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# Nanostructures of disordered 3D topological insulators

Romain Giraud

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# Nanostructures of disordered 3D topological insulators

## MÉMOIRE

soutenu le 20 octobre 2020

pour l'obtention d'une

**Habilitation de l'Université Grenoble Alpes**  
(Spécialité : Physique)

par

Romain GIRAUD

### Composition du jury

*Président :* Le président

*Rapporteurs :* Prof. Stephan ROCHE, ICN2 Research Scientist, ICN2/UAB (Barcelona, Spain)  
Prof. Sebastian GÖNNENWEIN, TU Dresden Professor, IFMP (Dresden, Germany)  
Dr. Michel VIRET, CEA Research Scientist, CEA SPEC (Saclay, France)

*Examineurs :* Prof. Sergio VALENZUELA, ICN2 Research Scientist, ICN2/UAB (Barcelona, Spain)  
Dr. Olivier FRUCHART, CNRS Research Scientist, CEA SPINTEC (Grenoble, France)  
Prof. Mairbek CHSHIEV, UGA Professor, CEA SPINTEC (Grenoble, France)



*"A ma Famille."*

*"En vérité, le chemin importe peu, la volonté d'arriver suffit à tout."  
Albert Camus, Le mythe de Sisyphe.*



## Acknowledgments

In this period of pandemic, I am particularly grateful to all members of this habilitation jury : SEBASTIAN GÖNNENWEIN, STEPHAN ROCHE and MICHEL VIRET for their review of my manuscript ; MAIRBEK CHSHIEV, OLIVIER FRUCHART and SERGIO VALENZUELA for their participation and contributions during this process.

Many thanks also to ADRIANA STOENESCU and ALAIN MARTY for their help in the organization of the defense.

At the time to get an accreditation to supervise research and PhD students, i would like to start these acknowledgments with some words about my professors. Although education is a process to develop an independent mind, some positively intrusive lessons helped make it faster. I am particularly thankful to ELIE BELORIZKY, and to JOSEPH LAJZEROWICZ who gave me some finger pain (but never a headache!) and significantly increased my usage rate of new pencils. Teaching physics to its quintessence and relate it sometimes to some History of Science or daily Life remains, to me, the ultimate education experience.

To become a condensed-matter physicist with some knowledge in mesoscopic physics, i learnt a lot from both JOËL CIBERT and LAURENT LÉVY. Their lessons have become some fertile grounds to develop the research deccribed in this manuscript, with an independent mind but also relying on solid grounds.

As a PhD student, i nurtured a *penchant* for doing research with a free mind, and developed by that time a strong taste to explore some physics outside the box but, say, close to the interface nevertheless. My doctor father BERNARD BARBARA has been a real mentor in this early quest, and i am indebted to him for his kindness and constant support in my academic career.

My research on spin-related charge transport in nanostructures started during my post-doctoral stay in Würzburg, and i am grateful to LAURENS MOLENKAMP for initiating my journey into the physics of (Ga,Mn)As.

Since then, i had the chance to work with, and sometimes supervise, many bright and friendly students and scientists.

At CNRS/LPN in Marcoussis, together with my colleagues ARISTIDE LEMAÎTRE and GIANCARLO FAINI, i impulsed the “MesoSpin” project with the idea to combine magnetism and mesoscopic transport in crystalline (Ga,Mn)As nanostructures. Among the too many people i could thank here, the members of the Phynano group : Carole, Marta, Edgar, Laura, Joseph, Laurent, Mara, Fabien, Carles, Hélène, Gwendal, François, Damien, Ulf, Karim, Bernard, Antonella, Fred, Yong, Guillemin, Jean-Christophe. And this is the laboratory where i have learnt how to repair a one-way valve with a latex glove...what i call the DOMINIQUE MAILLY’s school : “quick and dirty”...but always efficient !;-) Finding the crux of the problem and fixing it like MacGyver.

At the IFW Dresden, I had to change my research project and moved to novel topological insulators. I am grateful to BERND BÜCHNER for giving me this opportunity to build a new lab from scratch, and to further develop some expertise in low-noise magneto-transport measurements combining 3D superconducting magnets with  $^3\text{He}$ - $^4\text{He}$  dilution refrigerators. At the Institute for Solid State Physics, i set up the Quantum Transport group together with my colleague JOSEPH DUFOULEUR and since then we have a great collaboration, with passionate discussions always (and even one famous disagreement due to semantic only !). And, as a member of a famous winemaker family in Burgundy, wine is also another topic of interest to discuss about : remember when we desperately searched the entrance to attend a conference dinner in Würzburg

(being too late to enter the Würzburger Residenz) and finally found the wine caves..., or that time when i learnt that a good red wine can improve critical temperatures in superconductors. In this new journey to investigate 3D topological insulators, there are two persons being essential, our colleagues LOUIS VEYRAT and SILKE HAMPEL. Thanks for your great contributions and friendship! With the wine (or beer) comes the food...(yes, we are a german-french group)...and we are always missing Louis' delicious cakes [I can confess here that we -carefully- broke the rule -once only- not to introduce food in the laboratory, so as to taste liquid-nitrogen-cooled meringues. Worth the 'risk'!]. Novel magnetic topological insulators have brought some more chat about physics on the way to work...so many thanks to my colleague ANJA WOLTER-GIRAUD for our thorough discussions, always, and the way you appreciate my simple models, sometimes;-) Many thanks also to our IFW colleagues in general, and in particular to Hannes, Valentin, Aoyu, Arthur, Guillaume, Barbara, Sandra, Martin, Falk, Tino, Danny, Sebastian, Andy, Kevin, Katja, Kerstin.

In 2016, i resumed my CNRS position, joining SPINTEC. I am grateful to BERNARD DIENY, OLIVIER FRUCHART and LUCIAN PREJBEANU for their support and scientific goodwill. Many thanks to my colleagues from the SpinOrbitronics group, in particular to LAURENT VILA who is always a model for being both a very good and friendly physicist! Well...we also enjoyed some good wine (again in Würzburg!) and should certainly try some more beers as well! I am looking forward to working together on even more common projects -and maybe resume some old ones on (Ga,Mn)As, as well as with our other colleagues VINCENT BALTZ, MATTHIEU JAMET and FRÉDÉRIC BONELL. Many thanks also to many other colleagues, either at SPINTEC or at the PTA, and to our administration colleagues who always combine some good mood, a lot of understanding and efficiency : Rachel, Léa, Sabrina, Catherine, Céline and Adriana.

Beyond in-house cooperation, my recent research also benefited from great external collaborations with theoreticians (JENS BARDARSON, JÉRÔME CAYSSOL) and experimentalists (WALTER ESCOFFIER, GIACOMO PRANDO, NINI PRYDS, LAMBERT ALFF, EUGENIA TOIMIL-MORALES and ANNA ISAEVA), many thanks to all of you!

To finish, i would like to emphasize that my everyday thoughts go to my loving family, both in France and in Germany, and in particular to my wife ANJA who is giving me her forever strong support, trying her best not to complain too often about my too many travels and absence... Danke!

# Outline

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# Curriculum vitae



## 1 Personal Data

ROMAIN GIRAUD

born on 16 December 1975 in Lagny (77), France.

ADDRESS : Wittenbergerstr. 91, 01277 Dresden, Germany

FAMILY STATUS : Married, One child

AFFILIATION :

**Institution :** CNRS

**Since :** 01 October 2004

**Department :** Institute of Physics

**Position :** Chargé de Recherche

**CNU section :** 28<sup>ème</sup> section

**Laboratory :** SPINTEC

PROFESSIONAL ADDRESS :

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## 2 Work Experience

09/2002-09/2003	Post-Doc at the Joseph Fourier University, Grenoble (France), with Dr. J. Cibert, “Study of (Ga,Mn)N epilayers”
10/2003-09/2004	Post-Doc at the Technical University, Würzburg (Germany), with Prof. L. Molenkamp, “Classical spintronics in GaMnAs nanostructures and tunnel junctions”; <i>Fellow of the Alexander von Humboldt foundation</i>
Since 10/2004	Permanent CNRS researcher
10/2004–08/2010	Phynano Team, CNRS/LPN, Marcoussis (France); Leading the MesoSpin project for Quantum Spintronics
09/2010–10/2016	Secondment at the IFW, Dresden (Germany); Leading the Quantum Transport group at the Institute for Solid State Research (Prof. B. Büchner)
Since 11/2016	CNRS researcher at CEA/SPINTEC, Grenoble (France); Senior scientist in the SpinOrbitronics group, working on spin transport in topological insulator nanostructures and quantum spintronics in ferromagnetic nanostructures
Since 01/2019	Co-director with O. Fruchart of the International Research Project “SPINMAT” (SPINTEC/IFW Dresden)

### 3 Scientific Education

10/2002	DOCTORATE DEGREE, Dr. rer. nat. University Joseph Fourier, Grenoble “Quantum tunneling of weakly-coupled Ho <sup>3+</sup> magnetic moments”
10/1999–09/2002	PhD student at the Joseph Fourier University; CNRS grant
06/1999	ENSPG ENGINEER DIPLOMA INPG, Grenoble Institute of Technology
06/1999	MASTER DEGREE IN PHYSICS University Joseph Fourier, Grenoble “Ising ground state of individual rare-earth magnetic moments”
09/1996–06/1999	Student of the ENSPG (INPG Engineer school of physics)

### 4 Research expertise

RESEARCH FIELDS :

Mesoscopic physics, Nanomagnetism, Spintronics, Topological materials

EXPERIMENTAL SKILLS :

Nanofabrication (incl. e-beam lithography), mK cryogenics, Quantum transport measurements in vector magnetic fields, Hall-bar magnetometry

Research expertise in nanomagnetism and nanoelectronics, with a focus on spin-dependent quantum properties in nanostructures and/or heterostructures. Since 2010, topological insulators are mostly studied, either in low dimension (quantum wires) or in novel magnetic van-der-Waals heterostructures (topology and magnetism), with the goal to study novel quantum states of interest for spintronics, such as : the quantum anomalous Hall effect, Weyl semimetals, axion or Chern insulators.

### 5 Research supervision

Since 2005, i regularly supervised students and post-docs, who contributed with me to our main research activities, either on (Ga,Mn)As or on topological insulators.

POSTDOCS :

11/2004–09/2005	MARTA GRYGLAS Laboratory of Photonics and Nanostructures, Marcoussis “Ferromagnetic $p^{++}$ -GaMnAs/ $n^{++}$ -GaAs Zener-Esaki diodes”
Present situation :	Professor, Faculty of Physics, University of Warsaw
10/2005–07/2006	LAURENT VILA Laboratory of Photonics and Nanostructures, Marcoussis “Quantum transport in (Ga,Mn)As nanostructures”
Present situation :	Research Scientist, CEA, Grenoble

02/2008–10/2008 | FABIEN CHEYNIS  
Laboratory of Photonics and Nanostructures, Marcoussis  
“Ferromagnetic  $p^{++}$ -GaMnAs/ $n^{++}$ -GaAs Zener-Esaki diodes”  
Present situation : Assistant Professor, Physics Department, University of Aix-Marseille

06/2009–08/2010 | MARA GRANADA  
Laboratory of Photonics and Nanostructures, Marcoussis  
“Quantum transport in (Ga,Mn)(As,P) nanostructures”  
Present situation : Research Scientist, Centro Atómico, Bariloche

03/2014–04/2016 | BASTIEN DASSONNEVILLE  
IFW, Dresden  
“Quantum transport in  $\text{Bi}_2\text{Se}_3$  nanowires”  
Fellow of the AvH foundation  
Present situation : Research Fellow, PML/NIST, Boulder

PHD STUDENTS :

10/2006–09/2009 | EDGAR BRIONES  
LPN/University Paris XI, Orsay  
“Tunnel anisotropic magnetoresistance  
of ferromagnetic  $p$ -GaMnAs/ $n$ -GaAs Zener-Esaki diodes :  
PhD defense in Orsay on 10/2009  
Present situation : Research Scientist at the Jesuit University of Guadalajara

10/2012–12/2015 | LOUIS VEYRAT  
IFW/Technical University, Dresden  
“Quantum transport in 3D topological insulators nanostructures”  
PhD defense in Dresden on 05/2016  
Present situation : Research Fellow at EP4/TU Würzburg

10/2017–03/2021 | VALENTIN LABRACHERIE  
SPINTEC/IFW Dresden  
“Magnetotransport properties of  $\text{WTe}_2$  nanostructures”  
Present situation : Co-tutelle PhD Student UGA/TU Dresden

12/2017–05/2021 | AOYU TAN  
SPINTEC/IFW Dresden  
“Magnetic topological insulator nanostructures”  
Present situation : Co-tutelle PhD Student UGA/TU Dresden

## MASTER STUDENTS :

02/2012–06/2012	LOUIS VEYRAT IFW, Dresden “Magneto-transport properties of Bi <sub>2</sub> Se <sub>3</sub> nanowires” Master of Physics, University Paris XI, Orsay Present situation : Research Fellow at EP4/TU Würzburg
03/2012–09/2015	KUMAR PRATEEK IFW, Dresden “Aharonov-Bohm oscillations in Bi <sub>2</sub> Te <sub>3</sub> nanowires” Erasmus Mundus Master in Nanoscience and Nanotechnology, Leuven Present situation : PhD Student, Leiden University
03/2016–09/2016	VIVEKSHARMA PRABHAKARA IFW, Dresden “Novel candidates for topological insulators, Bi <sub>4</sub> I <sub>4</sub> and Bi <sub>4</sub> Br <sub>4</sub> ” Master in Nanoelectronic Systems, TU Dresden Present situation : PhD Student, EMAT, University of Antwerp
01/2016–12/2016	HANNES FUNKE IFW, Dresden “Magnetotransport studies of Bi <sub>2</sub> Te <sub>3</sub> nanostructures” Master in Physics, TU Dresden Present situation : SAW COMPONENTS Dresden GmbH
03/2017–08/2017	AOYU TAN IFW, Dresden “Spin transport in Bi <sub>2</sub> Te <sub>3</sub> nanowires” Master in Nanobiophysics, BIOTEC/TU Dresden Present situation : PhD Student, TU Dresden/UGA Grenoble
01/2019–09/2019	NARAYAN KUNCHUR IFW, Dresden “Magnetotransport properties of novel magnetic topological insulators” Erasmus Mundus Master in Nanoscience and Nanotechnology, Leuven Present situation : PhD Student, Max Planck Institute, Dresden

## UNDERGRADUATE STUDENTS :

Chunlin YU (06–08/2011)	Fangjia YAN (04–07/2012)
Hannes FUNKE (07/2013–12/2015)	Maria KHARLAMOVA (11/2015–08/2016)
Viveksharma PRABHAKARA (01–02/2016)	
Gurushiddappa NADUVINAMANE (06–09/2016)	
Aoyu TAN (07/2016–02/2017)	Joaquin CORNEJO (06–08/2019)
Marie-Paule OKINDA (06–07/2020)	

## 6 Collaborations

### RESEARCH ON DILUTED FERROMAGNETIC SEMICONDUCTORS :

- Joël Cibert, Henri Mariette (Grenoble)
- Charles Gould, Laurens Molenkamp (Würzburg)
- Aristide Lemaître, Giancarlo Faini (Marcoussis)

### RESEARCH ON LOW-DIMENSIONAL CONDUCTORS :

- Jean-Christophe Harmand, Dominique Maily (Marcoussis)
- Nini Pryds (Copenhagen)

### RESEARCH ON TOPOLOGICAL INSULATORS :

- Joseph Dufouleur, Silke Hampel, Anna Isaeva, Andy Thomas, Bernd Büchner (Dresden)
- Jens Bardarson (Stockholm)
- Jérôme Cayssol (Bordeaux)
- Walter Escoffier (Toulouse)
- Maria Eugenia Toimil-Morales (Darmstadt)
- Laurent Vila, Matthieu Jamet, Frédéric Bonell (Grenoble)

# Publication list

- [1] Chiorescu, I., Giraud, R., Jansen, A. G. M., Caneschi, A., and Barbara, B., “Phonon-assisted tunneling in the quantum regime of  $\text{Mn}_{12}\text{-ac}$ ”, *Phys. Rev. Lett.* **85**, 4807 (2000).
- [2] Barbara, B., Chiorescu, I., Giraud, R., Jansen, A., and Caneschi, A., “Mesoscopic Spin Tunneling in Molecular Crystals”, arXiv :cond-mat/0005268 [cond-mat.mes-hall] (2000).
- [3] Giraud, R., Wernsdorfer, W., Tkachuk, A. M., Maily, D., and Barbara, B., “Nuclear spin driven quantum relaxation in  $\text{LiY}_{0.998}\text{Ho}_{0.002}\text{F}_4$ ”, *Phys. Rev. Lett.* **87**, 057203 (2001).
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- [13] Giraud, R., Kuroda, S., Marcet, S., Bellet-Amalric, E., Biquard, X., Barbara, B., Fruchart, D., Ferrand, D., Cibert, J., and Mariette, H., “Structural and magnetic properties of a  $\text{Ga}_{0.986}\text{Mn}_{0.0014}\text{N}$  epilayer”, *Journal of Magnetism and Magnetic Materials* **272-276**, E1557 (2004).
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# Summaries



# Summary of the research activities and scientific project

My experimental research activities focus on quantum phenomena in magnets and low-dimensional conductors, mostly studied by electrical transport measurements at very low temperatures and in 3D vector magnetic fields. Very different physical systems, such as nanomagnets or topological insulator nanowires, only have in common to be characterized by a Hamiltonian with a well-defined symmetry. For different reasons, a rather weak coupling to their environment preserve the intrinsic nature of these quantum states, including their phase, and quantum interference determines their physical properties. In many cases, as for some discussed in this manuscript, it ultimately gives the possibility to reveal the influence of a geometric phase, the Berry phase, on the spectral properties or physical observables of weakly-coupled quantum systems, related to the topology of their Hilbert space.

What does this means in practice ?

During my PhD thesis with Bernard Barbara, the slow quantum dynamics of the smallest magnet ever, an atomic magnet, was revealed by staircase-like hysteresis loops at very low temperature. Abrupt magnetization jumps are due to quantum tunneling of the electro-nuclear magnetic moment of individual  $\text{Ho}^{3+}$  ions (Landau-Zener tunneling). In a well-defined crystal-field environment, the tunnel probability depends on the energy gap between eigenstates (induced by a symmetry breaking), which can thus be altered by the Berry phase. For a single magnetic moment, this physics can be described by an intuitive semi-classical picture, based on the evolution of the quantum states on a Bloch sphere, with some magnetic field-tunable interference that reveals Berry phase effects.

During my post-doctoral stays, I studied the magneto-transport properties of diluted ferromagnetic semiconductors, with carrier-mediated exchange interaction, using either Hall bars or tunnel junctions [one year in Grenoble, with Joël Cibert/Henri Mariette : Mn-doped GaN ; one year in Würzburg with Laurens Molenkamp : Mn-doped GaAs]. The magnetic anisotropy of ferromagnets such as (Ga,Mn)As is mediated by heavy holes, so that it retains the group symmetry of the nonmagnetic unit cell (a cubic symmetry, with a small degree of uniaxial anisotropy due to strains). With nearly no disorder, small magnetization switching fields are controlled by the nucleation of domain walls. In a simple tunnel junction (Ga,Mn)As/AlOx/Au, with a single magnetic layer, perpendicular magnetic domains lead to the bi-stable tunnel anisotropic magneto-resistance. The key property is that a rotation of the magnetization directly modifies the electronic band structure, which can be probed by transport measurements (local density of states, Berry phase).

