

Topological Insulator with Micro-Grating Structures

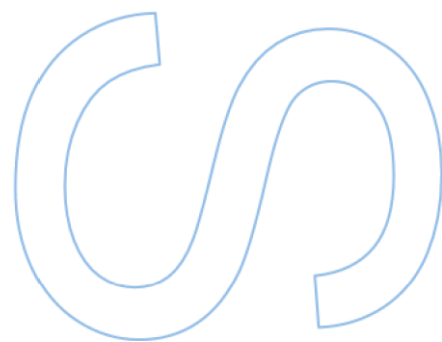
Carlos Eduardo Roque

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Department of Physics and Astronomy

Faculty of Sciences of University of Porto

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Carlos Eduardo Roque

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Supervisor

Prof. Dr. André M. Pereira, Faculty of Sciences of University
of Porto

Co-supervisor

Dra. Ana L. Pires, Faculty of Sciences of University of Porto



“ O caos é uma ordem por decifrar ”

José Saramago in *“O Homem Duplicado”*

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Resumo

Semicondutores não magnéticos, que servem como base à maioria dos dispositivos eletrônicos, dependem da eletrônica para processar informação. Embora tenham impulsionado as bases para os avanços técnicos das últimas décadas, existe atualmente uma enorme necessidade de tecnologia mais rápida, mais fiável e menos consumidora de energia. O sobreaquecimento, a dissipação de energia e as restrições à miniaturização destes materiais contribuem para o paradigma atual, conduzindo a um limite fundamental dos dispositivos eletrônicos tradicionais.

A Spintrónica, a disciplina da física que estuda como podemos utilizar o grau de liberdade do *spin* para recolher, transportar e processar informação, avança com uma solução promissora para todos estes problemas. Deste modo, esta área emergente abre a porta a tecnologias magnéticas inovadoras, revolucionando o armazenamento de dados, a computação e sensores. Dispositivos neste domínio são versáteis e podem integrar outros campos da física para melhorar a sua eficiência. Neste trabalho, propomos que a Plasmonica pode introduzir um grau adicional de liberdade nos dispositivos de spintrónica, com potencial para revolucionar o seu design e funcionalidade. Ao integrar materiais quânticos, como Isoladores Topológicos, e excitando modos plasmónicos nos mesmos, é possível remodelar o paradigma da tecnologia spintrónica, particularmente aplicando esta tecnologia a circuitos ópticos integrados.

Tendo isto em consideração, este trabalho foca-se, em primeiro lugar, na fabricação de estruturas de *grating* plasmónicas através de *thermal nanoimprint lithography* (T-NIL), utilizando como substrato termoplástico polimetilmetacrilato (PMMA). Fabricámos com sucesso estruturas de *grating* com periodicidades de $p=(0.725 \pm 0.001) \mu\text{m}$ e $p=(1.500 \pm 0.001) \mu\text{m}$ e optimizámos o respetivo procedimento de fabricação. Seguidamente, depositamos Bi_2Te_3 , um material com uma fase topológica bem conhecida, e Ag no topo das estruturas, através de deposição *DC-magneto sputtering*.

Adicionalmente, uma vez que as ressonâncias plasmónicas dependem de vários parâmetros geométricos, estas estruturas foram simuladas numericamente com o software COMSOL Multiphysics para analisar potenciais fenómenos de ressonância plasmónicos e a sua dependência com a variação de tais parâmetros. Identificámos três possíveis picos plasmónicos a 450 nm, 571 nm e 683 nm, e estudámos a dependência da frequência destes com a variação de espessura de Bi_2Te_3 e Ag entre 5 e 90 nm. Posteriormente, medimos as nossas amostras por espectroscopia UV-VIS-NIR, variando a espessura de Bi_2Te_3 entre 5 e 107 nm.

Por fim, foram medidas as propriedades de transporte, nomeadamente a dependência da resistividade com a temperatura e a magnetocondutância, no intervalo de 3K a 300 K e de -8 T a 8 T, respetivamente. Os resultados revelaram uma competição entre os efeitos de Localização Fraca e Anti-Localização Fraca, provavelmente atribuída à desordem decorrente da natureza amorfa do sistema em estudo.

Palavras-chave: Spintrónica, Isoladores Topológicos, Plasmónica, Amorfo

Abstract

Nonmagnetic semiconductors, which serve as the foundation for most electronic devices, rely on electronics to process information. Although they laid the groundwork for the technical advances of the last few decades, there is now a huge need for faster, more dependable, and less energy-dissipative technologies. Overheating, power dissipation, and the restrictions of miniaturizing these materials all contribute to the current apparatus, resulting in a fundamental limit of traditional electronic devices.

