature electrical based spin transport measurements of HDF exist. Here we propose to experimentally study spin and charge transport in multiterminal lateral devices based on $Bi_2Se_3$ and $Bi_2Te_3$ TIs where some interesting and observable effects can be measured due to the strong spin orbit coupling (SOC) in TIs. Room temperature gate tunable spin and charge transport measurements in ultrathin crystals of $Bi_2Se_3$ helps to understand the spin-charge coupling in the surface states. Recently, ultrathin $Bi_2Se_3$ crystalline films are obtained by "graphene-like" mechanical exfoliation due to the presence of van der Waals force between quintuple building layer blocks of the crystal from crystalline $Bi_2Se_3$ and $Bi_2Te_3$ [9-11].

Motivated by the recent observation of quantum interference and magnetoresistance in nanoribbons of TIs [7], we also propose to study suspended thin TI nanoribbons to gain access to bottom and top surface states whose coupling in thinner films (~6nm) [12, 13] result in an energy gap in the topologically protected metallic surface states of the $Bi_2Se_3$ film. The gapped surface states exhibit sizable spin-orbit splitting that potentially can be exploited

to study the direct or inverse quantum spin Hall effect (QSHE) at room temperature. Spin transport for various thicknesses will be investigated to understand the influence of the crossover between helical surface states to chiral edge states. Lithographically defined nanoribbons will be studied to understand and compare transport between the diffusive and (quasi) ballistic regimes.

## 5 Duration of the Project

4 years, starting from September 2010

## 6 Personnel

### 6.1 Senior-scientists

| Name | Task in Project | Time |
|---|---|---|
| Prof. dr.ir Bart J van Wees | Supervision and Management | 10 % |
| Prof. dr. Petra Rudolf | Supervision and Analysis | 5 % |
| Dr. N. Tombros | Supervision and Analysis | 10 % |

### 6.2 Junior-scientists and technicians

| Name | Task in Project | Time |
|---|---|---|
| PhD student (OIO) | Experiments and Analysis | 90 % |
| Johan Holstein | Technical Support | 5 % |
| Bernard Wolfs | Technical Support | 5 % |

## 7 Cost Estimates

### 7.1 Personnel Positions

One 'onderzoeker in opleiding' position for four years.

### 7.2 Running Budget

15 k€/year for conferences, summer schools and maintenance.

### 7.3 Equipment (k€)

For this project we can use equipments from our graphene research.

| Equipment | Costs (k€) |
|---|---|
| Stanford Research Systems SR830 DSP lock-in | ~ 5 |
| Edwards Ext Turbo pump (optional) | ~ 3 |
| Liquid Helium and Nitrogen | ~ 5 |
| $Bi_2Te_3$ and $Bi_2Se_3$ crystals | ~ 2 |
| TOTAL | ~ 15 |

### 7.4 Other Support

This project will be supported by Zernike Institute of Advanced Materials and Rijksuniversiteit Groningen. They will contribute in the positions of the technician, the senior scientists and purchase of pure $Bi_2Se_3$, $Bi_2Te_3$ and Ca doped $Bi_2Se_3$ samples.

### 7.5 Budget Summary (in k€)

The expenses are summarized in the following table:

| | | 2010 | 2011 | 2012 | 2013 | 2014 | Total |
|---|---|---|---|---|---|---|---|
| **Position** | PhD Student | 15 | 43 | 43 | 43 | 28 | 172 |
| | Postdocs | - | - | - | - | - | - |
| | Technicians | - | - | - | - | - | - |
| | Guests | - | - | - | - | - | - |
| **Costs** | Personnel | 15 | 43 | 43 | 43 | 28 | 172 |
| | Running Budget | 6 | 15 | 15 | 15 | 9 | 60 |
| | Equipment | 15 | - | - | - | - | 15 |
| **Total** | | 23 | 93 | 58 | 58 | 40 | 247 |

## 8 Research programme

### 8.1 Introduction

Present day electronic devices following Moore's law are fast approaching the physical limits of miniaturization. Therefore, ever-smaller transistors, capacitors and other memory elements with low power consumption are required. Spintronics is envisaged to tackle this problem by avoiding resistive heating in such small devices in a foreseeable future. Within this context TIs are an attractive option for dissipationless spintronic devices.

The recent theoretical prediction of the intrinsic quantum Hall effect (ISHE) and its experimental discovery has generated great interest in the field of spintronics [14]. The effect allows for direct spin accumulation without an external magnetic field and the resulting spin current is dissipationless which can be realized in TIs [15].