As a CNRS researcher at the Laboratoire de Photonique et Nanostructures, I initiated the « MesoSpin » project and started to investigate quantum phenomena in (Ga,Mn)As. The basic idea was simple : in single-crystalline magnets, with a uniaxial anisotropy and little disorder, the phase-coherence length can be long enough so that the influence of magnetic textures on quantum spin transport can be evidenced by quantum interference experiments. This was first confirmed by revealing the dephasing induced by magnetic domain walls and domains, modifying universal conductance fluctuations. In the saturated magnetization state, the exact orientation of the magnetization can be controlled by rather small magnetic fields and Berry phase effects were unveiled.

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From 2010 to 2016, I was seconded at the IFW Dresden, and had to give a new orientation to my work. Since then, my research is focused on novel materials having a non-trivial electronic band structure, with some intrinsic transport properties fixed by the Berry phase. 3D topological insulators are at the core of this research, and their properties are mostly investigated by quantum transport measurements. In particular, a specific expertise was developed to understand the influence of quantum confinement in the low-dimensional geometry of monocrystalline nanowires (additional Berry phase due to the curvature), as well as that of structural disorder. A first contribution was the evidence of the quasi-1D ballistic transport of surface modes, as a signature of the weak coupling of spin-helical Dirac fermions to their environment. This was a striking result, since real materials do have a large degree of disorder. Then, in follow-on contributions, two key ingredients were shown to drive this unusual situation in condensed-matter materials [ballistic transport in highly-disordered conductors], that are : i) the anisotropic scattering of 2D topological surface states [enhanced transport length] and, ii) the strong quantum confinement in nanowires [1D energy spectrum].

Since 2016, my research at SPINTEC is extended to magnetic topological insulators, and related topological quantum materials [Weyl; 2D van der Waals]. Combining magnetism and topology gives new routes to modify electronic band structures and thus to create novel quantum phases. First, the internal magnetic field [mostly, exchange interactions] can open gaps with a non-trivial topology and give a Chern insulator. Second, the magnetic order can add some more symmetries, in addition to crystal symmetries, which can lead to different topological states, such as the axion insulator. Third, finite-size effects are expected in thin films, e.g. due to different surface terminations in layered collinear antiferromagnets, and gapless quasiparticles can propagate along an atomic step. Generally speaking, (i) the control of the order parameter offers the interesting opportunity to tune electronic phases by applying some magnetic fields and, ultimately, (ii) some new concepts can be envisioned when considering magnetic textures, such as domain walls.

In this dissertation, a first part gives an exhaustive review of the results obtained on nanostructures of disordered 3D topological insulators, from nanoflakes to nanowires. Quantum transport measurements are used to evidence some unique properties of spin-helical Dirac fermions, due to their anisotropic scattering by disorder. In quasi-1D quantum wires, confinement in reduced dimension leads to quasi-ballistic transport, with a hollow-conductor geometry as for carbon nanotubes and semiconducting core-shell nanowires, but with strong disorder. The second part describes my on-going research project, with the aim to investigate dissipationless spin-polarized chiral edge states. Two directions are followed : i) the search for the perfectly-transmitted mode in (Bi,Sb)Te nanowires or in core-shell 3D TI heterostructures ; ii) the search for the quantum anomalous Hall and axion insulator regimes (among other topological phases) in novel magnetic topological insulators of the  $\text{MnBi}_2\text{Te}_4$  family, and related materials.

Some previous research, not reviewed in this dissertation, includes :

- (Ga,Mn)As at CNRS/LPN Marcoussis
- $\gamma\text{-Al}_2\text{O}_3/\text{SrTiO}_3$  at the IFW Dresden



# Résumé des travaux et projet de recherche



Mon activité de recherche est centrée sur l'étude des phénomènes quantiques en nanomagnétisme et nanoélectronique, principalement par des mesures de magnéto-transport réalisées à très basses températures et sous champ magnétique vectoriel 3D. Les systèmes étudiés peuvent être très différents, comme un nanoaimant isolé ou un nanofil d'isolant topologique, mais ils ont en commun d'être décrits par un Hamiltonien dont les symétries sont bien définies. Pour différentes raisons, leurs états quantiques sont faiblement couplés à l'environnement, ce qui préserve leur phase et permet d'étudier leurs interférences, à l'origine de certaines propriétés physiques. En particulier, l'évolution des états quantiques dans l'espace de Hilbert induit une phase géométrique (Berry) qui, pour des topologies spécifiques, peut modifier les observables ou induire de nouveaux états quantiques.

Comment ces effets ont-ils été observés dans ce travail ?

Dans mon travail de thèse en nanomagnétisme, sous la direction de Bernard Barbara, nous avons observé la dynamique quantique lente d'un aimant atomique à très basse température. Le renversement du moment magnétique électro-nucléaire d'un ion  $\text{Ho}^{3+}$  se fait par sauts quantifiés, traduisant un processus d'effet tunnel sous la barrière d'anisotropie (effet Landau-Zener-Stueckelberg). La dynamique est induite par un abaissement de symétrie, qui crée des superpositions d'états, et elle peut être modifiée, pour des orientations de champ bien particulières, par la phase de Berry (oscillations de l'amplitude de répulsion des niveaux quantiques). Dans une approche semi-classique, les états quantiques d'un moment magnétique sont décrits par des trajectoires sur une sphère de Bloch et la description des interférences reste relativement simple.

Pendant mon activité post-doctoral, j'ai étudié le transport électrique dans des semi-conducteurs magnétiques dilués :  $(\text{Ga},\text{Mn})\text{N}$  avec Joël Cibert/Henri Mariette, et notamment l'aimant  $(\text{Ga},\text{Mn})\text{As}$  avec Laurens Molenkamp. Les propriétés sont déterminées par la structure de bandes de ces matériaux cristallins, dont les détails dépendent de l'aimantation. Obtenus par croissance par épitaxie par jets moléculaires, l'ordre cristallin de films minces est excellent et, du point de vue du magnétisme, le renversement d'aimantation est contrôlé par la nucléation de parois de domaines, et de très larges domaines magnétiques peuvent être stabilisés. En transport classique, les modifications locales de la densité d'états au niveau de Fermi, lors de la rotation d'aimantation, peuvent être observées avec une simple jonction tunnel. L'anisotropie de magnéto-résistance tunnel induit un comportement binaire, similaire à celui des jonctions tunnel magnétiques en électronique de spin, cependant avec un seul contact magnétique.

Comme chercheur CNRS au laboratoire LPN à Marcoussis, j'ai poursuivi les études de jonctions tunnel en étudiant le cas des diodes Zener-Esaki (effet tunnel interbandes), permettant de réaliser la spectroscopie en énergie de l'anisotropie de magnéto-résistance tunnel et démontrer les effets de bande [résultats non publiés]. J'ai surtout développé le projet « MesoSpin », avec l'idée d'étudier le transport quantique dans des nanostructures ferromagnétiques  $(\text{Ga},\text{Mn})\text{As}$ , matériau pour lequel la longueur de cohérence de phase est particulièrement grande (faible désordre, absence de magnons de basse énergie) par rapport à celle des métaux ferromagnétiques. L'idée simple était d'observer les effets de déphasage par précession rapide des spins dans un champ d'échange. Nous avons ainsi pu

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mettre en évidence les fluctuations universelles de conductance directement reliées à une texture magnétique, puis montré l'influence de la phase de Berry.

De 2010 à 2016, j'ai été mis à disposition à l'IFW Dresden, et j'ai dû donner une nouvelle orientation à ma recherche. Les effets de phase de Berry sur la structure de bande des matériaux trouvaient alors une nouvelle dynamique avec la découverte des isolants topologiques 3D. Mon activité a essentiellement concerné l'étude de nanostructures de  $\text{Bi}_2\text{Se}_3$  et  $\text{Bi}_2\text{Te}_3$ , et particulièrement les nanofils quantiques. Par des mesures de transport quantiques (oscillations Aharonov-Bohm, fluctuations de conductance), nous avons pu démontrer des propriétés uniques des quasi-particules de Dirac texturées de spin, comme la forte anisotropie de diffusion des porteurs en surface (diffusion à 2D), à l'origine de longueurs de cohérence phase relativement longues malgré un fort degré de désordre, ou encore le transport quasi-balistique en présence de confinement quantique (modes de surface quasi-1D).

Depuis Octobre 2016, j'ai rejoint le groupe SpinOrbitronics du laboratoire SPINTEC, et mon activité est désormais étendue aux nouveaux matériaux topologiques et magnétiques, en lien avec les matériaux 2D à liaisons van der Waals, d'intérêt pour la spintronique. Le magnétisme offre des opportunités pour générer et contrôler de nouveaux états quantiques texturés ou polarisés de spin. Le champ d'échange permet de modifier la structure de bande d'isolants topologiques non magnétiques, en créant une bande interdite pour les états de Dirac de surface. Surtout, en présence d'un réseau magnétique, la symétrie est modifiée et des nouveaux états quantiques peuvent être induits et observés par des mesures de transport (effet Hall quantique anormal, quantification de la réponse magnéto-électrique). Dans des nanostructures, les détails micromagnétiques (terminaisons de surface, textures) agissent ainsi directement sur les propriétés des conducteurs topologiques, avec la perspective de les contrôler en agissant sur l'aimantation.

Dans ce mémoire, je décris d'abord l'activité réalisée autour des nanostructures d'isolants topologiques 3D, et l'étude du désordre microscopique. En particulier, le régime de transport quasi-balistique des modes de surface dans un nanofil quantique est discuté en détail. Dans mon projet de recherche, focalisé sur le transport balistique par des modes chiraux, deux approches sont poursuivies : i) la réalisation d'un mode chiral par modification du mode topologique, parfaitement transmis, dans un nanofil  $(\text{Bi,Sb})\text{Te}$  [potentiel électro-chimique proche du point de Dirac, ie. en régime à faible nombre de canaux de conduction] ; ii) la réalisation de canaux de bords en régime d'effet Hall quantique anormal, en champ nul, dans une nouvelle classe de matériaux topologiques magnétiques, de la famille  $\text{MnBi}_2\text{Te}_4$  (et matériaux van der Waals similaires).

D'autres activités de recherche antérieures, non discutées dans ce manuscrit, concernent :

- $(\text{Ga,Mn})\text{As}$  au CNRS/LPN Marcoussis
- $\gamma\text{-Al}_2\text{O}_3/\text{SrTiO}_3$  à l'IFW Dresden



# Research activities



# 1

## Preamble

My research on 3D  $\mathbb{Z}_2$  topological insulators started in 2011, with a collaboration with Dr. Silke Hampel (head of the “Chemistry at the nanoscale” group at the IFW/IFF). As an alternative to the mechanical exfoliation of large single crystals, high-quality monocrystalline nanostructures of  $\text{Bi}_2\text{Se}_3$  and  $\text{Bi}_2\text{Te}_3$  were obtained by a vapor transport technique, without any catalyst, giving access to nanostructures of different shapes (platelets, nanoribbons, nanowires) with well-defined facets. These grow flat onto the amorphous surface of a  $\text{SiO}_x/\text{Si}$  substrate, with a random distribution and a low-enough surface density, so that an individual nanostructure can be electrically connected by e-beam lithography and metal evaporation/lift-off techniques.

Together with my colleague Dr. Joseph Dufouleur, we investigated the magneto-transport properties of individual nanostructures, and used quantum transport methods as an unambiguous “tool” to unveil the specific behavior of topological surface states, despite their coexistence with a large bulk-carrier density. As an important finding, we demonstrated the anisotropic scattering of 2D spin-helical Dirac fermions by a static disorder, and distinguished it from the isotropic scattering of massive quasi-particles in the bulk. Then, our most important contribution has been the detailed study of surface-mode transport in narrow nanowires, so as to understand the origin of quasi-ballistic transport in highly-disordered 3D topological insulator quantum wires. Due to quantum confinement, all pairs of modes are gapped, so that they lose, in theory, their topological protection. We gave evidence for the mechanism at work that preserves the quasi-1D spectral properties, for all modes, and we showed that both quantum confinement and anisotropic scattering lead to a unique situation for mesoscopic conductors : they behave like ballistic conductors despite a huge degree of disorder.

These studies were conducted in the “high-energy” regime, with a large number of populated modes. For a chemical potential close to the Dirac point, a flux-periodic topological transition is predicted, due to the specific contribution of a single mode with perfect transmission. This helical mode, if evidenced experimentally, could be driven into a spin chiral mode under a moderate transverse magnetic field. The perspectives foreseen from such a purely ballistic and truly 1D channel are described in the scientific project.



## 2

# Nanostructures of disordered 3D topological insulators

### 2.1 $\mathbb{Z}_2$ topological insulators

The influence of geometric phases on electronic band structures is known for long, but this research found a new impulse when  $\mathbb{Z}_2$  topological insulators were identified as a new topological state of matter in 2D in 2005 (graphene, HgTe quantum wells). This quantum spin Hall regime was rapidly confirmed experimentally in materials with a strong spin-orbit coupling, and the concept of topological band theory was soon extended to 3D insulators. Since then, these are intensively studied both theoretically and experimentally (see, for instance, [42–44] for early reviews and [45] for a review focusing on quantum transport in 3D topological insulators).

Many  $\mathbb{Z}_2$  topological insulators are small-gap semiconductors, for which the spin–orbit coupling is strong enough to induce a band inversion, such that the bulk energy gap closes at the interface with a topologically-trivial insulator. It results in the formation of gapless electronic states, which host Dirac fermions propagating at edges or surfaces, depending on the materials’ dimensionality (2D or 3D, respectively). Because of the strong spin–orbit coupling and time-reversal symmetry (TRS), these electronic states have a specific spin texture, with spin-momentum locking and a reversed chirality for opposite interfaces. Therefore,  $\mathbb{Z}_2$  topological insulators (TIs) have gapless states with a band structure made of spin-helical Dirac fermions.

Remarkably, the direct backscattering is forbidden by the spin texture.

In 2D TIs, the suppression of backscattering leads to the ballistic motion of charge carriers with opposite spin flows, a hallmark of the quantum spin Hall state. This was predicted for HgCdTe heterostructures [46, 47], and soon confirmed by experiments [48].

In strong 3D TIs, topological surface states are robust against any kind of nonmagnetic disorder, but the backscattering of these quasiparticles is restored by multiple-event scattering. Importantly, this results in a finite transport length (the backscattering length), limiting quasi-ballistic transport to narrow nanostructures only. In addition to having a finite residual bulk-carrier conductivity, this initially made the evidence of surface Dirac fermions by charge transport measurements difficult, and gapless topological surface states were actually first unveiled by photoemission spectroscopy studies.



In 2010, the evidence of Aharonov-Bohm oscillations in  $\text{Bi}_2\text{Se}_3$  nanoribbons gave the first robust evidence by electrical transport for the existence of metallic surface states [49], although it gave no indication on the exact nature of these quasiparticles. Yet, it became clear that quantum transport measurements could become a choice technique to reveal the physics of spin-helical Dirac fermions in simple geometries of mesoscopic conductors such as nanowires or nanoribbons (see [39] and refs. therein).

In 2013, we reported on a study of decoherence in  $\text{Bi}_2\text{Se}_3$  nanowires, based on the analysis of the temperature dependence of Aharonov-Bohm oscillations [27]. This shed some new light on the specific nature of metallic surface states, and revealed their weak coupling to the environment. This property of spin-helical Dirac fermions results from their spin texture, which is the key to understand the quasi-ballistic transport of surface modes in 3D TI quantum wires. Further investigations were then planned as :

- Studies of wide nanostructures (flakes or wide ribbons) were first considered [32], so as to investigate the transport properties of 2D surface states. We revealed their anisotropic scattering [33], giving a significant enhancement of the transport length with respect to the case of bulk massive quasiparticles. This set the clear condition to realize ballistic transport in narrow nanostructures.
- Studies of narrow nanostructures (nanowires or narrow ribbons) were then conducted, and specific properties were revealed by quantum coherent transport in 3D vector magnetic fields [34]. This gave us a comprehensive and quantitative understanding of the multi-mode surface transport regime and shed light on the origin of the quasi-ballistic properties despite a strong disorder [36].

## 2.2 Bulk-Surface carriers

$\text{Bi}_2\text{Se}_3$  is the canonical representative compound of stoichiometric materials realizing the simplest form of a strong 3D topological insulator, with a single Dirac cone at the center of the Brillouin zone. This family of materials gave many opportunities to modify both the bulk band gap and the chemical potential, as done in  $(\text{Bi,Sb})(\text{Se,Te})$  compounds [50, 51]. Yet, a significant drawback of chalcogenides is the difficulty to achieve charge compensation, since charged defects can be massively created during the growth (especially anion vacancies, which are thermodynamically stable and have a small energy cost).

Due to a large residual bulk doping, the study of surface-state transport in disordered 3D topological insulators is therefore not straightforward. Considering the case of a highly-degenerate semiconductor such as  $\text{Bi}_2\text{Se}_3$ , with a bulk carrier density in the  $10^{19}\text{cm}^{-3}$  range (which roughly corresponds to about 1% of Se vacancies, acting as double donors), the crossover thickness  $t_c$  for which the surface conductance becomes comparable to the bulk conductance is roughly given by  $t_c = 2(n_s\mu_s)/(n_b\mu_b)$ . For a strong disorder typical for  $\text{Bi}_2\text{Se}_3$ , common to both surface and bulk states, the mobility of topological surface states is about one order of magnitude larger than that of bulk states (enhancement due to the anisotropic scattering of spin-helical Dirac fermions). Taking band bending into account [32], typical values for the surface and bulk carrier densities are  $n_s = 5 \times 10^{12}\text{cm}^{-2}$  and  $n_b = 5 \times 10^{19}\text{cm}^{-3}$ . This gives a value  $t_c = 20$  nm. Based on a realistic approximation (ignoring details of band bending due to a very short Thomas-Fermi screening length  $\lambda_{\text{TF}} \approx 3$  nm), this lower bound for  $t_c$  clearly shows that the control of topological surface-state transport in disordered 3D TIs requires to use thin nanostructures.

As most 3D topological insulators are van der Waals-layered materials, the first method used to prepare thin films has been the exfoliation technique, based on scotch tape, as routinely used to produce graphene. This method has the advantage to give access to many macroscopic crystals, including ternary or quaternary compounds, some of which being more or less charge compensated. Using a back-gate voltage, it becomes possible to tune the electro-chemical potential close to the Dirac point and deplete the bulk carriers. Due to the efficient screening of topological surface states, dual-gate devices were however needed to investigate the two-interface quantum Hall regime in BSTS nanoflakes [52]. Yet, the mechanical exfoliation also has some drawback. Firstly, it can result in severe damages and the surface is often dirty, with a much worse quality than that achieved from cleaved crystals. Secondly, the information on the crystalline orientation is usually lost. This is not really important for simple topological insulators such as  $\text{Bi}_2\text{Se}_3$ , with a single Dirac point in the center of the Brillouin zone, but it can be for materials with more complex topological invariants (with only some surfaces supporting topological states).