Spintronics, the discipline of physics that studies how we may use the spin degree of freedom to collect, transport, and process information, provides a promising solution to all these problems. In this way, this emerging field opens the door to innovative magnetic-based technologies, revolutionizing data storage, computation, and sensing systems. Spintronic-based devices are versatile and can employ other fields of physics to improve their efficiency. In this work, we propose that Plasmonics can introduce an additional degree of freedom in spintronic devices, with potential to revolutionize their design and functionality. By integrating quantum materials such as Topological Insulators and exciting plasmon modes on them, it is possible to reshape the paradigm of spintronics technology, particularly applying this technology to integrated optical circuits.

Taking this into consideration, the present work focuses firstly on the fabrication of plasmonic grating structures through thermal nanoimprint lithography (T-NIL), using as a thermoplastic substrate polymethyl methacrylate (PMMA). We successfully fabricated grating structures with periodicities of $p=(0.725 \pm 0.001) \mu\text{m}$ and $p=(1.500 \pm 0.001) \mu\text{m}$ and optimized the respective fabrication procedure. This was followed by the deposition of Bi_2Te_3 , a material with a well-known topological phase, and Ag on top of the grating structures, by DC-magneto sputtering deposition.

Furthermore, since plasmonics resonances depend on several geometric parameters, these structures were numerically simulated with the COMSOL Multiphysics software to address potential plasmonic resonances and their dependence with parametric sweeping. We identified three possible plasmonic peaks at 450 nm, 571 nm, and 683 nm, and we studied their frequency shift upon thickness sweeping between 5 and 90 nm of both Bi_2Te_3 and Ag. Later, we measured our samples by UV-VIS-NIR spectroscopy, ranging their Bi_2Te_3 thickness between 5 nm and 107 nm.

Finally, transport properties were measured, specifically, focusing on the temperature dependence of resistivity and the magnetoconductance, within the range of 3 K to 300 K

and -8 T to 8 T, respectively . The results revealed a competition between Weak Localization and Weak Anti-Localization effects, likely attributed to the disorder arising from the amorphous nature of the system under investigation.

Keywords: Spintronics, Topological Insulators, Plasmonics, Amorphous

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Symbols

t	Thickness	nm
λ	Incoming Light's Wavelength	nm
l_ϕ	Phase coherence length	nm
ρ	Resistivity	$\Omega \text{ m}$
σ	Conductivity	$\Omega^{-1} \text{ m}^{-1}$
$\Delta\sigma(H)$	Magnetoconductivity	e^2/h
α	Hikami-Larkin-Nagaoka model prefactor	
T	Temperature	K
n	Refractive Index (real part)	
κ	Extinction Coefficient	
p	Periodicity	μm
D	Crystallite Size	nm
A	Absorbance	
R	Reflectance	
T	Transmittance	
λ_L	Plasmon Limiting Wavelength	nm
λ_c	Plasmon Excitation's Wavelength	nm
ϕ	Grating's Wall Angle	$^\circ$
d	Dimensionality of the System	

List of Abbreviations

AC Alternated Current

ARPES Angle-Resolved Photo-Emission Spectroscopy

DC Direct Current

EELS Electron Energy Loss Spectroscopy

EWFD Electromagnetic Waves, Frequency Domain

FDTD Finite-Difference Time-Domain

FEM Finite Element Methods

FFT Fast Fourier Transform

FM Ferromagnetic

FMR Ferromagnetic Resonance

GR Magneto-conductivity

HLN Hikami-Larking-Nagaoka Model

IFIMUP Institute of Physics for Advanced Materials, Nanotechnology and Photonics

IR Infra-Red

ISHE Inverse Spin Hall Effect

MBE Molecular Beam Epitaxy

MR Magneto-resistance

NIL Nanoimprint Lithography

NIR Near Infra-Red

NM Non-magnetic

PDMS Polydimethylsiloxane

PMMA Poly(methyl methacrylate)

PPMS Physical Properties Measuring System

PS Polystyrene

QHE Quantum Hall Effect

QSHE Quantum Spin Hall Effect

SCI Spin to charge Inter-conversion

SEM Scanning Electron Microscopy

SERS Surface Enhanced Raman Spectroscopy

SHE Spin Hall Effect

SOC Spin Orbit Coupling

SPP Surface Plasmon Polariton

TI Topological Insulator

TM Transverse Magnetic

TRS Time-Reversal Symmetry

T-NIL Thermal Nanoimprint Lithography

TSS Topological Surface State

UV Ultra-violet

UV-NIL Ultra-Violet Nanoimprint Lithography

VIS Visible

WAL Weak Anti-Localization

WL Weak Localization

XRD X-Ray Diffraction

XRR X-Ray Reflectometry

2DEG 2D Electron Gas

Preface

Motivation

Nonmagnetic semiconductors, which serve as the foundation for most electronic devices, rely on the carrier charge to process information. Although they laid the groundwork for the technical advances witnessed over the last few decades, there is now a huge need for faster, more dependable, and less energy-dissipative technologies. However, overheating, power dissipation, and the restrictions of miniaturizing these materials, as depicted in Figure 1, all contribute to the current apparatus, resulting in a fundamental limit for traditional electronic devices [1]. This evolution is more evident in Figure 1, which shows the progressive miniaturization of non-magnetic semiconductor devices over time, shrinking from 3 μm (1990) to 3 nm (2022).