ISHE in HgTe based quantum wells was observed in the group of L. Molenkamp in Würzbürg [16]. They showed that transition between trivial insulating edge states to non trivial helical edge states is obtained at a critical thickness $d_c$=6.3nm of the HgTe quantum well. Subsequent nonlocal electronic transport experiments have confirmed the existence of spin polarized edge states in the quantum spin Hall state without the application of an external magnetic field as opposed to the ordinary quantum Hall edge states that are usually observed at high magnetic fields [17, 18].

This confirms that the quantum transport through the (helical) edge channels is dissipationless with counterpropagating spin states at the edge.
In these TIs, there are robust spin quantum Hall surface (in 3D) or edges states (in 2D) with strong SOC that move in opposite directions for opposite spins (see Fig. 1)

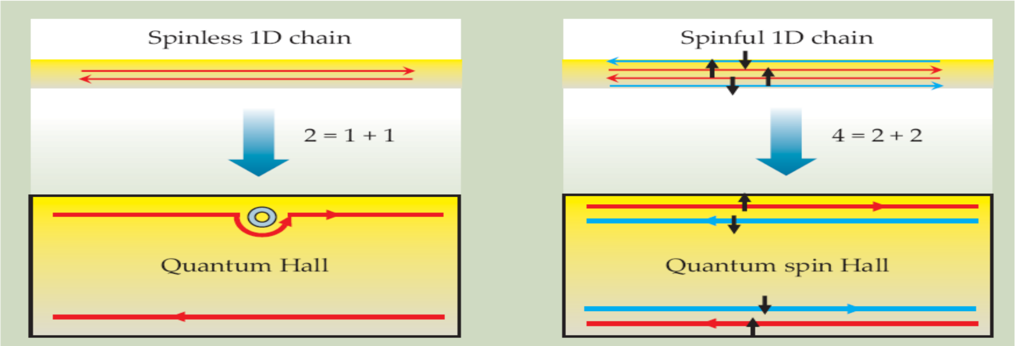
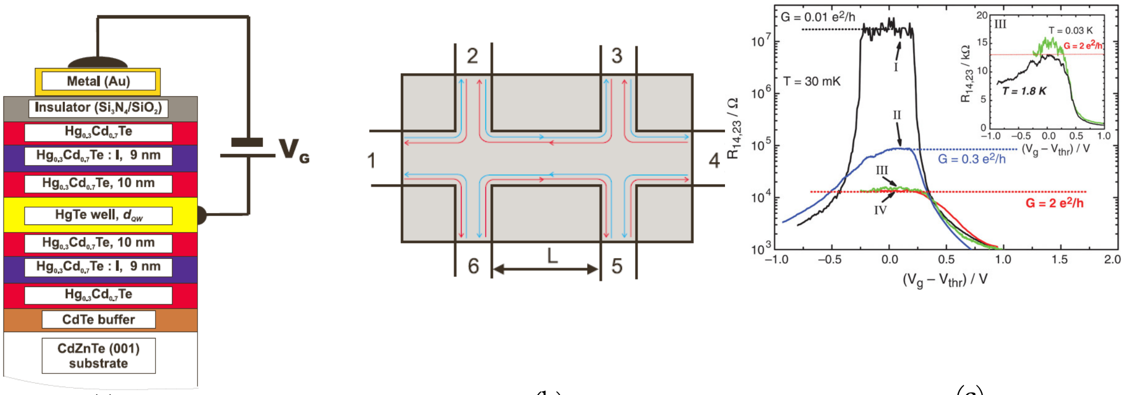


**Fig. 1: Illustration of the two quantum states of matter in 2D (a) shows the spinles Quantum Hall edge states that requires high magnetic field for its observation. Upper and lower edge states contain two oppositely movin states that are robust to scattering. (b) shows spin quantum Hall state with spin full 1D edge states observed without an external magnetic field due to strong SOC. Picture taken from ref. [19]**

So far edge channel transport in HgTe quantum wells is being reported only from the Würzburg group due to their expertise of more than a decade on growth of HgTe nanostructures. And yet growth optimization, gate controlling and measurements were difficult [20, 21]. Besides to this, the toxicity of Hg demands use of other room temperature TIs. This is where thin films obtained from families of Bi alloys come in. It has recently been confirmed by theoretical [1] and experimental works [8-10, 12, 22] that topologically protected spin Hall states exist in 3D topological insulators due to the strong spin orbit interaction. Since then it has attracted large interest due to the potential applications in spintronics and computational applications. This is also driven by the possibility of tuning the Fermi energy by high dielectric oxide top gate [23] and the possibility of achieving ultrathin $Bi_2Se_3$ films that preserve the topologically defined states. Currently thin films of $Bi_2Se_3$ samples can be produced by vapor phase epitaxy on Si(111) surface[24], by catalytic growth [25] and most interestingly by “graphene-like” mechanical exfoliation [6, 11].