Instead, single-crystalline nanostructures were grown by different bottom-up methods, each technique having its pros and cons. Ultra-thin layers prepared by molecular-beam epitaxy are well-adapted to tune the interface band structure by electrical fields, but films thicker than 20 nm tend to grow 3D, with many defects and grain boundaries. High-quality single-crystalline nanostructures with large aspect ratios can be grown by catalyst-assisted molecular-beam epitaxy or chemical vapor deposition, as well as by catalyst-free vapor transport, but ultra-thin structures are scarce. Very narrow quantum wires can also be prepared by electrodeposition in membranes, presently with some intrinsic limitations due to diameter fluctuations in ultimate nanomembranes and grains. In all cases, individual nanostructures can be randomly grown on or transferred onto  $\text{SiO}_x/\text{Si}$  substrates, so that an electric field can be created at the bottom interface by a backgate voltage.

## 2.3 Growth of $\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Te}_3$ by vapor transport

To obtain some nanostructures with a high crystalline quality and pristine surfaces, we have developed a new process to optimize the growth of  $\text{Bi}_2\text{Se}_3$  nanostructures by vapor transport. As sketched in Fig. 2.1, a sealed silica ampulla is used as an inert reactor in a home-built oven with a thermal gradient, so as to sublime the source material ( $T_2 \approx 500 - 600^\circ\text{C}$ ) and to transport the molecular flux ( $\text{BiSe}$  and  $\text{Se}_2$ ) towards a lower-temperature region ( $T_1 \approx 430 - 460^\circ\text{C}$ ) where elements recrystallize on the amorphous surface of the substrate. This method has a couple of advantages when compared to other techniques (electrodeposition, molecular beam epitaxy, Au catalyst-assisted chemical vapor deposition)[30]

- High-quality monocrystalline nanostructures are obtained with atomically flat facets ;
- There is no contamination from a catalyst and, most important, the growth proceeds within the substrate plane [contrary to Au-assisted chemical vapor deposition, which results in a “forest” of tilted nanostructures]. No further processing is needed, such as a transfer step ;
- Different shapes (large nanoflakes, long nanotibbons or nanowires) give access to a wide range of aspect ratios [suitable to investigate the physics of surface Dirac fermions with or without lateral quantum confinement] ;

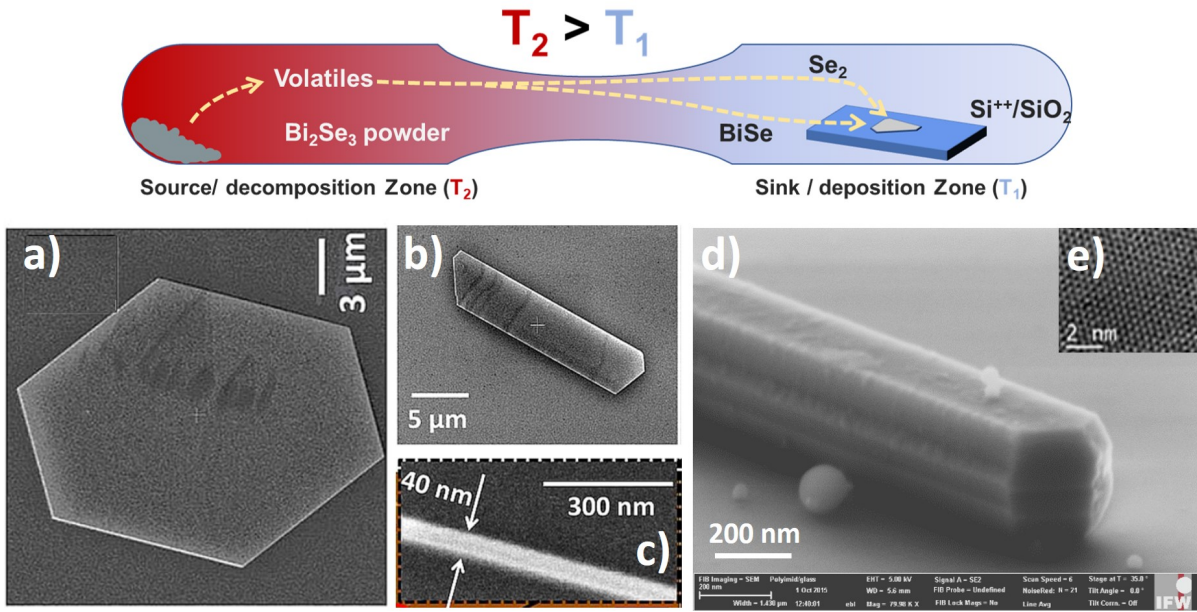


FIGURE 2.1 – Schematic of the principle of the vapor transport growth for  $\text{Bi}_2\text{Se}_3$  pure crystals. Thin nanostructures are obtained, such as, a) nanoflakes, b) nanoribbons, and c,d) nanowires, all with a high crystalline quality (as revealed by transmission electron microscopy images, e).

- The thickness can be as small as a couple of nm, whereas the nanostructures can be of hundreds of micron long, with a width that can be as large as tens of micron or as small as about 40 nm.

The high quality of as-grown nanostructures was confirmed by transmission electron microscopy including selected area electron diffraction, as well as by electrical transport measurements, and even high-resolution scanning electron microscope images revealed the surface facets (see Fig. 2.1d).

Still, two important limitations remain to date :

- Thicknesses below 20 nm are scarce ;
- The growth was mostly limited to binary compounds (dirty-metal regime).

Like for other groups, our studies of 3D TI nanowires were therefore limited to the “high-energy” regime, with the Fermi energy shifted some tens to hundreds of meV from the Dirac point. However, for quantum transport measurements, the residual bulk conductivity is not an issue, particularly in nanowires that have a well-defined cross section and periodic properties related to topological surface states only. Still, this limitation calls for new developments, so as to grow ternary compounds for instance, that are necessary to consider studies of the “low-energy” regime, as discussed in the research project.

## 2D topological surface states

### 3.1 Introduction

Single-crystalline 3D topological insulators host a 2D electron gas of massless Dirac fermions at their surfaces/interfaces with other materials. With their helical spin texture, these quasi-particles are different from those in graphene, and 3D TIs also offer an interesting alternative to semiconducting heterostructures with massive 2D fermions in a charge-accumulation 2D electron gas, which often requires a more complex stacking of epitaxial layers of materials with different energy gaps and electronic-band offsets.

In practice, the control of topological surface states in a 3D topological insulator also requires that of band bending at interfaces (due to charge transfer from the environment) and an important limitation to investigate topological surface-state transport actually came from that most materials are not true bulk insulators.

Moreover, in order to understand the physics of 3D TI quantum wires, it is first necessary to take a closer look at the scattering of 2D spin-helical Dirac fermions by a non-magnetic disorder. For a surface, the backscattering of quasi-particles is not suppressed since it remains possible by successive small-angle scattering events, over a scale given by the transport length,  $l_{tr}$ . Still, the spin texture favors forward scattering, so that the transport length can be much longer than the disorder correlation length (contrary Dirac fermions in graphene). As a consequence, the condition for ballistic transport can already be realized in nanostructures with dimensions close to the micron scale, and quantum devices with a simple geometry can be built from individual nanowires.

In our work, we mostly investigated the charge transport properties of  $\text{Bi}_2\text{Se}_3$  and  $\text{Bi}_2\text{Te}_3$  nanostructures, grown by vapor transport on  $\text{SiO}_x/\text{Si}$  substrates. These binary 3D TIs have a large residual bulk conductivity (with a very small bulk-carrier mobility, but a huge carrier density), so that the total conductance of thick nanostructures is dominated by bulk transport. This contribution is slightly reduced in  $\text{Bi}_2\text{Te}_3$ , but only high-energy surface quasi-particles can be studied because the Dirac point lies deep into the valence band, whereas the Fermi level is pinned near or above the top of the valence band. In  $\text{Bi}_2\text{Se}_3$ , the Dirac point lies within the bulk band gap, but the Fermi level is high above the bottom of the conduction band (due to too many Se vacancies), so that it can only be modified over a small energy range by an electrical gate (efficient electrostatic screening). Based on magneto-transport measurements in high magnetic fields, we investigated the

Shubnikov-de Haas oscillations of patterned micro-Hall bars in detail. Using an electrical back gate, it was possible to separate all three contributions to charge transport and reveal band bendings effects. Transconductance measurements gave further information, from which the transport length was inferred. The results revealed the anisotropic scattering of topological surface states, and explained the rather long phase-coherence length. Most importantly, the determination of  $l_{tr}$  confirmed that a crossover from diffusive to ballistic transport occurs at a lengthscale of hundreds of nm, in typical disordered 3D topological insulators. Reducing the degree of disorder in optimized materials would give ballistic transport in 2D, since the transport length could largely exceed the micron scale.

### 3.2 Band bending and interface charge transfers

In a degenerate semiconductor, electronic bands are flat in the bulk, but they bend over a short distance at interfaces (gap, extra charges). Because of the existence of gapless surface states, an upward band bending naturally occurs in a 3D topological insulator, due to a partial charge transfer from the bulk to interfaces. Therefore, the bulk-carrier density is lower at the vicinity of the surface states over a length scale given by the Thomas–Fermi screening length  $\lambda_{TF}$  (a couple of nm)[32, 33], and a depletion region could even exist if the chemical potential in the bulk is reduced. Using this band bending effect is indeed a very efficient way to deplete very thin flakes of 3D TIs (see [53], for instance).

We systematically studied the band bending in different nanostructures of  $\text{Bi}_2\text{Se}_3$  at low temperatures and in large magnetic fields ( $\mu_0 H$  up to 55 T) [32]. Using the electrical backgate dependence of the oscillations and considering the effective mass inferred from the temperature dependence of the Shubnikov–de Haas oscillations, it was possible to assign the different frequencies observed in the Fourier transform to the different charge carriers populations (see Fig. 3.1). This allowed us to determine the band bending in 3D TI nanostructures for a bulk charge density  $n_b$  ranging from  $n_b = 2.6 \times 10^{19} \text{cm}^{-3}$  down to  $n_b = 6.6 \times 10^{17} \text{cm}^{-3}$ . Two representative cases are discussed below.

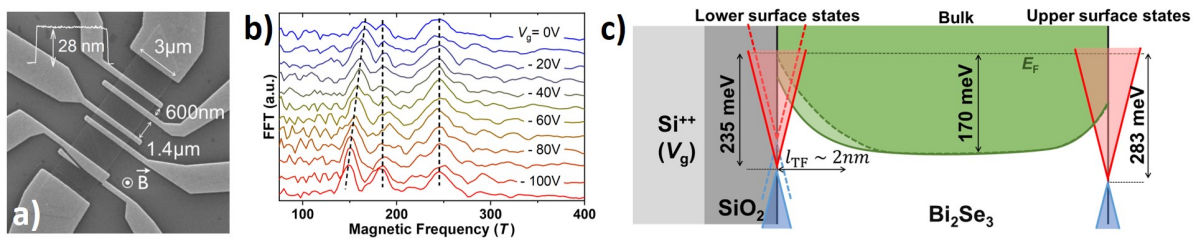


FIGURE 3.1 – (a) Scanning-electron microscope image of a highly-doped  $\text{Bi}_2\text{Se}_3$  nanoribbon, (b) fast-Fourier transforms of the longitudinal magneto-resistance, for different back-gate voltages, and (c) Evolution of the electronic band profile, with typical upward band bending at interfaces.

For a highly-doped nanoribbon ( $n_b = 1.4 \times 10^{19} \text{cm}^{-3}$ ;  $t = 28 \text{nm}$ ), an upward band bending was observed (Fig. 3.1c), as the result of a dominant charge transfer from the bulk. Still, the degree of band bending depends on the nature of the interface, which is different for the surface (native oxide) and for the interface with the substrate. This shows that some extra charge transfer comes from some local charges at the interfaces.

For a low-doped nanoribbon ( $n_b = 6.6 \times 10^{17} \text{cm}^{-3}$ ;  $t = 80 \text{ nm}$ ), band bending is controlled by the electrical doping at interfaces. A very strong downward band bending (of about 260 meV) was even evidenced at the bottom interface (see Fig. 3.2). Such a strong effect leads to the formation of a 2D electron gas that coexists with topological interface states, with two bands filled. These bands correspond well to massive quasiparticles confined in a triangular potential.

Therefore, the downward band bending points to the necessity of controlling both the residual bulk doping *and* the additional surface/interface doping to produce bulk-compensated structures. In particular, the very strong downward band bending near the interface with the substrate suggests that the interface with  $\text{SiO}_2$  can favor a strong interface doping, which could result either from a locally increased disorder in  $\text{Bi}_2\text{Se}_3$  or from dangling bonds.

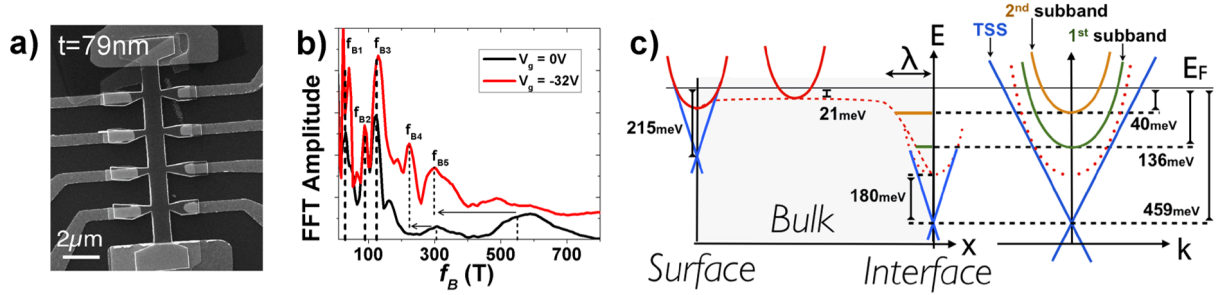


FIGURE 3.2 – (a) Scanning-electron microscope image of a Hall-bar patterned low-doped  $\text{Bi}_2\text{Se}_3$  nanoribbon, (b) fast-Fourier transform of the longitudinal magneto-resistance for two different back-gate voltages, and (c) evolution of the electronic band profile from the top surface to the bottom interface, typical for a low bulk-carrier density (downward band bending) and a large interface carrier density (coexistence of topological states with a Rashba 2DEG).

Important results are summarized as follows :

- Due to the large residual bulk density and the pinning of the Fermi energy in the conduction or valence band of materials with a large dielectric constant, the bulk contribution to the conductance is never negligible ;
- Besides, it usually controls the upward band bending at interfaces of the topological insulator (charge transfer of bulk carriers to empty gapless topological surface states) ;
- Downward band bending can, however, exist if a massive charge transfer from another origin is also present (surface/interface disorder, surface adsorbents, or electrostatic gate). Such a situation is more likely to happen for materials with a small bulk carrier density ;
- If the surface/interface density is very large (typically for  $n_s > 10^{13} \text{cm}^{-2}$ ), a Rashba charge-accumulation 2DEG of massive quasi-particles coexist with Dirac topological states.

### 3.3 Anisotropic scattering of topological surface Dirac fermions

Further important information on the scattering of spin-helical Dirac fermions by a non-magnetic disorder can be obtained from transconductance  $G(V_G)$  measurements [33]. It is indeed important to distinguish between two different scattering times and, accordingly, between two different length scales (see Fig. 3.3a) :

- The quantum lifetime of quasi-particles can be inferred from Shubnikov-de Hass measurements. It is associated with the mean-free path  $l_e$  between two successive scattering centers (thus to the microscopic disorder correlation length) and this momentum scattering time relates to the *quantum mobility* ;
- The transport time of carriers corresponds to the timescale for momentum backscattering, over a length called the transport length  $l_{tr}$ , related to the *transport mobility*, which determines the classical conductance. Whereas direct backscattering is forbidden by spin–momentum locking (giving dissipationless states in a 2D TI), it is allowed by multiple scattering processes in a disordered 3D TI, resulting in a finite transport length and a large  $l_{tr}/\xi$  ratio (Fig.3.4c).

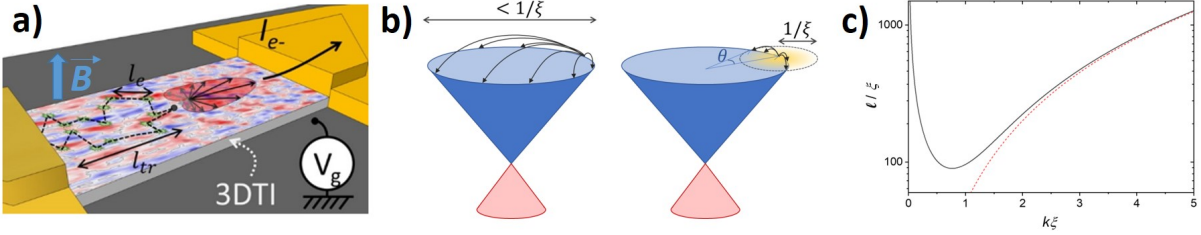


FIGURE 3.3 – (a) Anisotropic scattering by disorder. The backscattering transport length  $l_{tr}$  can be much longer than the mean-free path  $l_e$  ; (b) Schematic of the isotropic (left) and anisotropic (right) scattering by a disorder potential with a correlation length  $\xi$  ; (c) Dependence of the ratio  $l_{tr}/\xi$  with  $k\xi$  for a disorder strength  $g = 0.1$ , with a minimum value at about  $k\xi \approx 1$ . The red dashed line is the long-range disorder limit ( $k\xi \gg 1$ ) where  $l_{tr}/\xi \propto k^2\xi^2$ .

Using a thin  $\text{Bi}_2\text{Se}_3$  nanoribbon (Fig.3.4a), the backgate voltage allowed us to modify the upward band bending, induced by a rather large bulk carrier density, at the bottom interface (“lower surface state”). All microscopic transport parameters could be inferred from quantum magnetotransport (Fig.3.4b) and trans-conductance (Fig.3.4c) measurements [34]. Contrary to bulk carriers, for which  $l_e \approx l_{tr}$  is given by the disorder correlation length  $\xi$  (isotropic scattering due to a short-range disorder potential), the transport length of both upper and lower topological surface states was found to be much larger than the mean-free path, despite a limitation at about 200 nm probably due to the finite coupling with bulk carriers [larger values are expected for decoupled topological states and/or in materials with a larger disorder correlation length, such as  $\text{Bi}_2\text{Te}_3$ ].

The average number of scattering centers involved in a backscattering process is directly related to the ratio  $l_{tr}/l_e$ , and it can give some important information on the nature of both the scattering potential and the quasi-particles. Importantly, we revealed the long-range nature of disorder for spin-helical surface Dirac fermions, due to efficient electrostatic screening and spin-momentum locking, which results in strongly anisotropic scattering.