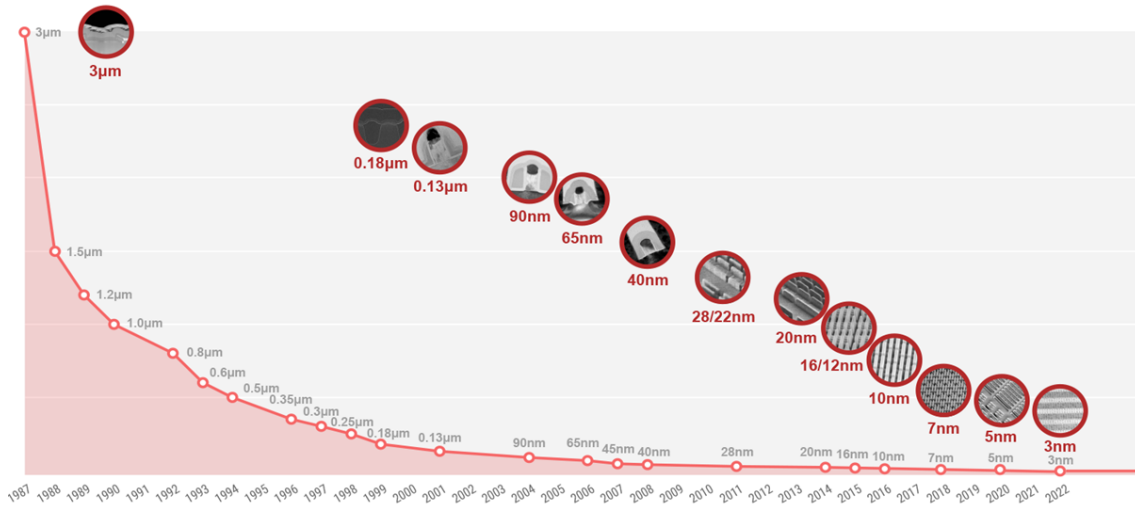


FIGURE 1: Representative Miniaturization of non-magnetic semiconductor devices over the last years. From ref. [2].

Spintronics, the field of physics that studies how we may use the spin degree of freedom to collect, transport, and process information, provides a promising solution to all these limits. This emerging field opens the door to innovative magnetic-based technologies, revolutionizing data storage, computation, and sensing systems. Spintronic devices are versatile and can employ other fields of physics to improve the efficiency of their magnetization response. Given the vast scope of this field, which encompasses a myriad of potential applications such as spin valves [3], magnetic tunnel junctions [4], magnonics

computing [5], among others, there exists a wealth of untapped synergies within the study of optics. A prime example is plasmonics, a specialized area that investigates the generation of plasmons. These quasiparticles embody collective electron oscillations in the conduction bands of metals, excited at specific resonant frequencies. Such plasmonic phenomena predominantly arise from the interplay between light and metal-dielectric interfaces [6]. This interaction between light and matter unveils a world of application when it comes to studying the next chip-scale technology [7], as it allows us to achieve higher operation speeds with smaller size devices. Figure 2 illustrates the operation speed as a function of critical dimensions of the plasmonics, photonics, electronics, and past technology.

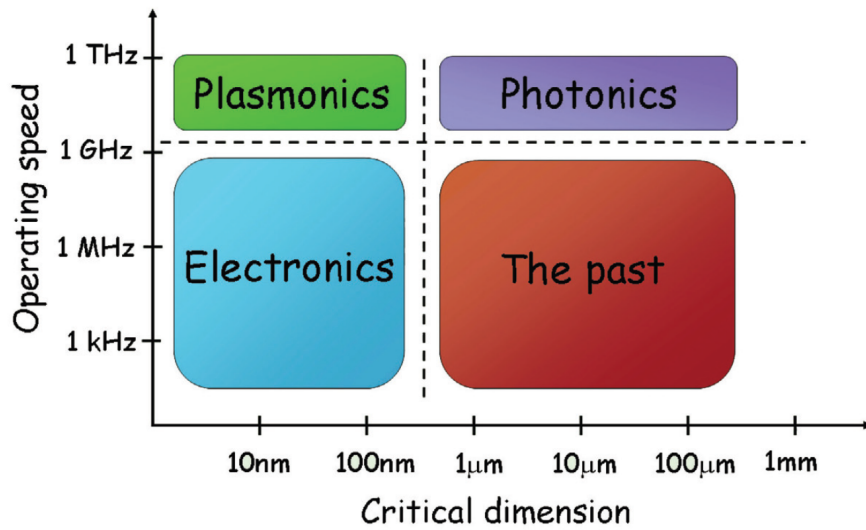


FIGURE 2: Comparison of photonics, electronics and plasmonics concerning the operating speed and critical dimensions for chip-scale devices. From ref. [7].