(a) (b) (c)

**Fig. 2: Quantum spin Hall effect in HgTe quantum wells. (a) shows the complex growth to obtain HgTe quantum walls. (b) shows the nonlocal geometry employed to measure quantum spin Hall edge states at low temperature thereby proving the robustness of the edge channels. (c) shows four probe conductance for thre different width of the quantum well for which transition between the ordinary insulator to quantum spin Hall edge channels is obtained or a critical width dc=6.3nm. pictures taken from ref. [5]**

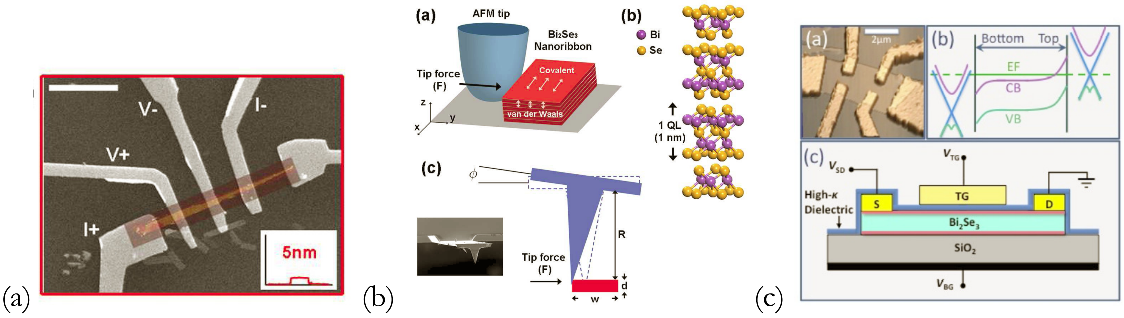
Now we propose the experimental study of spin transport in multiterminal lateral geometries and Hall bar geometries in suspended and non suspended samples of thin $Bi_2Se_3$ and $Bi_2Te_3$ TIs. Since our group has developed an expertise in spin transport measurements in graphene and other metallic systems, we believe that our proposed project will achieve the goal of studying spin transport in such materials at room temperature. $Bi_2Se_3$ and $Bi_2Te_3$ are therefore suitable to study charge and spin transport at room temperature and subsequently understand spin-charge coupled transport on the helical surface states of TIs.

We expect emergence of new magnetoresistance effects due to the strong SOC in our TI based lateral spin valve devices which are measurable and observable at room temperature. We analyze our measurements based on drift-diffusion model previously developed for lateral geometries but this time taking in to account the strong spin orbit interaction.

We also propose to study spin dependent thermal transport measurements in our lateral devices in view of the interesting high and low temperature thermoelectric applications of these two TIs [26, 27]. We also demonstrate tunability of spin (charge) transports and related effects by engineering devices with top or bottom gate voltages to control HDF in 2D and 3D TIs. Since both quantum Hall and spin quantum Hall states can be achieved in $Bi_2Te_3$ and $Bi_2Se_3$ TIs by varying the thickness of the film [12, 13], we also thrive to realize a 1D *p-n* interfaced lateral spin valve device by varying the global and local charge density a back gate and top gate respectively. We also like to put a recent prediction by C. Beenaker *et al.* into a test where the possibility of spin injection, precession and detection in the quantum Hall state of 2D TI (HgTe in their case) is proposed [28].

## 8.2 Device fabrication

Crystals of $Bi_2Se_3$, Ca doped $Bi_2Se_3$ and $Bi_2Te_3$ are widely available in the market for reasonable prices. Starting out with "graphene-like" mechanical exfoliation, we try to achieve ultrathin crystalline films following the procedure reported in ref. [10]. A typical crystalline film obtained with this method is shown in the figure below.



(a) (b) (c)

**Fig. 3: (a) SEM micrograph of thin crystalline $Bi_2Se_3$ of 5nm thick obtained by mechanical exfoliation. Lithographically defined contacts are deposited for charge transport measurements similar to the process of device fabrication in graphene. (b) Shows the method of mechanical exfoliation using contact mode AFM. Quintuple layers are ripped of the stack of $Bi_2Se_3$ by lateral force applied by the AFM tip. (c) top gated $Bi_2Se_3$ thin film of 17nm thick in a Hall bar geometry where ambipolar electric field has been claimed in ref. [23]. Inset shows schematic variation of the band structure along the z-direction showing the tunable Fermi energy of surface states by top gate.**

Thickness and homogeneities of the obtained thin films will be characterized following our well established graphene sample characterization techniques. To study transport in nanoribbons of TIs we follow well developed techniques employed by Popincuic *et al.* where graphene nanoribbons down to 50nm wide are achieved by few lithographic steps and usage of reactive ion etching [29].