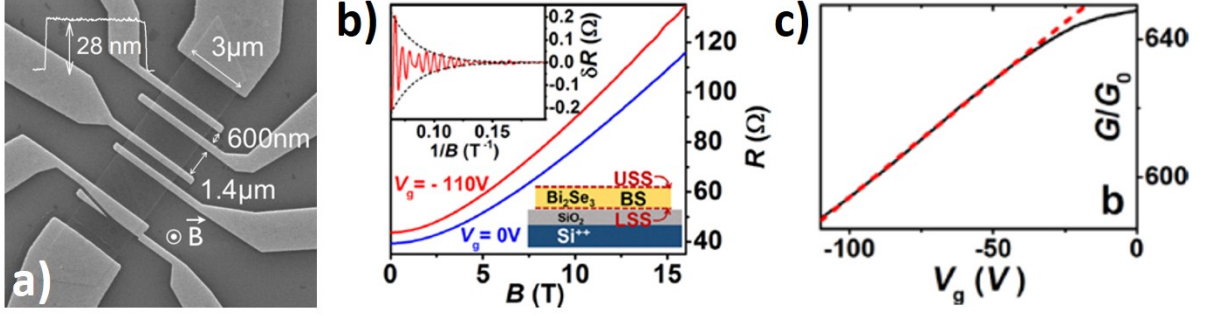


FIGURE 3.4 – (a) Scanning electron microscope image of a thin  $\text{Bi}_2\text{Se}_3$  nanoribbon with traversing ohmic contacts ; (b) Magnetoresistance with a perpendicular applied field, for two gate voltages. Upper inset : SdH oscillations after subtraction of the classical resistance background ; Lower inset : Schematic of the device (c) Backgate voltage dependence of the transconductance and linear fit from which the transport length of the lower surface states (bottom interface) is inferred.

This means that forward scattering is favored for such quasi-particles and that the transport length is strongly enhanced ( $l_{\text{tr}} \gg l_e$ ), as predicted by theory [54] and confirmed by our trans-conductance measurements [34]. Such a situation never happens in other materials within which charge carriers directly coexist with disorder (including Dirac fermions in graphene), where the scattering of quasi-particles by any kind of disorder is isotropic (that is,  $l_{\text{tr}} \approx l_e$ ). Only high-mobility AlGaAs 2DEGs can realize such a situation of anisotropic scattering. In this case, however, this is due to the spatial separation of free carriers and local ionized donors, and it is not possible to vary the degree of disorder over a wide range, so that there is little room to investigate quasi-ballistic transport (in other words, the transition from ballistic to diffusive transport is rather abrupt and happens already when a small amount of impurities are introduced in the system). This is not the case for spin-helical Dirac fermions, and this property is at the origin of their unique transport properties, particularly in nanostructures, with an extended range of parameters to study quasi-ballistic transport, and therefore the ballistic-to-diffusive crossover in mesoscopic conductors.

This enhanced transport length for topological surface states is also important for spin transport studies, as it gives a lower limit for the size of functional spintronic devices making use of the spin-momentum locking property. Indeed, due to the strong spin-orbit coupling, there is a direct correspondence between the momentum-backscattering transport length and the spin relaxation length. With wide  $\text{Bi}_2\text{Se}_3$  nanostructures, this scale is rather short ( $l_{\text{tr}} \approx 200$  nm) [33], so that, for instance, lateral spin valves could only be realized in the short-junction limit. This also shows that the true potential for the spin-to-charge conversion in highly-disordered  $\text{Bi}_2\text{Se}_3$  could still give an improvement in the conversion efficiency by two orders of magnitude with respect to state-of-the-art records, with an inverse Edelstein length  $l_{\text{IEE}}$  determined by the intrinsic transport length of the 3D topological insulator, whereas it presently remains limited by the spin/momentum relaxation below metallic ohmic contacts (with  $l_{\text{IEE}} \approx 2$  nm) [55].



### 3.4 Dimensionalities of charge transport

The enhancement of the transport length for topological surface states also has two fundamental consequences for the quantum transport properties of 3D TI nanostructures and the dimensionality of surface charge transport :

- The phase coherence length of TSS is also enhanced in the same ratio for diffusive 2D surface states in nanoplatelets or wide nanoribbons, so that mesoscopic transport can be studied in rather wide and long conductors (well beyond the micron size), despite relatively strong disorder. Since  $L_\varphi \gg l_{\text{tr}}$  ( $L_\varphi$  being determined by inelastic scattering), quantum corrections to the conductance due to diffusive phase-coherent transport can thus be revealed by magneto-transport measurements with magnetic fields as small as 100 mT ;
- The condition for ballistic transport in the transverse motion of surface carriers along the perimeter is more restrictive ( $L_p < 2l_{\text{tr}}$ ). Still, this corresponds to rather long values ( $L_p \lesssim 500$  nm), and therefore ballistic nanostructures can have a rather large cross section ( $S \approx 0.2 \mu\text{m}^2$ ). Contrary to the case of carbon nanotubes, magnetic flux-dependent periodic phenomena in these quantum wires (such as the Aharonov-Bohm interference) can therefore be studied in rather small fields, with periods in the range of some hundreds of mT to a couple of Tesla.

In the case of highly-disordered  $\text{Bi}_2\text{Se}_3$  nanostructures ( $l_e \approx 30$  nm), some large values of  $l_{\text{tr}}$  ( $>200$  nm) [33] and of  $L_\varphi$  ( $>2 \mu\text{m}$ ) [27] were found. This implies that the dimensionality of surface-state transport is reduced in narrow nanostructures (mostly nanowires) and that their band structure is modified due to quantum confinement (which even further reduces the scattering by disorder). It thus becomes important to distinguish between three different situations for the dimensionality of charge transport :

- No quantum confinement [ $l_{\text{tr}}$  is shorter than every dimensions]. Surface-state transport is diffusive and quasi-particles are 2D spin-helical Dirac fermions with a continuous spin-helical Dirac-cone band structure ;
- Transverse quantum confinement [the perimeter  $L_p$  becomes shorter than  $2 l_{\text{tr}}$ ]. Surface transport is quasi-ballistic if the distance  $L$  between contacts is longer than  $l_{\text{tr}}$  and trully ballistic otherwise. Even if the length  $L$  becomes much longer than  $l_{\text{tr}}$ , surface modes are quasi-1D channels in such long nanowires (becoming truly 1D only when they close), due to the minimum value of  $l_{\text{tr}}$  (maximum energy broadening in the 1D spectrum, independent of  $L$ ) ;
- Full quantum confinement [all dimensions are shorter than  $l_{\text{tr}}$ ]. Spin-helical Dirac fermions are fully localized in a short nanowire, which becomes a 0D quantum dot.

We remark that the dimensionality of quantum coherent transport is another quantity determined by comparing the dimensions of a mesoscopic conductor to the phase coherent length  $L_\varphi$ . Since  $L_\varphi$  is longer than  $l_{\text{tr}}$ , nanoribbons with a width  $W$ , such as  $l_{\text{tr}} < W < L_\varphi$ , have 2D spin-helical surface Dirac fermions but quantum coherent transport is 1D (which modifies the self-averaging of quantum interference in long conductors, for which the length  $L$  is longer than  $L_\varphi$ ).

# Quasi-1D topological surface modes

Signatures of the quasi-ballistic transport of topological surface states in 3D TI quantum wires can be revealed by the study of quantum corrections to the conductance. In a bulky nanostructure, spin-helical Dirac fermions propagate on the surface in a hollow-type electrical geometry, in analogy to carbon nanotubes (ballistic transport) or to the Sharvin-Sharvin metallic tubes (diffusive transport). Phase-coherent transport in the transverse direction (along the perimeter of the nanowire) thus gives rise to periodic Aharonov-Bohm oscillations in the longitudinal magnetoconductance, determined by a well-defined cross section  $S = \frac{L_p^2}{4\pi}$ , and their observation in wide  $\text{Bi}_2\text{Se}_3$  nanoribbons gave the first robust evidence of surface states by transport measurements [49]. Their topological nature was then confirmed by our study of decoherence at very low temperatures in narrow (quantum) nanowires [27], which revealed the unusual weak coupling to the environment and the ballistic motion in the *transverse direction*. Later, we investigated phase-coherent transport in the *longitudinal direction* in a study of conductance fluctuations [34], which revealed the subtle influence of disorder in the quasi-ballistic regime, leading to a non-universal behavior of quantum interference. A detailed understanding of the propagation of spin-helical 1D surface modes in presence of a Gaussian disorder showed that both the spin texture of Dirac fermions and their quantum confinement are responsible for the weak scattering by disorder [36], thus resulting in a weak coupling between quantum states. This is a necessary condition for their manipulation by radio-frequency fields, as well as for the study of specific properties related to a single topologically-protected low-energy mode, such as 1D chiral edge states or Majorana bound states [56].

## 4.1 Quantum confinement : 1D Dirac spectrum

In quantum wires ( $L_p < 2l_{\text{tr}}$ ), the surface-state band structure is modified by periodic boundary conditions imposed in the transverse direction of the nanostructure, leading to the quantization of the transverse momentum  $k_{\perp}$  (Fig. 4.1b). This situation is equivalent to the energy quantization of quasi-particles confined into a quantum well with infinitely-high potential barriers, and for Dirac fermions, the transverse energy becomes quantized with a constant energy-level spacing  $\Delta = \hbar v_F/L_p$  between successive transverse modes. Due to the winding of the wave function along the perimeter (curvature) and to the spin-momentum locking of helical Dirac fermions, an additional  $\pi$  Berry phase suppresses the topological protection of all energy modes in zero magnetic field (pairs of gapped states).

However, if a magnetic field is applied along the nanowire axis, the Aharonov-Bohm flux modifies the periodic boundary condition (which gives an overall shift of transverse quantization planes, thus tuning the energy spectrum). Importantly, this flux dependence can restore the topological protection periodically when the Aharonov-Bohm phase compensates the curvature-induced Berry phase, giving rise to a single gapless and linear mode with perfect transmission (Fig. 4.1c,  $\Phi = 1/2\Phi_0$ ), independent of disorder [57].

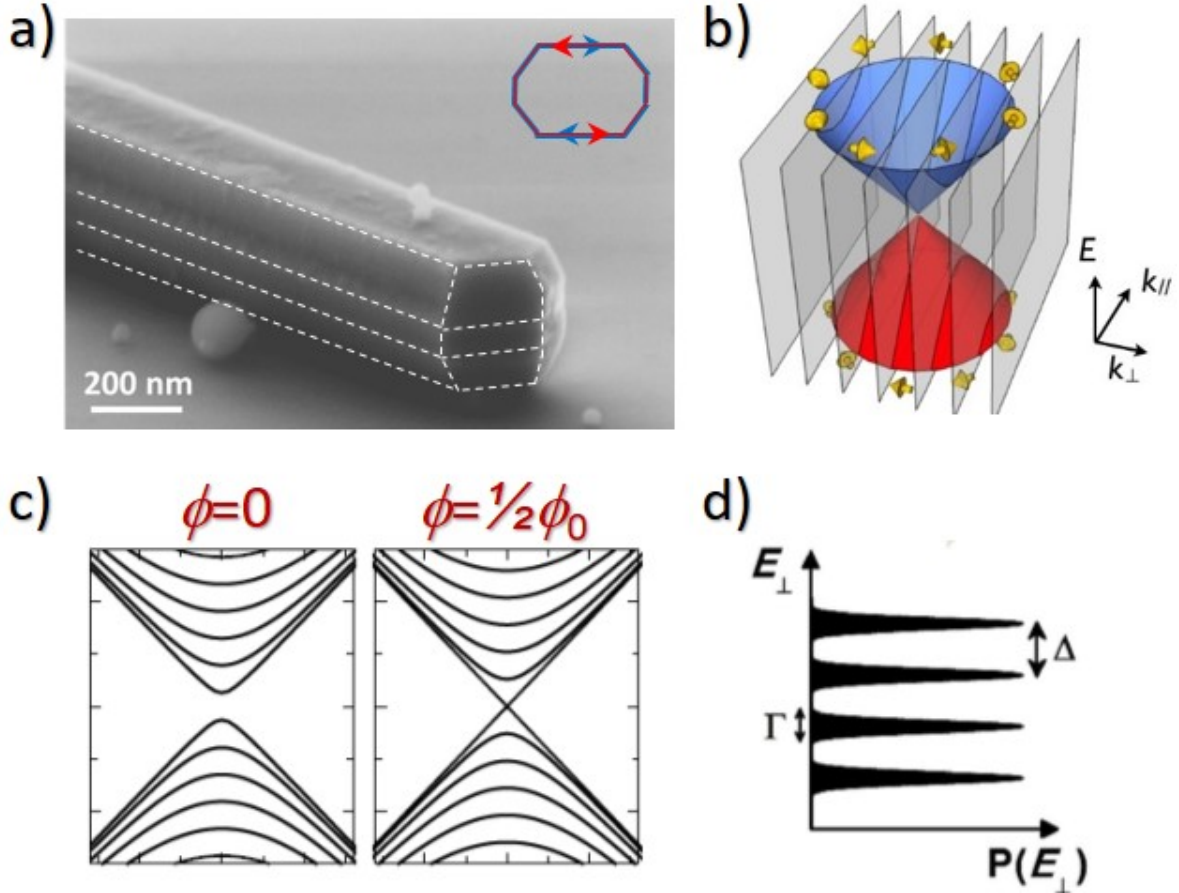


FIGURE 4.1 – (a) Scanning electron microscope image of a narrow Bi<sub>2</sub>Se<sub>3</sub> nanowire, with an electrical perimeter  $L_p = 300$  nm. Dashed lines separate different facets. Inset : schematics of the cross section and coherent winding of topological surface states; (b) Transverse-impulse quantization planes intersecting the spin-helical Dirac cone. By applying a magnetic flux, all planes are continuously shifted in the  $k_{\perp}$  direction; (c) Band structure for two values of the magnetic flux  $\Phi$ . For  $\Phi = 0$ , quantum confinement gives pairs of modes with a finite energy gap  $\Delta$ . For  $\Phi = 1/2\Phi_0$ , a single topological mode with linear dispersion appears (quantization plane intersecting the Dirac point); (d) Energy broadening  $\Gamma$  of the quantized transverse energy due to disorder [for 3D TI quantum wires,  $\Gamma$  is smaller than  $\Delta$ , even for relatively strong disorder].

In recent years, a couple of interesting studies suggested the influence of such a topological mode on quantum transport properties, particularly the Aharonov-Bohm (AB) oscillations [58–60]. These results raised some important questions since it was not possible to give a quantitative interpretation of the physical phenomena observed (Aharonov-Bohm

oscillations and non-universal conductance fluctuations) solely based on the contribution of this perfectly transmitted mode to the conductance. In particular, the *amplitude* of these quantum corrections to the conductance was always found much smaller than the conductance quantum  $G_0$ . It thus remained unclear whether these properties were a signature of a topological transition or were rather induced by all spin-textured modes, including dominant contributions from high-energy quasi-1D modes.

Actually, the quantum magneto-conductance is mostly due to a limited number of modes, those partially-opened modes with a quantized transverse energy close to the Fermi energy. Since most studies were conducted in the large- $N$  limit ( $E_F \gg \Delta$ ),  $N = \lfloor 2E_F/\Delta \rfloor$  being the number of occupied modes, the relative contribution of the topological surface mode is rather small. A full quantitative understanding required to describe the energy dependence of the transmissions for all surface modes, considering both disorder and interfaces with metallic contacts (see section 4.4 for details). In particular, the scattering of surface modes by disorder results in the energy broadening  $\Gamma$  of quantized modes (Fig. 4.1d), related to the transport length.

We evidenced that the quasi-ballistic regime is closely related to the condition  $\Gamma \ll \Delta$ , which is satisfied over an unusual broad parameters range (disorder strength, energy) in disordered 3D topological insulator nanostructures [36].

## 4.2 Quantum coherence I : Aharonov-Bohm oscillations

The quantum coherent transport of topological surface states in the transverse direction of a 3D TI nanostructure results in conductance oscillations when a longitudinal magnetic induction  $B_{\parallel}$  is applied (hence a magnetic flux  $\Phi = B_{\parallel} \times S^{\text{el}}$ , where  $S^{\text{el}}$  is the effective electrical cross section of metallic surface states). This is due to the flux-periodic evolution of the Aharonov-Bohm quantum interference giving successive conductance maxima (constructive interference) and minima (destructive interference).

Because the phase coherence length can be as large as a couple of microns (at very low temperatures), two different situations must be considered for coherent transport in the transverse motion of surface states :

- When  $L_{\varphi} \approx L_p/2$ , clear periodic oscillations of the conductance are directly visible in the longitudinal magneto-conductance  $G(B_{\parallel})$ . In this case (wide nanoribbons), only the fundamental- harmonic Aharonov-Bohm interference modifies the conductance, a behavior which already reveals that the phase averaging due to disorder is not efficient, despite a high point-defect density in most 3D TI materials.
- When  $L_{\varphi} \gg L_p$ , that is, either at very low temperatures or for short-perimeter nanowires (quantum wires), the periodic AB behavior is usually hidden in complex  $G(B_{\parallel})$  traces. This is due to the multiple-harmonic contributions to the transverse quantum interference (related to the multiple winding of coherent trajectories along the perimeter), and to the influence of disorder (phase shifts). Nevertheless, the periodic behavior can be revealed by a Fourier transform analysis.

### 4.2.1 Case of wide nanoribbons

We consider the long-perimeter limit, with  $L_\varphi \approx L_p/2$ .

For wide nanoribbons, periodic Aharonov-Bohm oscillations are directly visible in  $G(B_{\parallel})$  traces, with a rather small amplitude typical for the large- $N$  limit in a mesoscopic conductor, where  $N$  is the number of populated transverse modes, since  $\Delta = \hbar v_F/L_p$  is typically much smaller than  $E_F$ . As seen in Fig.4.2 for a wide  $\text{Bi}_2\text{Te}_3$  nanoribbon ( $E_F \approx 120$  meV;  $L_p = 940$  nm;  $\Delta = 2$  meV), the Aharonov-Bohm period,  $\delta B_{\text{AB}} = 150$  mT, directly relates to the electrical cross section of the nanostructure, with a value being slightly smaller than that given by its physical dimensions (the topological surface states being “buried” below a thin native oxide layer, typically 5 nm thick). The fast-Fourier transform of the  $G(B_{\parallel})$  trace thus gives a single peak at the AB frequency.