Plasmon modes exhibit several features, such as light confinement over nanometric volumes and resonance peaks over the optical spectrum, which can be tunable by manipulating the geometric properties of periodic structures. Moreover, interesting properties arise from these by applying an external magnetic field or even by exploring the magnetization of the material. Besides that, it is possible to excite these quasiparticles in Topological Insulators (TI) [8] and get even more exotic properties, since these materials by themselves also hold interesting quantum properties. Topological insulators are defined by having both insulating bulk and metallic protected surface edge states. By using them within the frame of plasmonics, it is possible to establish a link between the latter and spintronics, due to the spin-polarized plasmon modes excited on the material [9], opening in this way a path to a wide number of potential applications worth studying that could potentially revolutionize the technology we live with.

Research Objectives and Thesis Outline

The main objective of the proposal thesis is to study whether plasmonics modes in topological insulator micro-grating structures alter the already known quantum properties of the material, namely the observation of the Weak Anti-Localization (WAL) effect and the spin dynamics of the system. The topological insulator material chosen for this purpose is Bi_2Te_3 , since their topological features have already been studied within the research group and as there is a lack of studies within plasmonics in this material. This compound will be deposited onto micro-grating structures fabricated by nanoimprint lithography (NIL) on a polymethyl methacrylate (PMMA) substrate. The fabricated samples will be characterized by Scanning Electron Microscopy (SEM), X-Ray Diffraction, X-Ray Reflectivity, and UV-VIS-NIR Spectroscopy. By doing this, a deeper insight into the plasmonic events in the samples can be acquired. This approach will provide a clearer understanding of whether the Surface Plasmon Polaritons (SPP) waves excited on the structures enhance the spin-to-charge conversion mechanism or influence the Weak Anti-Localization (WAL) effect arising in the magnetoresistance data.

Communications

1. **Roque, Carlos E.**; Moreira, Mafalda; Pires, Ana L.; Pereira, André M.. "Control of Injected Spin Current into a Topological Insulator: Where Spintronics Meet Magnetoplasmonics". Poster presented in Magnetism in Portugal 2024 - School of Metrology in Magnetism, Lisbon, Portugal, 02/2024
2. **Roque, Carlos E.**; Fernandes, Bruno; Moreira, Mafalda; Pires, Ana L.; Pereira, André M.. "Micro-grating Structures of Topological Insulators: A Connection Between Spintronics and Plasmonics". Oral Presentation presented in DFA Week, Department of Physics and Astronomy, Faculty of Sciences of University of Porto, Porto, Portugal, 04/2024.
3. **Roque, Carlos E.**; Fernandes, Bruno; Moreira, Mafalda; Pires, Ana L.; Pereira, André M.. "Micro-Grating Structures of Topological Insulator and Gd: A Journey Through Spintronics and Magnetoplasmonics". Oral Presentation presented in IJUP - Encontro de Investigação Jovem da U.Porto, Porto, Portugal, 05/2024.
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Poster presented in 2024 IEEE Magnetics Society Summer School, Taipei, Taiwan, 06/2024,

5. **Roque, Carlos E.**; Moreira, Mafalda; Fernandes, Bruno; Pires, Ana L.; Pereira, André M.. "Micro-Grating Structures of Topological Insulator: From Plasmonics To Spintronics". Oral presentation presented in CMD31 - General Conference of the Condensed Matter Division of the European Physical Society, Braga, Portugal, 09/2024.
6. **Roque, Carlos E.**; Moreira, Mafalda; Fernandes, Bruno; Rodrigues, Ana Rita O., Pires, Ana L.; Pereira, André M.. "Does Plasmonics Influence Topological Surface States? A Case Study of Bi_2Te_3 ". Oral presentation presented in 2024 Around-the-Clock Around-the-Globe Magnetics Conference (AtC-AtG), Online, 10/2024.

1. State of the art

This chapter will discuss the main properties and phenomena exhibited by topological insulators and how these can be linked with plasmonics to explore exotic phenomena and develop new spintronics' technologies.