### 8.3 Measurement techniques

We perform spin transport measurements similar to our previous spin transport measurement in graphene based spin valve devices [29-32]. This mainly consists of charge and spin transport measurement in a multiterminal spin valve devices to confirm existence of spin transport in TIs. For this, combination of ferromagnetic/normal non magnetic electrodes will be used to detect spins with/without spin sensitive contacts. To understand the charge and spin transport, we first prepare prototype devices with non magnetic contacts as shown in Fig. 4. After this we perform both types of transport measurements by introducing a spin sensitive ferromagnetic contact in combination with normal contacts. At later stages spin injection and detection by ferromagnetic contacts will be performed and comparison will be made with the results obtained earlier. These results help in understanding the influence of magnetic electrodes on the HDS as wel us giving information on the spin transport parameters in a TI.

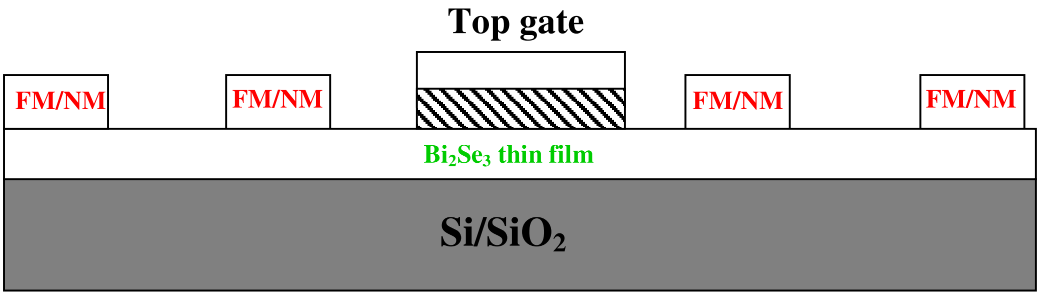


**Fig. 4: Schematic picture of a multiterminal lateral spin valve device. Spin sensitive ferromagnetic contact (FM) or normal (FM) contacts deposited on thin film of $Bi_2Se_3$ and $Bi_2Te_3$ deposited on $Si/SiO_2$ substrate. Back gate is used to globally control the total induced charge carriers in the TI where as the top gate is used to locally control the induced charge carriers in the TI to obtain different regimes of p-n junctions.**

Hanle spin precession measurement—which is typically used to extract the spin diffusion coefficient, spin life time and spin relaxation lengths—will not be performed. This is because perpendicular magnetic fields applied to the quantum spin Hall states removes time reversal symmetry [5] and thus HDFs. Rather, since an in plane magnetic field does not destroy the quantum spin Hall states, we perform length dependent spin valve measurement for different distances between injector and detector to be able to extract the spin relaxation lengths from a drift-diffusion spin model that takes into account the strong SOC. Since HDFs are robust to any impurity scattering, we expect the spin relaxation time in such devices to be similar to the momentum scattering time. These types of study helps to understand what kind of spin relaxation mechanisms are dominant.

## 8.4 Goal of the proposal

The general goal of this proposed research is the realization of quantum spin Hall transistor made of $Bi_2Se_3$ and $Bi_2Te_3$ TIs with multiple terminals. Spin valve devices with different distance between injector and detector electrodes will be prepared. Result of such measurement will be used for subsequent realization of spin transport in TIs and two other proposed works by S.H.Chen *et al.* [14, 33] where they showed possibility of achieving spin accumulation in a TI from a precessing ferromagnet.

Here we present a list of tasks that are directly or indirectly related to the general goal of the proposed work.

1. fabrication of ultrathin crystalline films of $Bi_2Se_3$ and $Bi_2Te_3$ of few quintuple layers by using mechanical exfoliation which are then deposited on $Si/SiO_2$ substrate. Similar device characterization which is established in our group for graphene research will be used (for example optical microscopy, AFM and Raman spectroscopy)
2. charge transport measurement in two-and four probe geometry for various thicknesses of films.
3. observation of ISHE in ultrathin TIs made of $Bi_2Se_3$ and $Bi_2Te_3$
4. study of the influence of surface non magnetic dopants
5. study the effect of coupling of top and bottom surface states as the thicknesses of the samples get smaller (less than 6 quintuple layers)
6. measure spin transport at low temperature and room temperature based on multilateral spin valve geometry
7. Study inverse spin Hall Effect by spin pumping from precessing magnetization into $Bi_2Se_3$ and $Bi_2Te_3$ based 2D TI [14, 33].