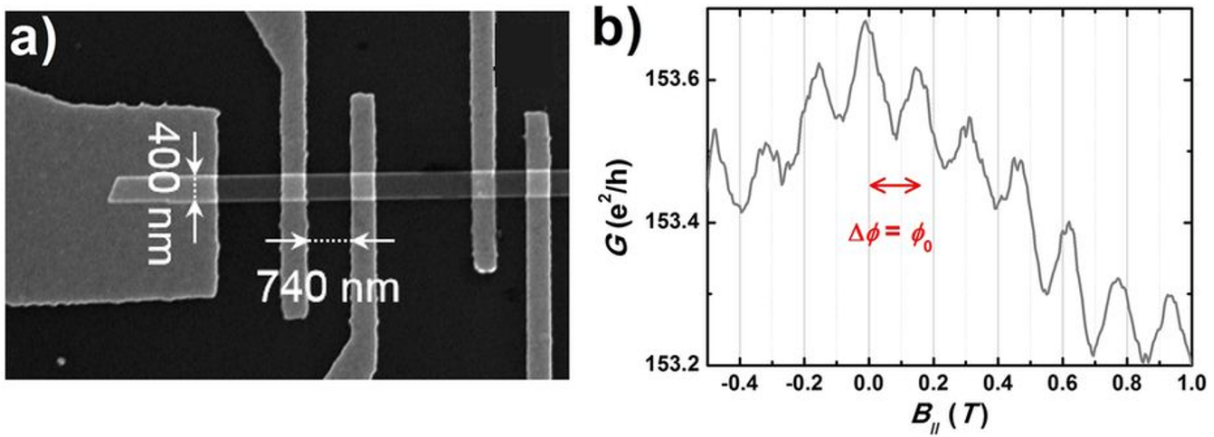


FIGURE 4.2 – (a) Scanning electron microscope image of a  $\text{Bi}_2\text{Te}_3$  nanowire with a rather large perimeter  $L_p = 940$  nm (width  $w = 400$  nm, height  $t = 70$  nm) and ohmic CrAu contacts; (b) Aharonov-Bohm periodic oscillations (fundamental harmonics), with a period  $\delta B_{\text{AB}}$  that directly relates to the nanowire’s electrical cross section ( $\delta B_{\text{AB}} = \Phi_0/S^{\text{el}}$ ).

According to theory, the overall phase shift of this sine evolution due to the AB quantum interference depends on both the degree of disorder and the energy of Dirac quasiparticles [57]. In most cases, the Fermi energy is very large and AB oscillations are phase locked with a conductance maximum in zero magnetic field, as found in many experiments and confirmed by theory. Yet, theory predicts the opposite situation (conductance minimum for a zero magnetic flux) when the chemical potential is near the Dirac point. The overall energy dependence of this phase shift can be quantitatively obtained from models taking explicitly disorder into account, and it allowed us to reveal an oscillatory behavior that is directly related to quantum confinement (see 4.4). For lower temperatures (longer  $L_\varphi$ ) or for narrower nanoribbons, roughly when  $L_\varphi \approx L_p$ , additional Altshuler-Aronov-Spivak (AAS) oscillations develop in addition. These correspond to quantum interference related to the complete winding of coherent paths along the perimeter, with time-reversed coherent loops so that this contribution is never damped by disorder, which is the usual situation found in (diffusive) mesoscopic conductors.

### 4.2.2 Case of narrow nanowires : quantum confinement

We consider the short-perimeter limit, with  $L_p < 2l_{tr} \ll L_\varphi$ .

For narrow nanostructures, the conductance modulation due to both AB and AAS interferences results from a complex mixing of high-order harmonics (multiple windings of coherent loops), with harmonic-dependent phase shifts induced by disorder and varying relative amplitudes due to quasi-ballistic transport. The periodic-flux dependence of the longitudinal magneto-conductance is therefore hardly visible in most  $G(B_{\parallel})$  traces, though it can still be when low-order harmonics remain dominant (as shown in Fig.4.3b).

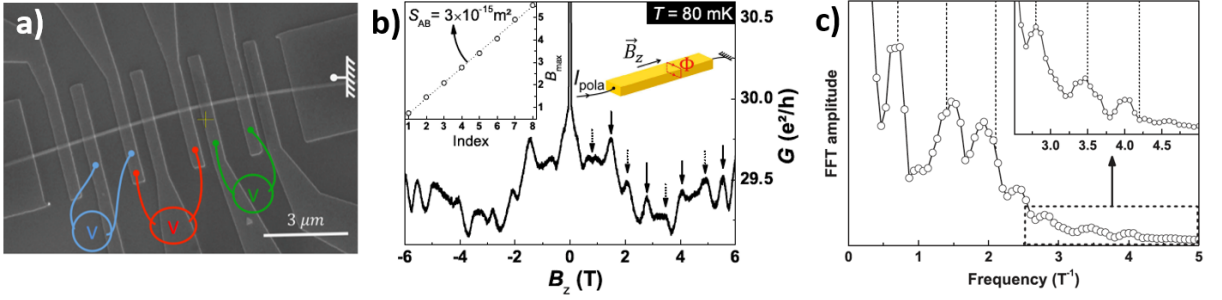


FIGURE 4.3 – (a) Scanning electron microscope image of a narrow Bi<sub>2</sub>Se<sub>3</sub> nanowire with a rather short perimeter  $L_p = 280$  nm and ohmic Al contacts; (b) Aharonov-Bohm periodic oscillations, with the first two harmonics being visible directly in the  $G(B_{\parallel})$  trace; (c) Fast-Fourier transform revealing higher-order harmonics, up to  $n = 6$  at very low temperature.

Since this periodic behavior is specific to topological surface states (with a flux-periodic energy spectrum), it can always be unveiled by a careful fast-Fourier transform (FFT) analysis, provided that enough oscillations are measured (that is, when the field range largely exceeds the fundamental AB period). For a micron-long Bi<sub>2</sub>Se<sub>3</sub> quantum wire with perimeter  $L_p = 280$  nm, up to six harmonics were clearly resolved at very low temperature, as seen in Fig.4.3c) [39]. For short wires ( $L \sim L_\varphi^{\text{BS}}$ ), we also remark that a complication comes further from that aperiodic conductance fluctuations due to bulk carriers coexist with surface periodic AB oscillations [although  $L_\varphi^{\text{BS}} < L_\varphi^{\text{SS}}$ , the self-averaging of coherent bulk transport is reduced at very low temperatures due to their charge transport dimensionality  $d = 3$  and to longer  $L_\varphi^{\text{BS}}$  values]. Besides, because  $G(B_{\parallel})$  curves are measured over a finite field range, the FFT of bulk aperiodic conductance fluctuations often results in a non-monotonous background, possibly giving “peaks” but with no relation to a periodic behavior, contrary to that of  $G(B_{\parallel})$  changes due to the AB interference. The ballistic nature of the transverse motion in such quantum wires results in an unusual temperature dependence of the phase coherence length  $L_\varphi^{\text{SS}}$ , with a  $1/T$  behavior observed for all harmonics. This is the signature of both ballistic transport ( $L_\varphi = v_F \tau_\varphi$ ) and a decoherence time  $\tau_\varphi \propto 1/T$  limited by a weak coupling to fluctuations of the environment [27]. All other scenarios based on decoherence limited by either the Nyquist noise or the thermal noise give a very different power-law dependence. An extra signature of the quasi-ballistic regime is also found when considering the relative amplitude of AB harmonics. Contrary to the case of a diffusive mesoscopic conductors, their amplitudes are not increasingly smaller for higher orders  $n$  and they cannot be described by an exponential damping behavior

related to the ratio  $L_\varphi/L_n$ , where  $L_n = n * L_p$  [27]. This is due to disorder and to both geometric and contact effects, which all influence details of the quantum interference for different quantum coherent paths, in the quasi-ballistic regime [36]. In general, it remains difficult to investigate details of the AB oscillations in this regime, due to the complex mixing of all harmonics in the presence of disorder, which varies for different configurations of the microscopic disorder (as obtained by thermal cycling at room temperature of a given mesoscopic conductor).

### 4.3 Quantum coherence II : non-universal conductance fluctuations

More information about the weak scattering of quantized surface modes by a non-magnetic disorder can be obtained by studying the (static) conductance fluctuations due to the longitudinal motion of surface carriers in a highly-disordered 3D TI quantum wire. Contrary to the case of a diffusive mesoscopic conductor, their statistical properties such as the conductance variance are not universal and they can vary when the quantized Dirac band structure is modified by an Aharonov-Bohm flux [34]. Using a 3D vector magnet, we could vary independently the longitudinal field (tuning of the energy spectrum; transverse motion) and the transverse magnetic field (probing the aperiodic conductance fluctuations due to disorder; longitudinal motion). This provides the complete mapping of quantum interference, as seen in Fig.4.4a). It was shown that the absolute amplitude of the conductance variance  $\widehat{Var}G = \delta G_{\text{rms}}^2 = \langle G - \langle G \rangle \rangle^2$  has a periodic modulation with the magnetic flux (Fig.4.4b), a property specific to surface transport (well-defined cross section). This behavior is well captured by numerical calculations (Fig.4.4c and d), which also reproduce the correct amplitude of this modulation  $\text{mod}(\widehat{Var}G)$ . We evidenced that nonuniversal conductance fluctuations are the signature of the weak coupling between transverse quantized modes induced by disorder, and we inferred the amplitude of the disorder broadening  $\Gamma$  from the temperature dependence of the modulation  $\text{mod}(\widehat{Var}G)$ , in rather good agreement with numerical calculations [34]. Because  $\Gamma \ll \Delta$ , quasi-ballistic transport is a common property to all populated surface modes, each of them giving a significant contribution to the conductance (close to  $G_0$ ) as compared to that of the perfect transmission case ( $G_0$ ). Besides, the sharp evolution of  $\widehat{Var}G$  with the energy of successive transverse modes (Fig.4.4c) suggested that only a limited number of partially-opened modes (nearby  $E_F$ ) contribute to the conductance fluctuations, due to a rapid energy dependence of the transmission for all channels.

### 4.4 Quasi-ballistic transport : disorder and transmissions

To evidence that this interpretation is actually very general to all quantum corrections to the conductance in 3D TI quantum wires, it is important to calculate the energy dependence of the transmissions for all modes, taking disorder into account but also interfaces with metallic contacts. This was done in a comparative study of numerical calculations with an analytical model that captures the main property common to all quasi-1D spin-helical surface modes, that is, the suppression of scattering due to quantum confinement

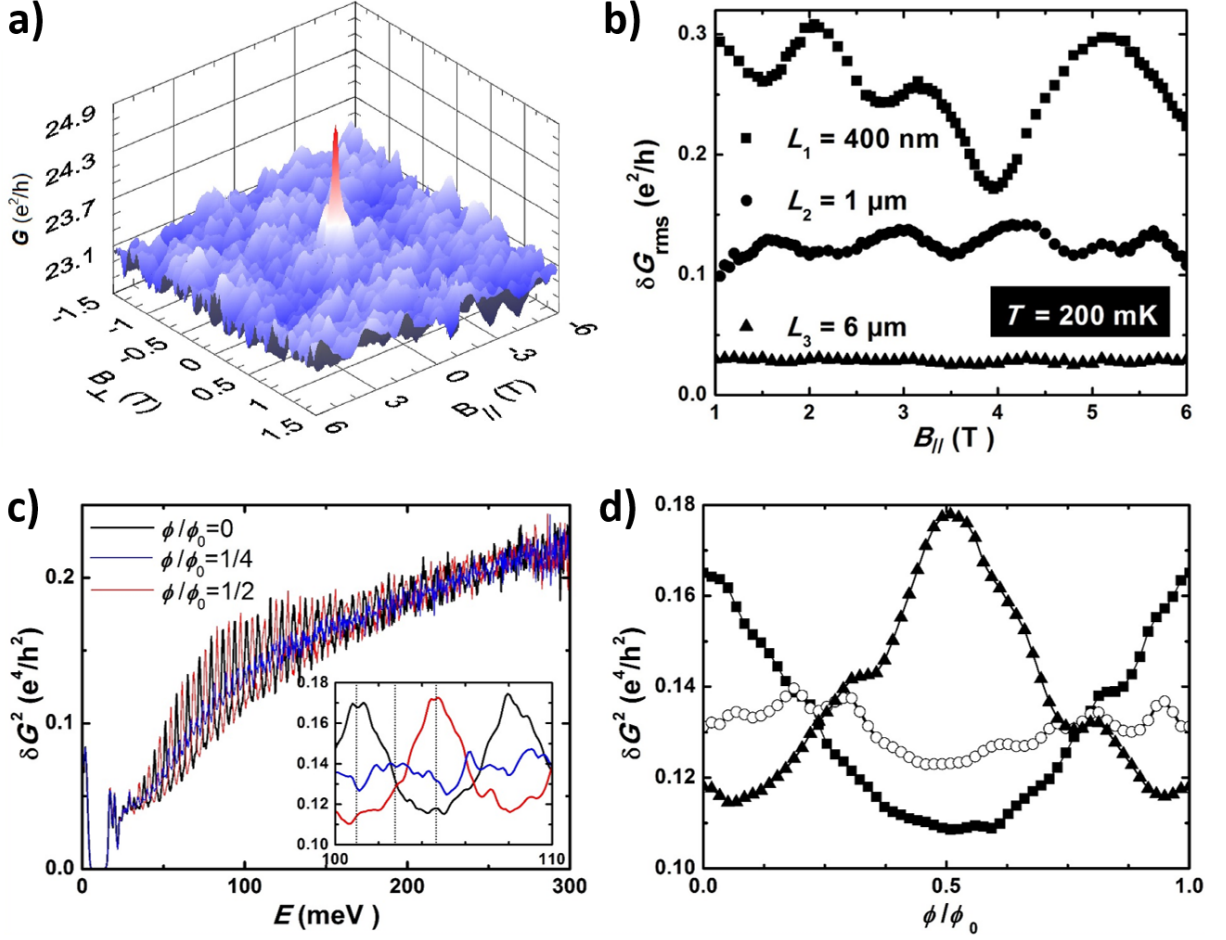


FIGURE 4.4 – (a) Quantum corrections to the conductance of a  $\text{Bi}_2\text{Se}_3$  quantum wire mapped over a large range of longitudinal magnetic fields (magnetic flux) and transverse magnetic fields, at very low temperature; (b) Flux-dependence of the conductance variance revealing a non-universal behavior and a periodic evolution, a signature of the quasi-ballistic transport of Dirac surface modes; (c and d) Numerical calculations of the energy (c) and flux (d) dependences of the conductance variance in a disordered 3D TI quantum wire, showing periodic evolutions due to quantum confinement of all transverse modes.

[34]. The set of transmissions  $T_i$  represents the mesoscopic code of a coherent conductor, from which all important quantities can be calculated, the simplest one being the total conductance  $G = G_0 \sum_i T_i$ . Importantly, the transmissions were found to nearly reach unity for all modes when their longitudinal kinetic energy exceeds  $\Delta$  (Fig.4.5). The same (rapid) evolution was found even for high-energy modes, though over a slightly broader energy window, thus explaining why quasi-ballistic transport properties exist for many modes over a broad energy range. This also shows that diffusive longitudinal transport is realized only for conductor lengths that largely exceed the transport length. Contrary to the case of 2D quasi-particles with isotropic scattering, for which the transition from the ballistic to the diffusive regime is rather abrupt ( $l_e < L < l_{tr}$ , with  $l_{tr} \leq 2l_e$ ), the quasi-ballistic regime in 3D TI quantum wires exists over a wider parameter range



$(l_{\text{tr}}/2 < L < \alpha l_{\text{tr}}$ , with  $l_{\text{tr}} \gg l_e$  and  $\alpha$  is related to the aspect ratio  $L/L_p$ ). This unusual behavior, related to the enhanced transport length, is due to both the spin texture of Dirac modes (anisotropic scattering) and to their large confinement energy in quantum wires, both favoring the weak scattering of quantized modes by disorder.

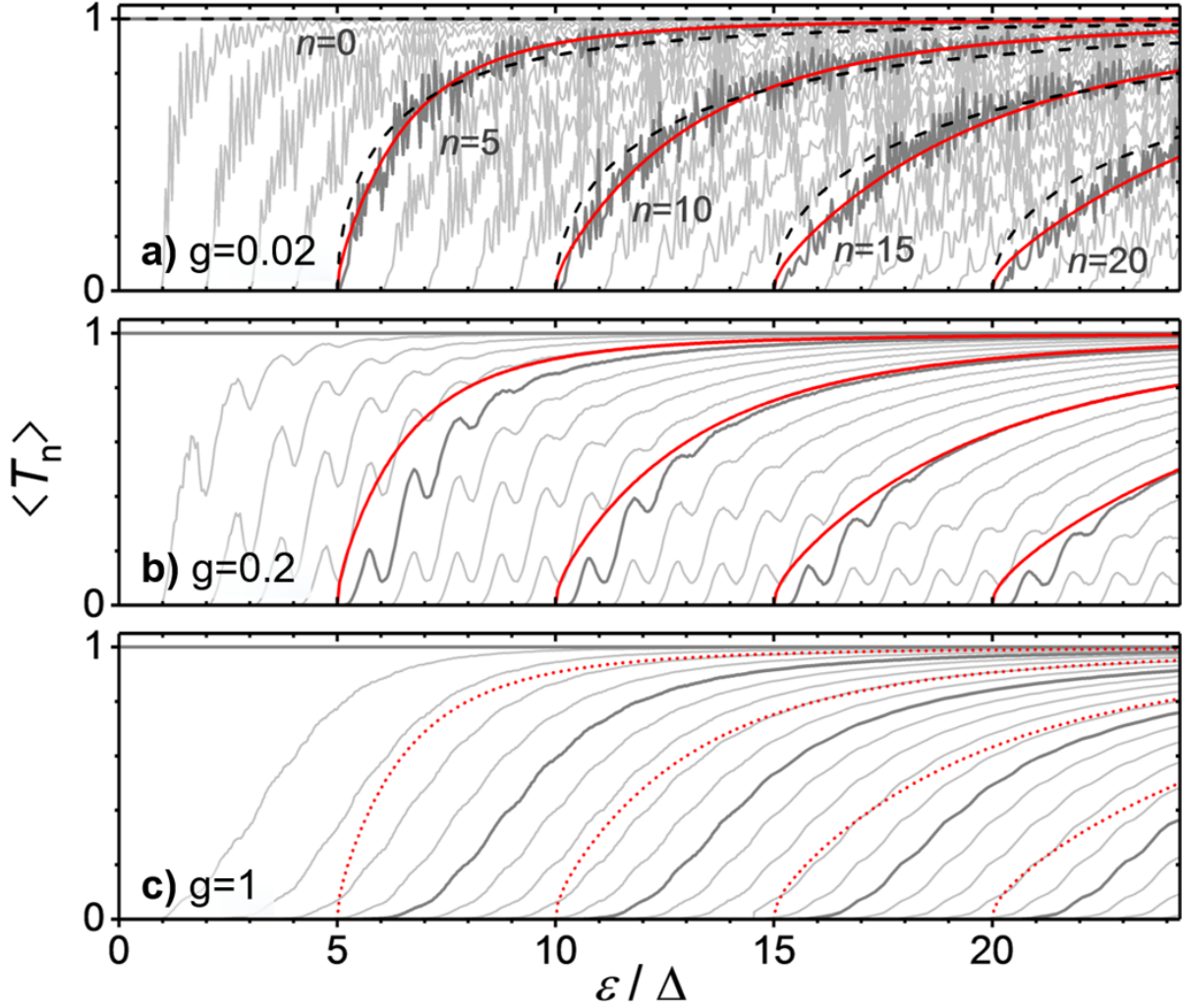


FIGURE 4.5 – Energy dependence of the surface-mode transmissions in disordered 3D topological insulator quantum wires, for three values of the disorder strength  $g = 0.02$  (a),  $g = 0.2$  (b), and  $g = 1$  (c).