1.1. Topological Insulators

1.1.1 Origin

Topological Insulator (TI) research first emerged from an interest in the Quantum Hall Effect (QHE), discovered by Klaus von Klitzing in 1980 [10], and what materials are better to hold it. Topological Insulators represent a class of materials that exhibit the Quantum Spin Hall Effect (QSHE), the spin-polarized version of the QHE. The quantum spin hall states cannot be described in classical band theory by the symmetries of the quantum system [11]. Instead, these are defined by a set of properties, called topological invariants, that do not change as the system evolves adiabatically, such as the number of conducting edge modes, which is then observed as a quantized hall conductivity, as is the case in the QHE. A whole new realm of physical phenomena and discoveries became possible because of these effects. Throughout the subsequent decade, the mathematical concepts of topology were developed regarding the QHE and super-fluid phase transitions' description by David Thouless, Duncan Haldane, and Michael Kosterlitz, granting them the Nobel Prize in Physics in 2016 [12]. Following their work, Charles Kane and Eugene Mele proposed in 2005 a model for a graphene lattice with high Spin Orbit Coupling (SOC) [13] in which QSHE emerges. In this effect, the conducting edge states come as Kramers' doublets due to SOC, with one conducting state associated with spin up electrons and the other with spin down. The number of conducting edge states is proportional to the quantized hall conductance. The experimental realization of the QSHE was achieved in 2007 when studying a HgTe quantum well [14], a 2D TI material. Later, this discussion was extended to a new class of materials, the 3D TI, representing a topologically unique new state of matter.

1.1.2 Properties

The TI band structure has peculiar features. TI exhibit isolating properties in the bulk together with topologically protected metallic surface states [15]. These are described by

a 3D Dirac Hamiltonian, as stated in equation 1.1, where $\hbar$ is Planck's reduced constant, v_F is the Fermi velocity along the surface, and $\vec{\sigma}$ is the vector of the Pauli matrices.

$$\mathcal{H}(\vec{k}) = \hbar v_F \vec{k} \cdot (\vec{\sigma} \times \hat{z}) \quad (1.1)$$

Due to the spin-moment locking of the states, which emerges directly from the Dirac formalism, the surface states are protected by time reversal symmetry (TRS). This description also means the edge states are spin-polarized: electrons with opposite spins move in opposite directions, and travel without backscattering [16], as illustrated in Figure 1.1 (A). One way of explaining how these edge states emerge is through bulk band inversion and consequent hybridization in the bulk due to SOC. In TI materials the bandgap closes at high symmetry points of the Brillouin zone in a structure called Dirac cone. This structure, can be observed by Angle-Resolved Photo-Emission Spectroscopy (ARPES) measurements and exhibits chiral spin texture. An illustration of the Dirac cone is presented in Figure 1.1 (B).

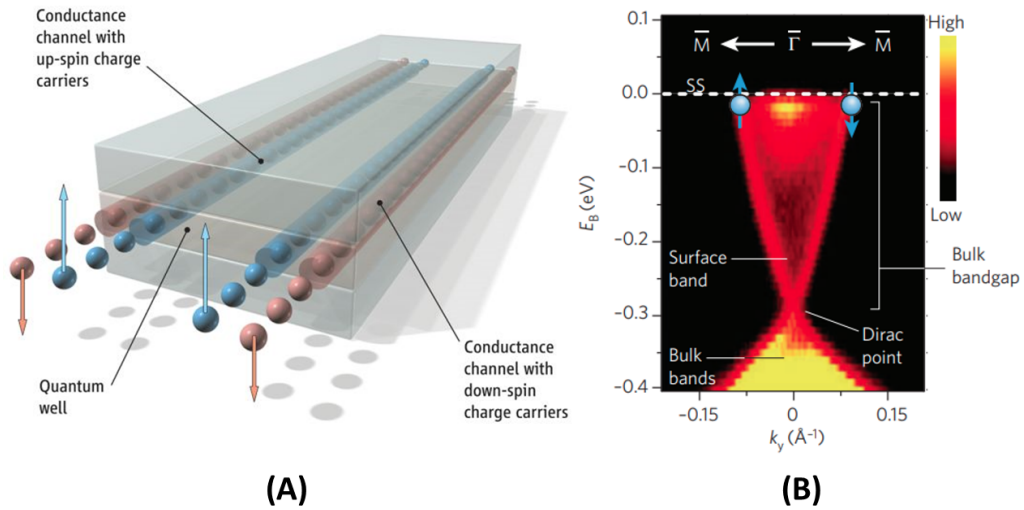


FIGURE 1.1: (A) Illustration of the conducting edge states with opposing spins in QSHE and (B) data of the Bi_2Se_3 ARPES measurement showing the cross-section of its band structure, where at the Γ points within the first Brillouin zone, the band gap closes in a Dirac cone. From ref. [14] and [15] respectively.