## 8.5 Collaborations

This project benefits from collaborations with different research groups within and outside the institute. The interdisciplinary nature of Zernike institute of advanced materials means easy access to facilities in the whole institute is possible. We have already set up collaboration with the group of prof. Dr. ir. Paul van Loosdrecht in the usage of Raman spectroscopy for characterization of our graphene and ultrathin $Bi_2Se_3$ and $Bi_2Te_3$ crystalline films. The group of prof. Petra Rudof will be responsible for characterization of our ultrathin films using their surface study facilities including photoemission spectroscopy. We are also planning to set up several collaborations outside our institute. We plan to set up collaboration with the group of prof. S. Balandin of the university of California—Riverside who have recently managed to obtain ultrathin films of $Bi_2Se_3$ and $Bi_2Te_3$ samples. They have characterized thin films using Raman spectroscopy to clearly identify the thickness of thin TI films. Useful exchange of resources and ideas to optimize our own device preparation procedures will be part of collaboration. In addition, samples may be requested from them to perform our electrical spin transport measurements.

### 8.6 Plan of Work

The plan of work is summarized in the following table:

| Time (months) | Activity |
|---|---|
| **1st year** | • Preparation and optimization of ultrathin films of $Bi_2Se_3$ by mechanical exfoliation. In this phase, different methods of mechanical exfoliation will be tried. The cheapest of all is the scotch-tape method. Other recently proposed methods of exfoliation using contact mode AFM will also be employed. Here optical microscopy, AFM and Raman microscopy are used to identify the thickness of films.<br>• Fabrication and optimization of lithographically defined contact deposition for electrical charge and spin transport. |
| **2nd year** | • Measure and confirm existence of spin Hall effect for various thicknesses and collect data to interpret crossover between 3D surface states and 2D edge states<br>• Spin and charge transport measurements on various thicknesses of $Bi_2Se_3$ and Bi2Te3 films |
| **3rd year** | • Preparation of nanoribbon TIs and study of charge and spin transport in the diffusive and (quasi) ballistic regime. |
| **4th year** | • Study inverse spin Hall effect in $Bi_2Se_3$ and $Bi_2Te_3$ thin films as proposed by S.H.Chen *et al.* [14, 33] (optional)<br>• Follow up of research<br>• Thesis writing |

All measurements will be performed both at low temperature and room temperature. Of course, deviations from this scheme might occur depending on the problems or interesting new physics encountered.

## 9 Infrastructure

The Physics of nanodevices group has a variety of processing and measuring facilities. For this project the most important facilities are:

- A cleanroom equipped with general processing facilities such as a mask aligner, an e-gun evaporator, resist spinner, ovens, optical microscope and a reactive ion plasma etcher
- A Raith e-line electron beam lithography system
- A JEOL inspection scanning electron microscope
- A digital instruments Nanoscope IV room temperature scanning probe microscope

We are planning to extend our sample preparations and measurements facilities by using:

- Home built ‘IV meetkast’ for electrical measurements.
- Oxford cryostat for our low temperature measurements which is part of our big graphene research

The group of prof. Dr. Ir. Paul van Loosdrech has a Raman spectroscopy setup which will be used for our TI thin film characterization. Other measurement setups which are not mentioned here may be used depending on need.

## 10 Application perspective in industry, other disciplines or society

Since helical Dirac fermions easily transport along the surface or edges without any dissipation, the experimental realization of a TI based spin devices will be a milestone in future spintronics and fault tolerant computations. The experimental realization of room temperature TI based field effect devices based on $Bi_2Se_3$ and $Bi_2Te_3$ will hence revolutionize future spintronic devices that are free of dissipation solving the current problems in miniaturization of conventional electronics.

Moreover, at the end of this project, fundamental information on charge and spin transport will have been gained even if experimental realization of a TI spin valve device is not achieved. The proposed work will also be a first step towards realizing spin field effect transistors that require strong SOC for gate tunability as proposed by Datta and Das [34].

Oscillatory crossover from two-dimensional to three-dimensional TIs surface states means careful engineering of thicknesses of mechanically exfoliated thin films can result in the realization of trivial insulating and non trivial quantum spin Hall states in the same material without requiring high magnetic fields[12].

In addition to the proposed work’s importance in understanding basic transport properties of a TI, it will also facilitate the study of some exotic phenomena like the existence of magnetic monopoles and Majorana fermions in ferromagnetic and superconducting heterostructures of TIs which are so far holy grails of fundamental physics [3, 35].

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