## 4.4.1 Scattering by disorder and contacts

Considering the scattering by disorder as due to a random potential of energy barriers (Gaussian disorder, with a correlation length  $\xi$ , see Fig.4.6a), it is possible to give an analytical description of the transmissions of high-energy modes propagating between two transparent ohmic contacts, for different degrees of disorder from the clean limit (ballistic, Fabry-Pérot) to the dirty limit (diffusive) [36]. In the quasi-ballistic regime, we found that the conductance is determined by the interfaces with metallic contacts (similarly to a clean conductor) and not by details of the microscopic disorder in the quantum wire. Due to the quantum confinement of Dirac fermions with evenly-spaced energy levels, the energy dependence of the conductance can oscillate at low energies (whereas it has a linear dependence at high energy, as for the 2D limit) and the average transmission per mode only depends on the nature of the contacts. For an intermediate disorder strength  $g = 0.2$ , Fabry-Pérot interferences are suppressed by efficient phase averaging, and inter-mode scattering results in an oscillatory energy dependence of the transmission, due to the increased density of states at the onset of a nearby mode and because disorder broadening remains smaller than the energy level spacing. This can be directly seen in the energy dependence of the transmission of a high-energy mode in a  $\text{Bi}_2\text{Se}_3$  quantum wire (using a realistic value of  $\xi$ ), as shown in Fig.4.6b). For very large values of  $g$ ,  $\Gamma$  exceeds  $\Delta$  and charge transport becomes diffusive.

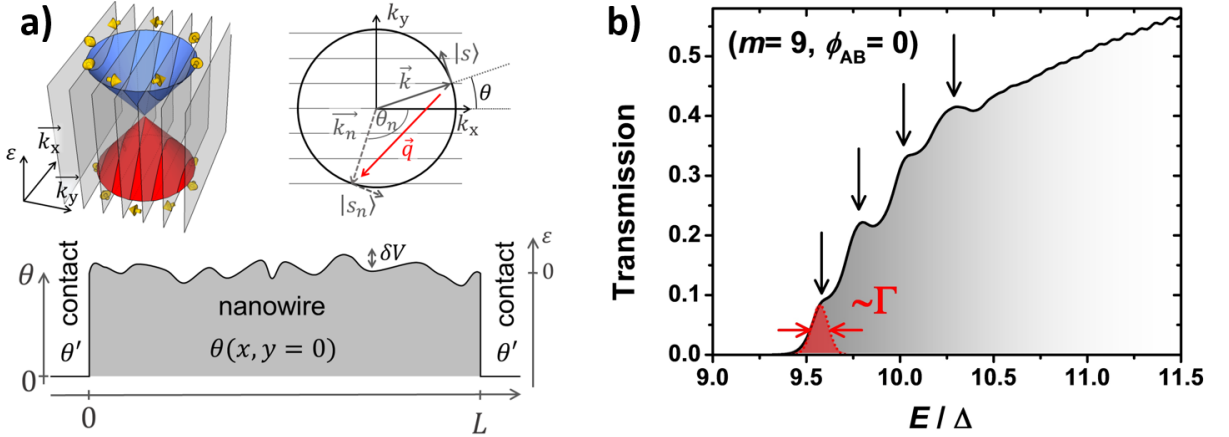


FIGURE 4.6 – (a) Quantized band structure of surface modes for a magnetic flux  $\Phi = 1/2\Phi_0$  and inter-mode scattering induced by a random disorder potential  $\delta V$ ; (b) Energy dependence of the transmission of the  $m = 9$  quantized mode for  $\Phi = 0$ , showing resonances due to disorder-induced inter-mode mixing, with an energy broadening  $\Gamma$ .

To understand why the quasi-ballistic regime exists over a broad range of parameters, it is important to consider the energy dependence of the transport length [see [36] for details], as shown in Fig.4.7 for different  $g$  values. Contrary to the case of massive quasiparticles,  $l_{\text{tr}}$  does not vanish at low energy for Dirac fermions in 1D. Instead, it diverges and a similar behavior occurs at high energy, due to the anisotropy of scattering. As a consequence, the transport length has a minimum value that depends on the strength of disorder. For a given disorder correlation length  $\xi$ , this minimum value is obtained for  $k\xi \approx 1$  and the values of  $l_{\text{tr}}^{\text{min}}$  can be much larger than the transverse dimensions of

the nanostructure for all energies, for a broad range of  $g$  values, so that the condition for quasi-ballistic transport is always fulfilled for such highly-disordered 3D topological insulator quantum wires. Good agreement was found between this simplified analytical model and numerical calculations, for that details of the microscopic disorder do not affect the conductance in this regime, which is mostly determined by metallic contacts.

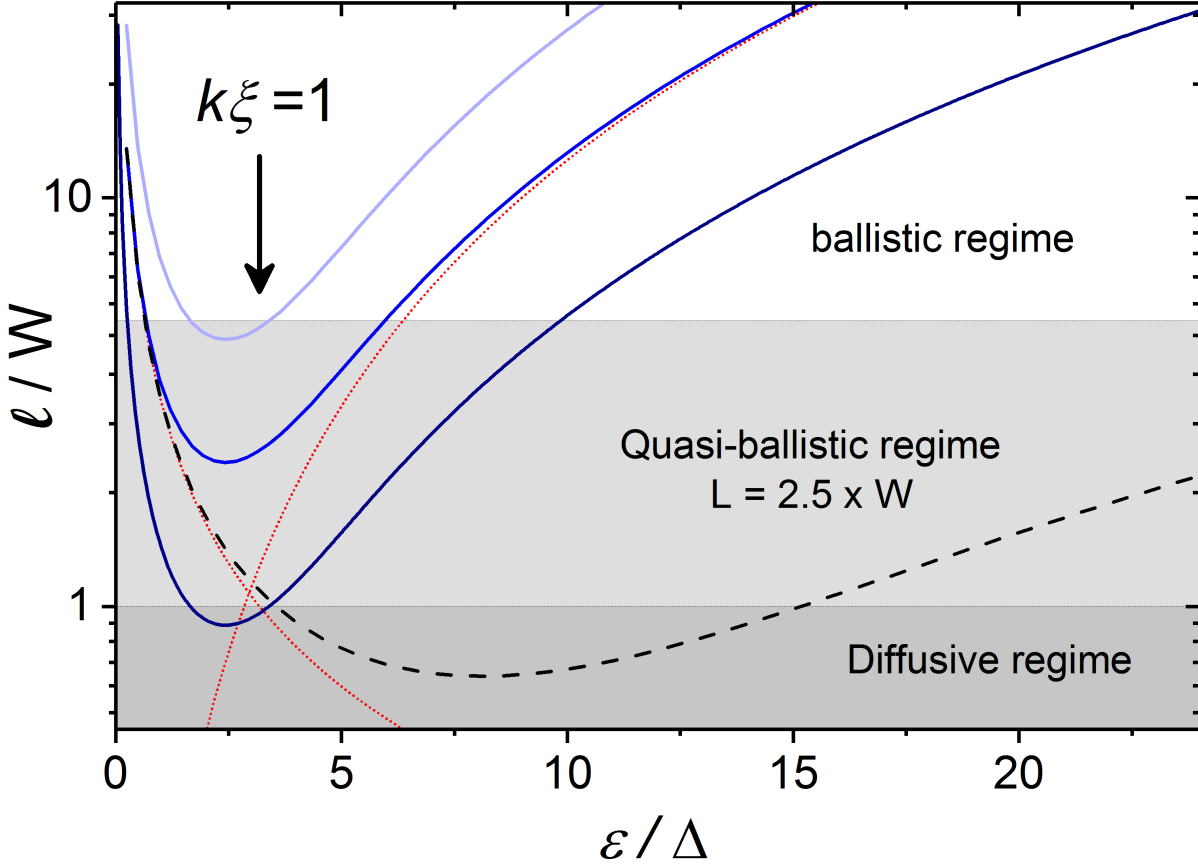


FIGURE 4.7 – Energy dependence of the transport length  $l_{\text{tr}}$  in a quantum wire with a perimeter  $W = L_p$  and a transverse energy quantization  $\Delta$ , calculated for  $\xi/W = 0.05$  and  $g = 0.1$  (light blue),  $0.2$  (blue), and  $0.5$  (dark blue). Red dotted lines show asymptotic behaviors related to the divergence of  $l_{\text{tr}}$  at low or high energies, due to the density of states or to the anisotropy of scattering, respectively. In all cases, the transport of all surface modes is ballistic or quasi-ballistic.

#### 4.4.2 Quantitative derivation of the AB amplitude

Based on the transmissions calculated for different values of the magnetic flux (giving different quantized energy spectra), the energy dependence of Aharonov-Bohm oscillations can be calculated. As seen in Fig.4.8a), their amplitude decreases with the energy of surface modes and a good quantitative agreement with experiments was found at high energies. The oscillatory behavior reported in experiments [58–60] is also well reproduced, as well as energy-periodic phase shifts [60] which are actually due to the quantized band structure (Fig.4.8b) [36] and *not* to the perfectly-transmitted mode (as reported by other groups).

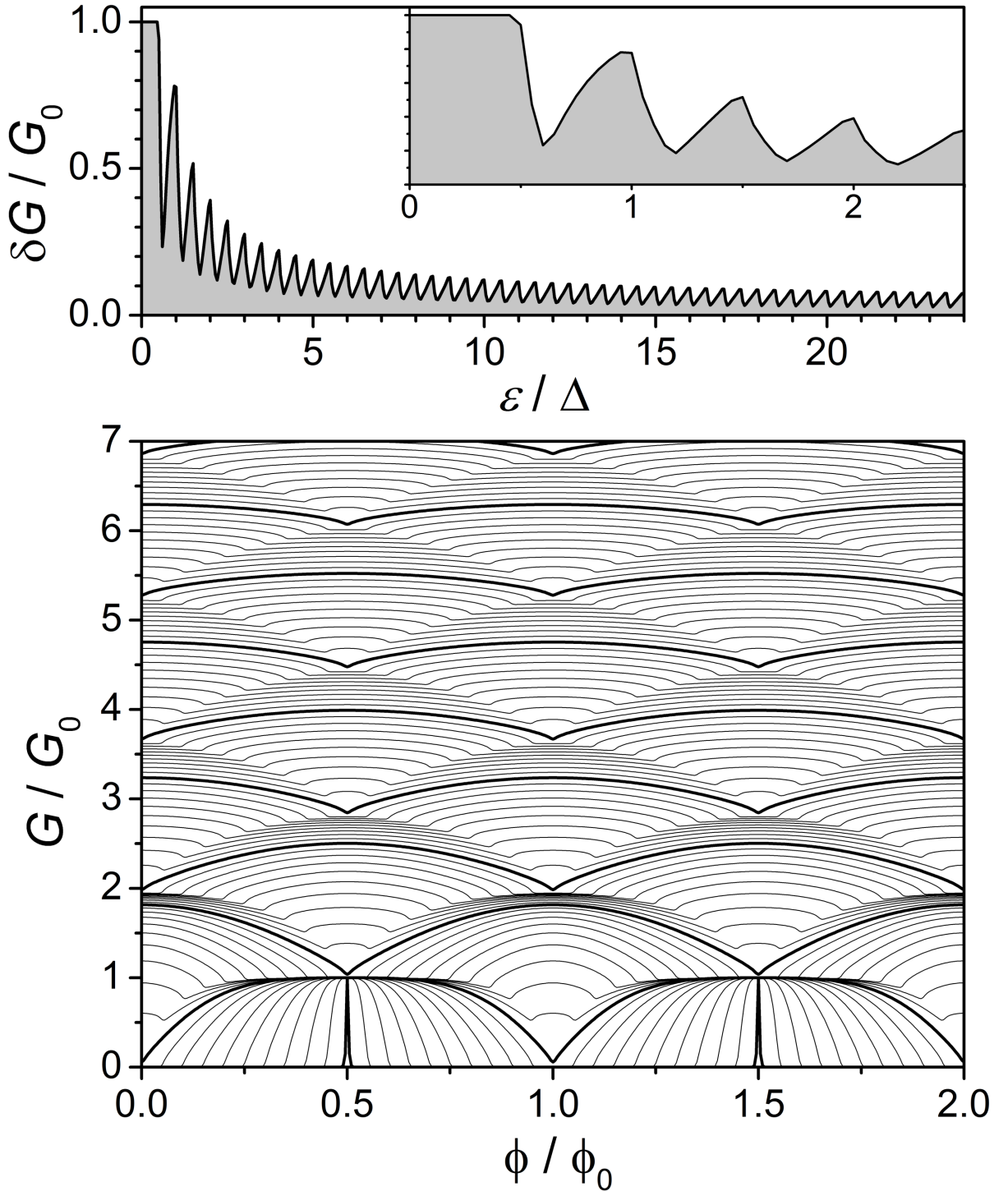


FIGURE 4.8 – (a) Energy dependence of the Aharonov-Bohm amplitude, rapidly decreasing from the conductance quantum  $G_0$  (contribution of the topological mode only) to a fraction of  $G_0$  (contribution of higher-energy transverse modes); (b) Flux dependence of the conductance for different energies, from 0 to  $4.5 \Delta$  (successive thin lines correspond to an energy change  $0.05 \Delta$  and thick lines to multiple values of  $1/2 \Delta$ ). Phase shifts are due to the quantized energy spectrum of surface modes. The influence of the topological mode is seen only for very low energies.



## Conclusions and perspectives

The weak coupling of surface states in 3D topological insulator quantum wires, due to both their spin texture and the quantum confinement of Dirac fermions, gives unique opportunities to control novel quantum states in mesoscopic conductors. In particular, dissipationless chiral states could be induced in a simple device geometry, using a nanowire.

Yet, there is still a technological barrier hindering the control of a small number of transverse quantized modes, with an energy close to the Dirac degeneracy point, mostly due to some intrinsic limitations in conventional 3D TI materials. Although the  $\text{Bi}_2\text{Se}_3$  family offers many advantages (tunable band structure in solid solutions of ternary compounds and high-quality single-crystalline nanostructures), it remains difficult to achieve surface transport only, and, importantly, to tune the Fermi energy near the Dirac point.

- Presently, our efforts to reach the small- $N$  limit are still based on simple strategies :
- either to *reduce the chemical potential*. The vapor-transport method is now used to grow ternary compounds  $\text{Bi}_{2-x}\text{Sb}_x\text{Te}_3$ , with the potential to significantly reduce the bulk carrier density.
  - or to *increase the quantum confinement energy*. Ultra-narrow nanowires can be grown by electro-deposition, and our study of the weak localization already confirmed their rather long phase-coherence length [37].

However, the next generation of electronic devices based on 3D topological insulators will necessarily be developed from advanced functional heterostructures. An important challenge will be to control the interface band bending, with a good interface quality, and to optimize the coupling between metallic contacts and interface Dirac fermions. This is particularly true for spin transport experiments, which require to minimize the momentum/spin relaxation below the contacts in order to use the intrinsic potential of electronic states with spin-momentum locking. Toward this goal, new growth and nanofabrication methods need to be envisioned, in combination with those already used to prepare high-quality single-crystalline nanostructures (vapor transport, vapor-liquid-solid epitaxy, and chemical-vapor deposition). Novel techniques already gave some promising results, for instance to realize highly-transparent superconducting contacts, by in-situ stencil lithography on ultra-thin films grown by molecular beam epitaxy, and investigate topological superconductivity [61]. Also, atomic layer epitaxy holds promise to prepare core-shell lateral nanostructures with compatible materials, such as  $\text{Sb}_2\text{Se}_3$  and  $\text{Bi}_2\text{Se}_3$ , being more adapted to control the electro-chemical potential at the interface with a topological insulator.



# Research project





## 6

# Introduction

Topological materials in 2D offer the possibility to control 1D dissipationless edge states, with some potential for quantum spintronics and quantum interferences devices.

For fundamental studies, this gives a platform to investigate the grounds of quantum mechanics at the mesoscopic lengthscale, as already initiated for instance with edge-state interferometers operating in the quantum Hall regime of AlGaAs 2DEGs.

For applications, these concepts hold promise for low-power electronics and interconnection, but also give the possibility to combine electrons, photons and phonons in coupled quantum devices that would all operate with topological edge states.

Among the three topological Hall effects establishing the so-called “quantum Hall trio”, electronic phases with spin-polarized states are particularly interesting since they are naturally prone to ballistic transport at a large scale (weak coupling to their environment). Different from the orbital quantum Hall effect (QHE), the spin-orbit based quantum spin Hall (QSH) and quantum anomalous Hall (QAH) phases exist in zero magnetic field. Since their discovery in 2007 (QSH) and 2013 (QAH), the degree of quantization of their conductance has been probed in different materials. A striking difference rapidly emerged however. Though measured with high-quality semiconducting heterostructures (II-VI or III-V), the QSH always showed a rather large deviation to the perfect quantization at  $G_0 = 2e^2/h$ , up to about 10%. On the contrary, the QAH was evidenced in diluted magnetic topological insulators (with both structural and magnetic disorder) and a perfectly-quantized conductance was always found, by different groups, with an accuracy within the ppm range for experiments conducted in non-metrology laboratories. To date, there is no other effect in zero magnetic field rivaling the stability of the QAH. Yet, there is still some room left to improve the measurement accuracy and test its true robustness, in comparison to the ultimate metrology grade as realized in the QHE regime. This also calls for new materials, beyond diluted magnetic topological insulators, so as to decrease the degree of disorder and also to increase the operating temperature.

Topological materials in 3D give some alternatives to realize chiral edge transport, and this is the main purpose of this research project. Two different strategies are followed :

- Opening a trivial band gap by the quantum confinement of surface Dirac fermions in narrow nanostructures, and driving the chiral mode by applying a moderate magnetic field ;
- Opening a band gap with a non-trivial topology for surface Dirac fermions in a 3D topological insulator, mostly by combining magnetism and topology.

More specifically, nanostructures of nonmagnetic 3D topological insulators could be used for spin-based quantum information, for instance, either for ballistic interconnects (long nanowires) or local spin filters (short nanowires, behaving as quantum “dots”).

Recently discovered antiferromagnetic topological insulators have the potential to go well beyond, as they open a new route to create novel topological electronic states, and to tune them by controlling the magnetization with small magnetic fields. Since these are van-der-Waals layered magnets, this research also has some link with the emerging work on the van-der-Waals growth of high-quality 2D magnetic heterostructures.

# Chiral mode in 3D TI quantum wires

## 7.1 Towards novel 3D TI heterostructures

As already mentioned, it is not straightforward to observe the conductance quantization in simple 3D TI nanostructures, for that they have a finite residual bulk conductivity. Still, there are strategies to increase the surface-to-bulk conductance ratio and, most important, to lower the number of occupied surface modes, with the goal to tune the surface chemical potential with an electrical gate.

This could be done in two realistic ways :

- With ultra-flat nanoribbons and a dual planar electrical gate (planar growth ; vapor transport on  $\text{SiO}_x/\text{Si}$ ). This simple approach would only need some ex-situ post-growth processing ;
- With narrow nanowires and a circular electrical gate (vertical growth ; Au-catalyst assisted VLS method). As-grown nanostructures can be transferred under high vacuum to coat them with an amorphous dielectric, before a metal layer is evaporated and the nanofabrication process (transfer, contacts) is completed ex-situ.