The presence of surface states on this class of materials may lead to the observation of the weak anti-localization (WAL) effect, which is typically observed at cryogenic temperatures in materials with high SOC, when the phase coherent length l_ϕ is larger than the mean free path of electrons. In this way, the electrons can go through multiple scattering phenomena while preserving their phase rather than being dephased. When this is the case, due to the constructive interference of the electron's wavefunctions, the WAL

effect arises [17], leading to a significant decrease in the electrical resistance of the sample. The quantum correction disappears when magnetic fields are applied [18], since external magnetic fields destroy TRS and introduce a phase shift between partner closed paths. Coincidentally, it is the application of a magnetic field that allows the effect to be observed as a characteristic V-shaped cusp centered at zero field in magnetoresistance measurements. Crucial information can be extracted from these curves by using the Hikami-Larkin-Nagaoka (HLN) model [19]. This model predicts theoretically the positive correction to the longitudinal magneto-conductivity due to the WAL effect, stated in equation 1.2, where $B_\psi = h/8e\pi(l_\phi)^2$ is a characteristic magnetic field that depends on the phase coherence length, Ψ is the digamma function, α is the coefficient that distinguishes the type of localization ($\alpha=1/2$ for each spin channel in WAL) and H is the applied magnetic field.

$$\Delta\sigma(H) = \frac{\alpha e^2}{\pi h} \left(\ln \left(\frac{B_\psi}{H} \right) - \Psi \left(\frac{1}{2} + \frac{B_\psi}{H} \right) \right) \quad (1.2)$$

The α parameter is expected to be equal to -1 in 3D TI, holding contributions of both decoupled surface states [18]. However, it has been found values of $\alpha = -0.5$, which is commonly associated with indirect coupling between surface states mediated by bulk carriers [20]. When considering the Weak Localization (WL) effect, characterized by the negative correction to the longitudinal magnetoconductivity, we expect values of α between 0 and 1/2 [21]. The phase-coherence length, on the other hand, is related to the level and origins of dephasing disorders in the system. Its magnitude enables us to address the quantity of phase-shifting scattering centres, and it follows a dependence with temperature of $L_\phi \propto T^{-p/2}$. The p parameter allows us to identify which dephasing mechanism dominates the system, whether it is 2D ($p = 1$) or 3D ($p = 2$) electron–electron interactions (EEI), or even electron–phonon interactions ($p=3$) [18]. Ideally we would expect decays of $L_\phi \propto T^{-0.5}$, which would mean that the surface states are dominating the conduction channels, however decay relations of $L_\phi \propto T^{-0.75}$ are commonly found, associated with 3D electron scattering. Therefore, the bulk can indeed contribute to these mechanisms, making it more difficult to differentiate the signal from the surface states, and therefore, the HLN model had to be re-adapted [20]. Equation 1.3 includes a new term that is added in order to take these considerations into account, acting as a background term.

$$\Delta\sigma(H) = \frac{\alpha e^2}{\pi h} \left(\ln \left(\frac{B_\psi}{H} \right) - \Psi \left(\frac{1}{2} + \frac{B_\psi}{H} \right) \right) + \beta H^2 \quad (1.3)$$

This β parameter takes not only into account classic cyclotronic $\beta_c = -\mu_H^2 G_0$ contribution, with μ_H being the Hall mobility and G_0 the zero-field conductance, but also quantum contributions β_q associated with spin-orbit scattering and elastic scattering events, defined in equation 1.4, where B_{SO} , B_e and B_ϕ are related with the spin-orbit relaxation length, mean free path of the electrons, and the decoherence length [17].

$$\beta_q = -\frac{e^2}{24\pi h} \left(\frac{1}{B_{SO} + B_e} \right)^2 + \frac{3e^2}{48\pi h} \left(\frac{1}{(4/3)B_{SO} + B_\phi} \right) \quad (1.4)$$

However, the observation of this effect does not necessarily imply the presence of surface states, since it is only considered as a signature emerging in high-SOC materials. Moreover, the absence of this effect does also not imply the non-existence of surface states, since only a ARPES measurement could probe that. In the case where it is not possible to perform this technique, additional characterization could be made in order to address other possible signatures of the surface states, such as the SdH oscillations [22]. The field of TI has grown quickly, namely in 3D structures, since the fabrication of 2D TI is constrained to quantum well heterostructures of small band gaps (≈ 4 -10 meV) which, therefore, show high bulk conductivities [23]. As the material needs to hold high SOC to exhibit quantum spin Hall states, alloys of heavy elements are in the spotlight within this field of research. The first ones to be discovered were the $\text{Bi}_x\text{Se}_{1-x}$ family of compounds [24] in 2008, leading to the formation of the first generation of 3D TI. In the years afterwards, materials from the same elements, such as Bi_2Te_3 , Bi_2Se_3 , Sb_2Te_3 were also studied as also 3D TI, leading to the second generation of these materials. Since then, the (Bi,Sb)-Te alloys continue to be a hot topic within this field as they are already extensively studied as room temperature thermoelectric (TE) materials [25].

1.1.3 Spin to Charge Interconversion Mechanisms in Topological Insulators

Given their high SOC, TI are also known for holding both the Spin Hall Effect (SHE) and its inverse effect (ISHE). The SHE is a spin to charge conversion mechanism, thus, by applying a charge current to the material, it is generated a spin current in the perpendicular direction. What happens within the material for this phenomenon to occur is that the electrons injected, that display both spin up and down, will interact with the lattice and electronic structure of the material. Due to intrinsic and extrinsic mechanisms, such as skew-scattering and side-jump contributions [26], the electrons will be deflected, and this deflection will occur in opposite directions depending on the spin polarization of the electrons [27]. In this way, spin up and spin down electrons will be localized in the sample

edges. However, as we are dealing with non-magnetic samples, the spin populations of either spin up and down polarizations will be the same, therefore, there won't be a charge unbalance within the sample; instead, there will be spin accumulation on both edges, which creates a spin current, as illustrated in Figure 1.2 (A).

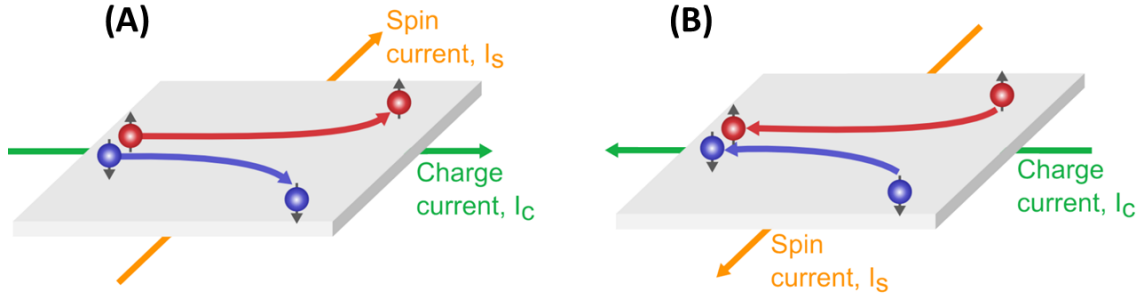


FIGURE 1.2: Schematics of spin to charge interconversion mechanisms present in high SOC materials: SHE (A) and ISHE (B). From ref. [28]

Since there is no charge unbalance, it is not possible to measure any SHE voltage, which adds a difficulty in studying these types of effects. Therefore, new ways of addressing this phenomenon that do not rely on electronic physical behaviour were advanced, and the first attempts to measure SHE successfully were by taking advantage of light interaction with the material, for example, using Scanning Kerr rotation measurements [29].

The SHE was first proposed in 1971 by Mikhail Dyakonov and Vladimir Perel [30], followed by its inverse effect in the following year. The ISHE is a spin to charge conversion mechanism, where pure spin currents (with opposite spins moving in opposite directions) are scattered and deflected towards the same direction due to the same mechanisms within the SHE. In this way, there will be charge accumulation on one side of the sample, creating a charge current. This charge current can be detected as a measurable ISHE voltage with a nano-voltmeter [26]. Figure 1.2 (B) shows a scheme of the inverse effect, and equation 1.5 expresses both these effects in terms of the spin hall angle θ_{SH} , which quantifies the efficiency of the conversion process [31], the charge current density $\vec{J}_c$, the spin current density $\vec{J}_s$, the carrier's spin direction $\hat{s}$, which is perpendicular to both the spin and charge currents, the charge of the electron e , and the Planck's reduced constant $\hbar$.

$$\text{SHE : } \vec{J}_s = \theta_{SH} \frac{\hbar}{2e} \vec{J}_c \times \hat{s} \quad \text{and} \quad \text{ISHE : } \vec{J}_c = \theta_{SH} \frac{2e}{\hbar} \vec{J}_s \times \hat{s} \quad (1.5)$$

These effects arise in many materials holding high SOC, but one advantage of using TI is that θ_{SH} appears to be 10 to 1000 times bigger in these materials, namely in Bi_2Se_3

[32, 33]. In order to induce pure spin currents within the Topological Insulator (TI), it is imperative to address the concept of spin injections, a phenomenon extensively examined in the context of TI [34]. One of the ways spin injection can occur is by placing a ferromagnetic (FM) material with a non-magnetic (NM) material, and applying an electrical voltage across them. In this scenario, electrons that possess spin polarization originating from the FM material exhibit a finite likelihood of migrating into the non-magnetic material. Therefore, as a response to the applied voltage, instead of only having a drift charge current $\vec{j}^{Q,Drift}$, there is a drift spin current $\vec{j}^{S,Drift}$, as defined in equation 1.6.

$$\begin{cases} \vec{j}^{Q,Drift} = \frac{\hbar}{2} \vec{e}_{spin} \frac{1}{-e} (\sigma_+ - \sigma_-) \vec{E} \\ \vec{j}^{S,Drift} = (\sigma_+ + \sigma_-) \vec{E} \end{cases} \quad (1.6)$$

Equation 1.6 takes into account the conductivity of both spin channels (σ_+ , σ_-), together with the applied electric field $\vec{E}$. A schematic representation of the spin injection is elucidated in Figure 1.3 (A) and (B).