In both cases, best would be to have a rather low bulk carrier density, so as to avoid a strong electrostatic screening by topological surface states. With this aim, novel ternary  $\text{Bi}_{2-x}\text{Sb}_x\text{Te}_3$  nanostructures could be good candidates. Very recently, some progress was made by our collaborator Silke Hampel (IFW Dresden), who succeeded in growing alloyed nanostructures with a composition range spanning from  $x = 0$  to  $x = 1$ .

Yet, the difficult control of charge transfer at interfaces clearly calls for the next generation of 3D TI nanostructures, based on the hetero-epitaxy of lattice-matching materials, as planar epilayers or vertical core-shell nanostructures. The catalyst-assisted vapor-liquid-solid (VLS) growth has been well developed for III-V materials (Fig.7.1), and nanowires have laterally-confined 2DEGs with quasi-1D transport properties [Lucot2011]. A similar strategy was applied to grow core-shell heterostructures of thermoelectric materials that belong to the  $\text{Bi}_2\text{Se}_3$  family (Fig.7.1), such as  $\text{Sb}_2\text{Se}_3/\text{SbS}_3$  nanowires made by VLS and atomic-layer epitaxy (ALE) [62]. In collaboration with Andy Thomas (IFW Dresden), similar structures could be grown, combining a topological insulator ( $\text{Bi}_2\text{Se}_3$ ) and a trivial insulator ( $\text{Sb}_2\text{Se}_3$ ). This would give access to nanostructures with a good quality of

interfaces (reduced interface doping) and a dielectric shell that can be used to add an efficient electrical gate. Though extremely interesting, such developments are challenging and therefore probably more adapted to longer-term projects.

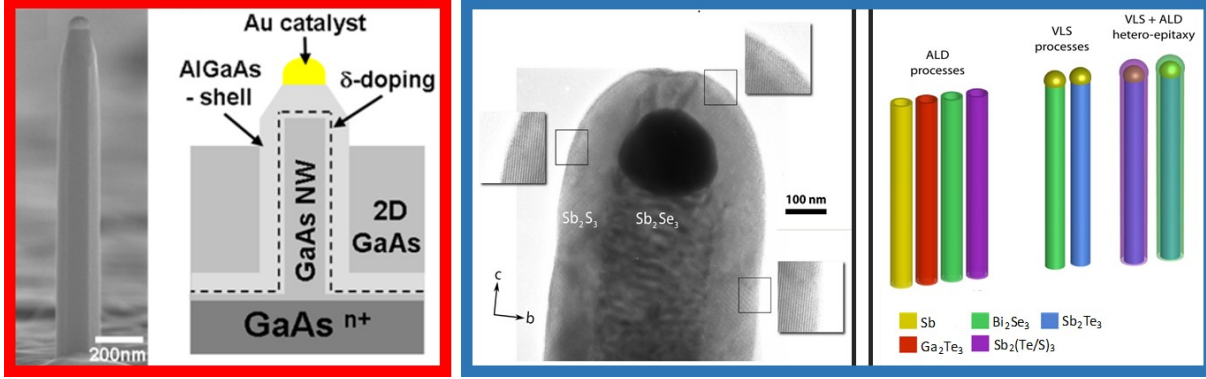


FIGURE 7.1 – (Left), Nanowire of an AlGaAs core-shell heterostructure, realizing a 2DEG hollow conductor (with a hexagonal symmetry). From [26]; (Right), Combined VLS/ALE growth of  $\text{Sb}_2\text{Se}_3/\text{SbS}_3$  nanowires, and other chalcogenide heterostructures. After [62].

## 7.2 Energy spectroscopy of quasi-1D transport

Using gate-tunable 3D TI nanostructures, it will become possible to realize the energy spectroscopy of the quantum transport properties. This would give the ultimate quantitative test to confirm the prediction of the model we developed (and discussed in 4.4.2) to describe the transport properties of quasi-ballistic surface modes in 3D TI quantum wires, and to unveil the single topological mode.

Most important, this would allow us to evidence :

- The energy dependence of the amplitude of Aharonov-Bohm oscillations, as shown in Fig. 4.8 ;
- The conductance quantization expected for quasi-1D quantum wires ;
- The “true” topological transition due to the flux-periodic appearance and vanishing of the zero-transverse-energy topological mode (with a perfect transmission), whereas previous reports were actually do to quantum confinement only.

In the small- $N$  limit, the conductance will be dominated by the contribution of a single mode, the only topological one, with a perfect transmission, independent of the disorder potential. This perfectly-transmitted mode is the key to create novel topological states in 3D TI quantum wires, such as Majorana bound states in presence of  $s$ -wave superconducting contacts, or a chiral edge state induced by a magnetic transverse field.

### 7.3 Perfectly transmitted topological mode

With the goal to investigate the physics of 3D TI quantum wires close to the Dirac point, best results could be obtained in long and ultra-narrow nanostructures [37], since low-energy modes other than the topological mode have a reduced transmission due to disorder (minimum of the backscattering length, so that  $l_{\text{tr}} \ll L$  and  $G \ll G_0$ ). Furthermore, since the transport of surface modes is quasi-ballistic, it will become important to optimize/control the coupling between metallic contacts and the transverse wave function of a given mode. In particular, the amplitude of probability can have an azimuthal angle dependence, which varies from one mode to another, so that quantum transport properties will ultimately depend on the exact geometry of the mesoscopic conductor.

To date, there is no study of the small- $N$  limit, and this clearly remains a big challenge. In order to evidence the influence of the topological mode on the conductance, it will be necessary to set the mesoscopic conductor in specific conditions :

- In the small- $N$  limit (less than about 4–5 modes populated), the relative contribution of the topological mode to the magneto-conductance will dominate ;
- In long-wire limit (regime  $L \gg l_{\text{tr}}$ ), the transmission of all modes but the topological one will be reduced.

### 7.4 Helical to Chiral transition

Remarkably, the low-energy spectrum can be modified by a large-enough transverse magnetic field [56]. For a rectangular cross section (Fig. 7.2), a striking property is related to the evolution of the topological mode from a helical state to a chiral edge state, when a moderate transverse magnetic field is applied. The specific orbital response of such 3D TI quantum wires corresponds to an intermediate situation between the quantum spin Hall in a 2D TI and the quantum Hall effect in 2DEGs.

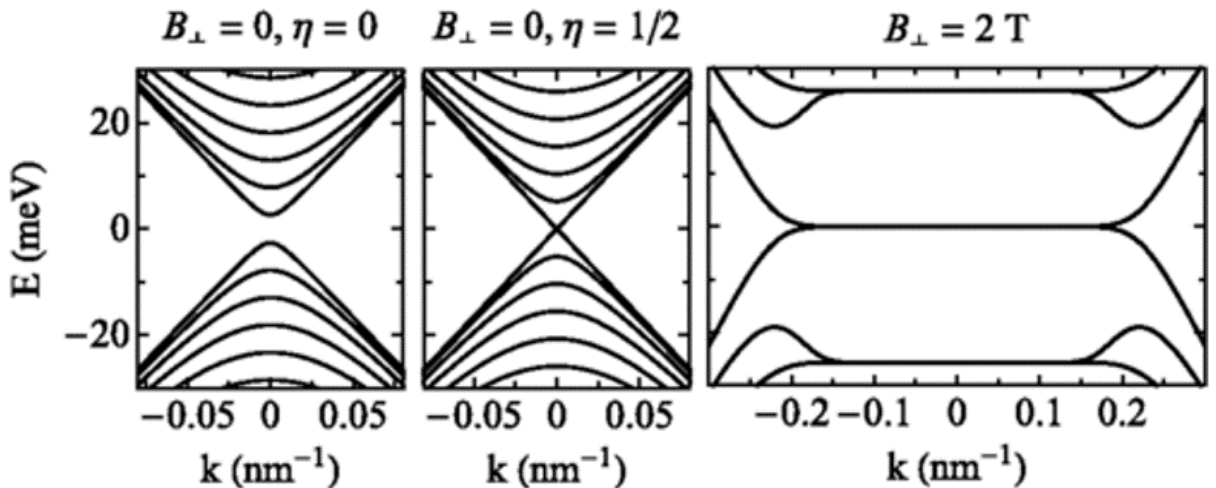


FIGURE 7.2 – Energy spectrum of a 3D TI quantum wire with a rectangular cross section ( $h = 40$  nm;  $w = 160$  nm) for  $\eta = \Phi/\Phi_0 = 0$  (left) and  $\eta = 1/2$  (center). A transverse field creates a chiral edge states without dispersion, independent of  $\eta$  (right). After [56].

The control of low-energy quantum states in 3D TI nanostructures would offer novel opportunities for their quantum manipulation as well as for spin filtering, by tuning the quantum states with an electric or a magnetic field.

Importantly, when coupled to metallic electrodes with gapped excitations, the topological mode generates novel quantum states with an intrinsic topological protection, such as Majorana bound states or spin-polarized edge states in the quantum anomalous Hall regime, in a simple device geometry [56]. These could be important for quantum dynamics studies, with limited decoherence.

## 7.5 More confinement...

Non-topological low-energy modes are also interesting for their energy tuning, by a gate voltage or a magnetic flux, which is associated with a continuous change of their spin state between nearly-orthogonal states. Besides, these can be either 1D extended states (long quantum wires) or 0D localized states (short quantum wires, that is, for  $L < l_{tr}$ ).

As shown in Fig.7.3, a simple nanofabrication process could allow us to create such quantum “dots”, for which the longitudinal quantum confinement will start to exceed the energy broadening. Indeed, short quantum wires can be simply obtained from long wires by using a pair of metallic contacts with a small spacing, and then cover the M/TI/M segment with a dielectric that can be used as a hard mask and also to create a self-aligned electrical top gate.

Of interest for quantum spin transport, this short conductor would be a tunable spin filter, since each of its quantum states has a specific spin orientation that can be selected for resonant tunneling, within the energy range  $\Delta$  ( $\sim 10$  meV), and which can be modified either by the electrical gate or by a longitudinal magnetic field.

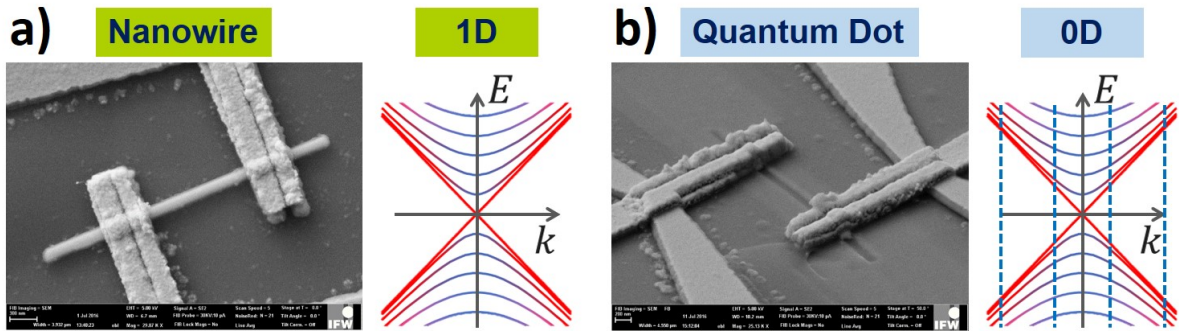


FIGURE 7.3 – Quantum confinement of topological surface states in 3D topological insulator nanowires, with **a)** extended quasi-1D surface modes (long wires) or **b)** localized OD surface states (short wires). Due to spin-momentum locking, spin states are well defined, and the spin projection can vary from perpendicular (red colored) to colinear (blue colored) to the wire axis.

# Chiral modes in magnetic 3D TIs

## 8.1 Introduction

Novel topological electronic phases can emerge when magnetism and topology are combined in a material. For  $\mathbb{Z}_2$  topological insulators, this happens when an internal exchange interaction adds to the band inversion mechanism. In particular, long-range magnetic order can induce a phase transition to another topological phase if :

- The magnetization is homogenous ; it breaks time-reversal symmetry and can couple topological interface states of opposite spin helicity. This creates a non-trivial gap in the topological-state band structure, and induces spin-polarized edge states : the quantum anomalous Hall phase ;
- The magnetization is not homogenous but has a periodic structure, like in antiferromagnets for instance. In some cases, this extra symmetry can modify the topology of the electronic band structure and generate novel phases, such as the axion insulator or a Weyl semimetal.

The influence of an exchange field was first considered in the work of Shoucheng Zhang, suggesting to turn the QSH phase in II-VI quantum wells into the QAH phase by adding magnetic dopants [63]. Later, the influence of an extra symmetry added by the magnetic long-range order was discussed in the case of antiferromagnetic topological insulators [64]. This physics is rich since both the Néel vector and details of the magnetic order at interfaces control the nature of the topological phase. Interestingly, due to a parity effect related to the magnetic surface termination, physical surface edge steps are predicted to host dissipationless chiral electronic modes.

In all cases, novel topological electronic phases can be controlled by the magnetization, via its orientation or its order parameter, and it becomes crucial to harness and engineer the magnetic properties of topological materials, down to the microscopic level.



## 8.2 Novel magnetic topological insulators

### 8.2.1 Magnetic van-der-Waals heterostructures

With the discovery of 2D ferromagnetism in atomically-thin magnets [65, 66], van-der-Waals layered materials and heterostructures can now include magnetic properties. Since some crystals also have a nontrivial topology of their band structure, such as members of the  $\text{Bi}_2\text{Se}_3$  family, this class of materials seems ideal to search for novel magnetic topological insulators.

Interestingly, this approach gives a unique way to combine many new concepts that arose from the graphene-driven research activities in the last decade. Ultimately, there could be some specific interest for spintronic devices to mix 2D materials with vanishingly-small spin-orbit coupling like graphene (long-range spin transport, intrinsic magnetism) and others having topological electronic states induced by strong spin-orbit interactions. A remarkable result that strengthens this link is the recent realization of the QAH phase in twisted bilayer graphene on boron nitride, and the possibility to switch the chirality with small electrical currents [67].

Generally speaking, such intrinsic magnetic properties are best to give some homogenous magnetization and result in stable topological phases, in particular as compared to diluted magnetic topological insulators. Besides, they offer an interesting alternative to other approaches based on magnetic proximity effects that usually suffer from more difficulty to control some properties at interfaces, such as unavoidable spin relaxation.

### 8.2.2 Intrinsic magnetic 3D topological insulators

In this research project, we target a specific class of layered magnetic topological insulators, the MBT compounds, which derives from the  $\text{Bi}_2\text{Se}_3$  family. These materials offer a couple of advantages, that are :

- High-quality single crystals can be obtained by thermodynamic growth, for all possible elements [M is a  $3d$  or a  $4f$  element ; B is Bi or Sb ; T is Te, Se or S] and compositions, and then exfoliated ;
- They can realize the weak coupling regime of antiferromagnets, with inter-layer exchange interactions that can be even smaller than achieved in multi-layered synthetic antiferromagnets ;
- Magnetic structures can vary from collinear antiferromagnets to uniform metamagnetism, and other magnetic orders are possible ;
- Their anisotropy could be increased by interfacing with other 2D magnetic insulators, or by using rare-earth elements ;
- They can be grown by van-der-Waals molecular epitaxy and integrated in a stack of compatible materials.

Most studies are presently dedicated to Mn-based compounds.

The magnetic base unit is a single  $\text{MnBi}_2\text{Te}_4$  septuple layer, formed by the intercalation of a MnTe magnetic layer within the nonmagnetic  $\text{Bi}_2\text{Te}_3$  (quintuple layer), as shown in Fig. 8.1a). It is a 2D ferromagnet (intralayer coupling  $J_F < 0$ ) with a perpendicular anisotropy  $K_U$  that stabilizes an out-of-plane magnetization.

Stacks of septuple layers form the bulk  $\text{MnBi}_2\text{Te}_4$  compound, with an antiferromagnetic interlayer coupling ( $J = J_{AF} > 0$ ). Recently, it was shown to realize the first antiferromagnetic topological insulator [68], with an  $A$ -type ground state, as the result of the specific symmetry related to its magnetic structure [64, 70]. This antiferromagnet breaks TRS but a new symmetry  $S = \Theta\tau_{1/2}$ , where  $\tau_{1/2}$  is a half-unit cell translation Fig.(Fig. 8.1b), gives a  $\mathbb{Z}_2$  topological insulator, similar to strong 3D TIs.

Interestingly, it is also possible to grow related crystals that have  $N$  units of the nonmagnetic  $\text{Bi}_2\text{Te}_3$  spacer in between 2D ferromagnetic layers and therefore a reduced inter-layer exchange coupling  $J$  (Fig. 8.1c). In particular, pure phases of two crystals,  $\text{MnBi}_4\text{Te}_7$  ( $N = 1$ ) and  $\text{MnBi}_6\text{Te}_{10}$  ( $N = 2$ ) were obtained by our collaborator Anna Isaeva (TU Dresden).

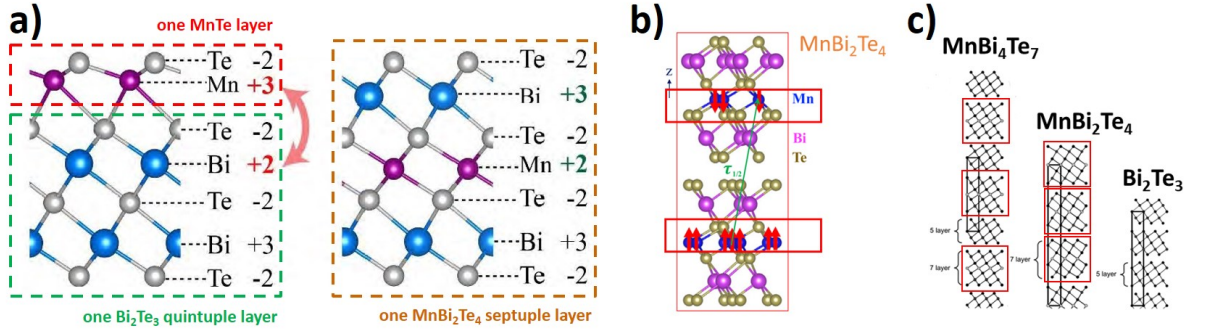


FIGURE 8.1 – (a), Formation of a  $\text{MnBi}_2\text{Te}_4$  septuple layer by the intercalation of a MnTe magnetic layer within the nonmagnetic  $\text{Bi}_2\text{Te}_3$ ; (b), Symmetry modified by the two magnetic sublattices with opposite magnetization in the antiferromagnetic state. The system is invariant by  $S = \Theta\tau_{1/2}$ , where  $\tau_{1/2}$  is a half-unit cell translation and  $\Theta$  is the TRS symmetry; (c) Stacks of magnetic septuple layers and nonmagnetic quintuple layers give antiferromagnets with different inter-layer exchange interaction strength. Ferromagnetic septuple layers are red framed. Adapted after [74] and [75].

As for transport properties, similar issues related to disorder in chalcogenides occur. Although  $[\text{MnBi}_2\text{Te}_4][\text{Bi}_2\text{Te}_3]_N$  are ideally stoichiometric magnetic topological insulators, these materials are highly  $n$ -doped and the temperature dependence of their in-plane resistivity is similar to that of dirty metals. Whereas the QAH phase can be evidenced in ultra-thin layers (no bulk conductivity), this remains an important limitation to investigate other insulating phases in thicker films, such as the axion insulator or the Chern insulator. Solutions exist and our collaborators are working in the chemical doping and optimization of crystals.

### 8.3 Magnetic properties of MBT materials

Another important aspect is to control the micromagnetic state of the “devices” studied. This can be done already, even with metallic samples. Different magnetic phases can be obtained in the Mn-based MBT compounds studied. We have already revealed that such crystalline MBT magnetic multilayers are actually textbook systems that realize the weak-coupling regime of uniaxial antiferromagnets, except  $\text{MnBi}_2\text{Te}_4$ , with robust metamagnetic properties controlled by their perpendicular anisotropy [38].

#### 8.3.1 Magnetic phases

Below their Néel temperature, the typical signature of an *A*-type collinear antiferromagnet, a spin-flop transition, is observed in both  $\text{MnBi}_2\text{Te}_4$  and  $\text{MnBi}_4\text{Te}_7$ .

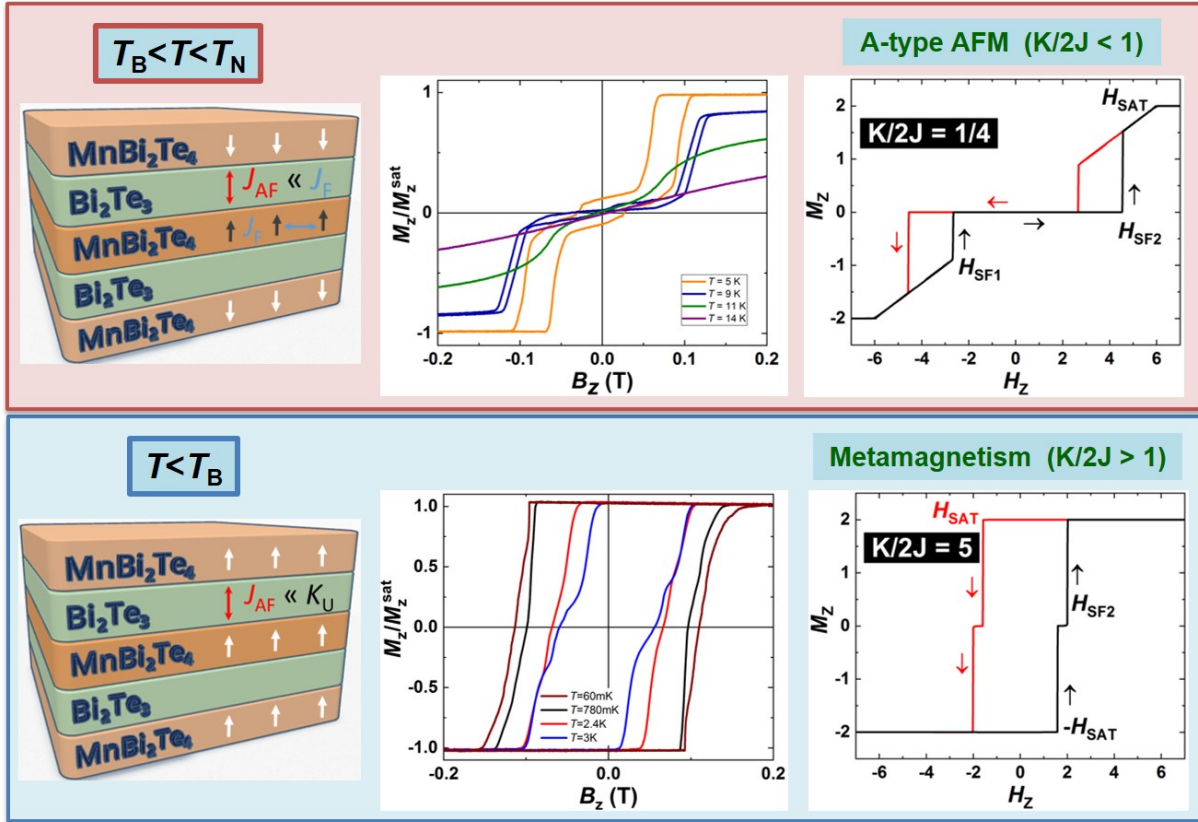


FIGURE 8.2 – Evolution of the collinear antiferromagnetic state (upper panel), with spin-flop transitions, to the metamagnetic state (lower panel) with spin-flip transitions and a fully saturated magnetization in zero field at low temperature, defining a blocking temperature  $T_B = 3$  K. The results are well reproduced by a bi-layer Stoner-Wohlfarth model, with different  $K/J$  ratios.

However,  $\text{MnBi}_4\text{Te}_7$  undergoes another transition to a bistable metamagnetic state at lower temperatures, with a fully saturated remnant magnetization below about 3 K and abrupt spinflip transitions (Fig. 8.2). This evolution is well described by a magnetic bilayer Stoner-Wohlfarth model with an interlayer exchange coupling  $J$  and a temperature-dependent effective anisotropy  $K$  related to the single-layer uniaxial anisotropy  $K_U$ .

This finding of metamagnetism is very general for van der Waals 2D-layered ferromagnets with a weak interlayer exchange coupling as compared to their uniaxial anisotropy strength. In the limit of a large  $K/J$  ratio, the model suggests a direct phase transition from paramagnetism to metamagnetism, with a saturated magnetization at remanence up to the blocking temperature  $T_B$  of the 2D ferromagnet base unit, with an upper bound given by the critical temperature of the magnetic base unit. Our study highlights the importance of the magnetic anisotropy to control the formation of different magnetic states, that is important to stabilize topological electronic phases.

### 8.3.2 Perpendicular magnetic anisotropy

The perpendicular anisotropy of Mn-based MBT layered crystals probably results from the crystal-field environment of Mn atoms, which is an elongated octahedron with an in-plane cubic symmetry and an out-of-plane uniaxial symmetry. As a consequence, the local anisotropy is mostly controlled by Te-Mn-Te dumbbells that favor a perpendicular magnetic anisotropy. This mechanism dominates in thick samples, and preliminary measurements already showed that there is still room for improvement if structural disorder can be reduced [post-growth annealing can for instance increase the blocking temperature of the metamagnetic phase from 3 K to 5 K]. Particularly important for thin layers, the interface anisotropy could be increased in well-chosen heterostructures.

#### 8.3.2.1 Optimization of bulk crystals

From structural analysis, it is now known that Mn-based MBT have a significant degree of intermixing between nearest-neighbor Mn-Bi pairs. This naturally leads to a decrease of the perpendicular anisotropy. Present efforts of our chemist colleagues are dedicated to :

- Optimize the growth conditions, so as to reduced the density of Bi-antisites ;
- Grow Sb-doped MBT compounds, in order to reduce the bulk carrier density ;
- On a longer term, consider the doping with rare-earth elements that can substitute cations without charge doping.

In MBT compounds, the exact nature of the exchange interaction remains elusive (superexchange, carrier-mediated,...), although there are signs that the magnetic properties are “preserved” in ultra-thin layers. A better understanding based on optimized crystals, or controlled electric/magnetic doping, would also shed some new light on the different mechanisms that can modify the anisotropy (possible contribution from the exchange anisotropy).

#### 8.3.2.2 Planar heterostructures

Another way to better understand the microscopic origin of the magnetic anisotropy is to modify this property by creating an interface with the magnetic topological insulator and another insulator with a large anisotropy and/or a higher critical temperature. For instance, it was recently predicted that a monolayer of  $\text{CrI}_3$ , a 2D ferromagnetic insulator with a perpendicular anisotropy and  $T_C = 45$  K, will have a strong exchange coupling at its interface with the first septuple layer of  $\text{MnBi}_2\text{Te}_4$  [76] and could be useful to stabilize

the QAH in  $\text{MnBi}_2\text{Te}_4$  thin films at temperatures well beyond the Néel temperature of the magnetic topological insulator. Furthermore, the overlayer insulator can have a large enough energy gap to add an electrical top gate

Such an engineering of the magnetic interface is a very promising route to design and optimize some functional topological devices, but it will of course require some advanced fabrication techniques in order to preserve the quality of the interface. This could be done by transfert methods under Argon atmosphere, as used for transition-metal dichalcogenides heterostructures, or by the direct van-der-Waals epitaxy of the heterostructure.

## 8.4 Topological phases

A specific physical property of a strong 3D topological insulator is the existence of a quantized magneto-electric coupling term (axion electrodynamics) that is “hidden” by the opposite chirality of opposite surfaces.

In an antiferromagnetic topological insulator with a perpendicular magnetization at each interface, surface states are gapped but the overall Hall conductance depends on the relative orientation of the interface magnetizations, giving either an axion insulator (Chern number  $C = 0$ ) or a QAH insulator (Chern number  $C = 1$ ). In addition to the topological magnetoelectric effect, dissipationless chiral edge modes exist at each surface step that defines an interface between the two different insulators.

In remarkable theoretical contributions [69–71], it was predicted that a material with topological properties and a colinear magnetic order with a perpendicular anisotropy can realize many different topological phases. For  $\text{MnBi}_2\text{Te}_4$ , depending on the dimensionality (2D for ultra-thin layers, 3D for thicker multilayers), but also on the local magnetization at interfaces, eight different topological states were identified (Fig. 8.3a). Furthermore, novel topological phases were also predicted in the canted state [73], a magnetic state that can be induced by rather small fields (with a limitation due to the anisotropy only for materials with  $N \geq 1$ ), with tunable band gaps and topological quantum phase transitions.

For a single material...this is quite a lot !

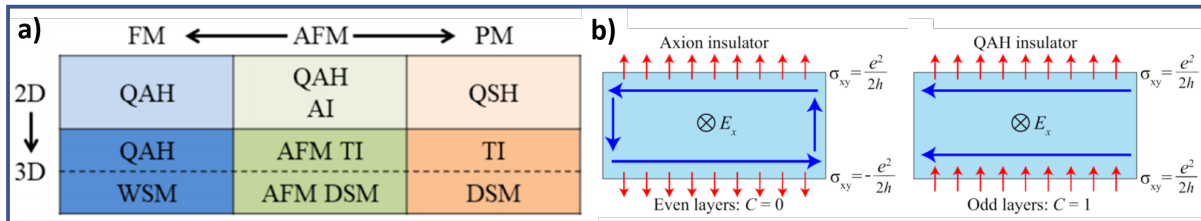


FIGURE 8.3 – **a)**, The eight different quantum states predicted in  $\text{MB}_2\text{T}_4$ , depending on their magnetic state (ferro FM, antiferro AFM and paramagnetic PM). For thin films (2D) : QAH, AI (axion insulator), QSH. For thicker films (3D) : AFM TI, TI, Chern insulator, Weyl or Dirac semimetal ; **b)**, Even/odd parity effect of gapped surface states with a half-quantized Hall conductance, giving either an AI or a QAH state. After [70].

The basic unit itself, the single  $\text{MnBi}_2\text{Te}_4$  septuple layer, is a trivial insulator and a 2D ferromagnet with perpendicular anisotropy. But for two or more layers, the axion electrodynamics modifies the topological nature of the gap.

Recently, remarkable signatures of the quantization were indeed observed in ultra-thin layers of  $\text{MnBi}_2\text{Te}_4$  exfoliated from bulk crystals, with the report of the quantum anomalous Hall effect [77, 78] and even of the axion insulator phase [79]. In addition, it was possible to tune the quantized magneto-electric response by applying a back-gate voltage. Still, some results are rather complex and very intriguing, in particular when considering the energy and magnetic field dependence of the transverse resistance. It will also be important to clarify the nature of the hysteretic behavior and the interplay between the outermost layers and others.

In this project, we aim at reproducing these results with ultra-thin layers of  $\text{MnBi}_4\text{Te}_7$  and try to evaluate the influence of the uniaxial magnetic anisotropy on the temperature stability of the QAH state, as well as its evolution in the canted regime. Another important issue will be to clarify the energy dependence of the quantized response and the influence of the residual bulk conductivity. For this reason, we are already trying to work with Sb-doped MBT samples. We also remark that a comparative study of the 124 and 147 compounds is always interesting since the surface terminations can be very different, and also since outermost layers are crucial to stabilize the QAH state, as well as a possibly stronger surface anisotropy.

## 8.5 ...and beyond

Because van-der-Waals layered magnetic compounds have rather small exchange interactions, these crystals offer a very interesting playground, so as to test theoretical grounds describing the interplay between (topological) electronic states and magnetism in quantum materials, beyond the existence of topological phases. Different from the case of most antiferromagnets, their magnetization can be “manipulated” by 3D-vector magnetic fields, with rather small magnetic fields and all over the Bloch sphere.

As for other magnetic crystals, the influence of the Berry phase can be also probed in the metallic regime of bulk MBT compounds. This could be studied by considering the anomalous Hall response or the planar Hall effect. A more direct technique would be to investigate quantum interference in nano-Hall bars, as previously done with  $(\text{Ga},\text{Mn})\text{As}$  [31].

Conversely, magnetic textures, such as domain walls, can also be expected to add a spin-dependent phase to coherent quasiparticles propagating in the topological insulators. A domain wall will separate surface domains with opposite magnetizations. Bulk and surface quasi-particles will behave differently. In the latter case, a narrow domain wall (large anisotropy) could behave for chiral edge state as equivalent to a quantum point contact for QHE edge states, with a tunable transmission if its width can be modified (by a constriction or a side gate for instance).

Back to on-going studies and opened questions about the magnetic properties of the  $[\text{MnBi}_2\text{Te}_4][\text{Bi}_2\text{Te}_3]_N$  family, there are two important aspects that deserve some specific consideration : i) the nature of the inter-layer exchange interaction (and the possibility to change it to a ferromagnetic coupling), and ii) the study of the anomalous Hall effect.

### 8.5.1 Inter-layer exchange coupling

Ab-initio calculations considered the case of compensated magnetic topological insulators, for which the dominant magnetic coupling is due to (high-order) super-exchange interactions [68, 70]. For the inter-layer exchange interaction, both antiferromagnetic or ferromagnetic couplings are possible, also in synthetic heterostructures [72]. However, in most “real” materials, the conductivity is that of highly-degenerate semiconductors, also in the direction transverse to van-der-Waals planes, so that carrier-mediated exchange interactions could also play a non-negligible role.

As suggested by theory, the hysteretic behavior of MBT compounds is often interpreted in term of a 3D ferromagnetic phase. Recently, we gave evidence that this is not the case for the  $[\text{MnBi}_2\text{Te}_4][\text{Bi}_2\text{Te}_3]_N$  family, although other materials could actually undergo a true phase transition to a ferromagnetic state.

Presently, it is difficult to predict the magnetic phases of layered MBT materials. To better understand the mechanisms at work, it will be necessary to study :

- The influence of the chemical potential. Sb-doped Mn-based MBT samples will allow us to tune the bulk residual density, and clarify the nature of the inter-layer exchange interaction ;
- The perpendicular anisotropy. Bulk effects (reduced Mn/Bi intermixing, chemical doping with isovalent rare-earth elements) ; Surface anisotropy.

Based on our initial studies, we aim at investigating in detail the thickness dependence of the magnetic properties (bulk vs. surface anisotropy) and we also consider to enhance the surface anisotropy by combining the magnetic topological insulators with highly-anisotropic 2D ferromagnets.

### 8.5.2 Anomalous Hall effect

Another important aspect that remains rather overlooked is the nature of the anomalous Hall effect in bulk MBT crystals (or thick layers). As for other magnetic crystals with disorder, there can be two contributions, the extrinsic one due to impurities and the intrinsic one due to the Berry phase. On the one hand, these layered materials do combine topology and magnetism, so that a large Berry-curvature contribution can be expected from their inverted band structure. On the other hand, there is a finite degree of disorder, which can not only contribute to the extrinsic AHE but can also average out the intrinsic AHE component.

We remark that the AHE in  $\text{MnBi}_2\text{Te}_4$  was successfully tuned by a gate voltage in ultra-thin layers by two groups [77, 80], both showing a sign reversal, which actually could be the signature of a dominant spin contribution at the Te-Mn-Te-Bi-Te “interface” (as observed for  $\text{MnTe}/(\text{Bi,Sb})\text{Te}$  heterostructures [81]).

To better understand this problem, it will be interesting to investigate the evolution of the AHE with the electro-chemical potential in structures exfoliated from Sb-doped Mn-based MBT samples, for different compositions, so as to modify the chemical potential to a nearly-compensated regime and finally tune the potential with an electrical gate. Also important, it will be possible to compare the results obtained in canted antiferromagnets with those measured in the homogenous metamagnetic state with a tilted magnetic field.

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## Résumé

Mon activité scientifique porte principalement sur l'étude des phénomènes quantiques en nanoélectronique et nanomagnétisme, notamment en basse dimension dans des nanostructures, et particulièrement des propriétés de magnéto-transport dépendant du spin de nouvelles phases topologiques (isolants topologiques, semi-métaux de Weyl,...).

Ce manuscrit décrit mes travaux de recherche sur le transport de charges dans des nanostructures d'isolants topologiques 3D, avec la mise en évidence de la diffusion anisotrope des états de surface (fermions de Dirac 2D texturés de spin) et, surtout, l'étude du confinement transverse dans des nanofils quantiques et du transport quasi-1D quasi-balistique, malgré un fort désordre intrinsèque.

Le projet scientifique s'articule autour de deux stratégies permettant de réaliser le transport balistique 1D, par des canaux de bords texturés de spin : les modes de surface confinés dans des nanofils quantiques 3D TI, et les canaux de bord dans des isolants topologiques magnétiques. Dans ces derniers, la nature de la structure de bande pourrait être contrôlée par l'aimantation, manipulable en champ magnétique faible, voire par une grille électrostatique. Un objectif est la réalisation de conducteurs balistiques chiraux, d'intérêt pour les technologies de l'information classique et quantique (interconnexion,...).

**Mots-clés:** nanostructures, isolants topologiques, magnétisme, électronique de spin.

## Abstract

My experimental research activities focus on spin-related quantum phenomena in magnets and low-dimensional conductors, mostly studied by electrical transport measurements at very low temperatures and in 3D vector magnetic fields. In particular, novel quantum electronic phases are investigated in nanostructures of topological materials, such as strong 3D topological insulators (3D TIs) induced by spin-orbit coupling, for which the influence of the geometric Berry phase can lead to spin-textured gapless quasi-particles.

In this dissertation, a first part gives an exhaustive review of the results obtained on nanostructures of *disordered* 3D topological insulators, from nanoflakes to nanowires. Quantum transport measurements are used to evidence some unique properties of spin-helical 2D Dirac fermions, due to their anisotropic scattering by disorder. In quasi-1D quantum wires, confinement in reduced dimension leads to quasi-ballistic transport, with a hollow-conductor geometry as for carbon nanotubes and semiconducting core-shell nanowires, but with strong disorder. The second part describes my on-going research project, with the aim to investigate 1D dissipationless spin-polarized chiral edge states. Two directions are followed : i) the search for the perfectly-transmitted mode in (Bi,Sb)Te nanowires or in core-shell 3D TI heterostructures ; ii) the search for the quantum anomalous Hall and axion insulator regimes (among other topological phases) in novel magnetic topological insulators of the  $\text{MnBi}_2\text{Te}_4$  family, and related materials.

**Keywords:** nanostructures, topological insulators, magnetism, spintronics.