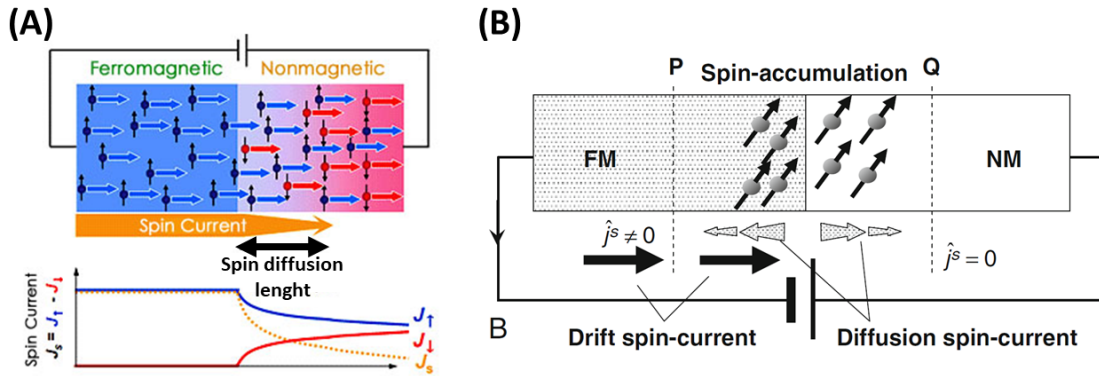


FIGURE 1.3: Schematics of a spin injection from a FM to a NM material (A) and spin scattering mechanisms that contribute to the spin diffusion length (B). From ref. [35] and [36]

From Figure 1.3 (B) and defining two points P and Q that are located far away from the interface between the FM and NM material, at the P point there is a well-defined spin current, since it is located in the ferromagnet. At the point Q, it is assumed that the spin current is zero, since during their flow, electrons will repeatedly scatter around scattering centres and will lose their original spin orientation in a process called spin flipping. Therefore, due to the conservation laws of the spin, there will be spin accumulation at the interface between the two materials [36]. This spin accumulation will not be solely localized at the interface, instead, it will diffuse to the adjacent portion of material. Therefore, there will also be a diffusion spin-current, as defined in equation 1.7, in which (D_+ , D_-),

(s_+, s_-) and (ρ_+, ρ_-) are the diffusion constant, spin density, and charge density for each spin channel.

$$\begin{cases} \vec{j}^{Q, Diffusion} = -D_+ \vec{\nabla} \rho_+ - D_- \vec{\nabla} \rho_- \\ \vec{j}^{S, Diffusion} = -D_+ \vec{\nabla} \vec{s}_+ - D_- \vec{\nabla} \vec{s}_+ \end{cases} \quad (1.7)$$

Once these electrons get to the NM layer, they will suffer spin relaxation and lose their spin polarized orientation at some point, while travelling away from the interface between the FM and NM materials [36]. The parameter that quantifies how much a spin current can hold its polarization into the NM material is the spin diffusion length λ_{spin} , defined in equation 1.8, in which $\tau_{\pm}$ is the spin relaxation time.

$$\lambda_{\pm} = \sqrt{D_{\pm} \tau_{\pm}} \quad (1.8)$$

For common materials studied in spintronics, this parameter ranges between 5 to 1000 nm. However, by taking advantage of the topological surface states (TSS) features in TI, this value can be increased to values bigger than $100\mu\text{m}$ [37]. Besides this method, there are other approaches that enable the injection of a spin current into the topological insulator, such as spin pumping [34].

To further amplify these properties, it is conceivable to couple these systems with other physical phenomena. Within the scope of the dissertation, focus was given on investigating the potential for enhancing spin-control mechanisms by studying plasmonics phenomena in TI.

1.2. Plasmonics Systems

Plasmonics, which stands as a prominent research focus in contemporary technological applications, has a historical foundation dating back to 1900, when Paul Drude introduced a model to describe the transport properties of conducting electrons in metals [38]. This model enabled us to calculate the dielectric function of metals, stated in equation 1.9, in which ω is the frequency, $\Gamma = v_F/l_e$ is the damping term, in which l_e is the electron's mean free path and v_F is the fermi velocity of the metal, and $\omega_p = \sqrt{ne^2/m\epsilon_0}$ is the bulk plasmon frequency, in which e is the charge of the electron, n is the electronic density, and m_e is the electron's mass.

$$\epsilon_r(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + \Gamma^2} + i \frac{\Gamma \omega_p^2}{\omega(\omega^2 + \Gamma^2)} \quad (1.9)$$

The optical properties of the metals can be derived from equation 1.9. This relation describes accurately noble metals in the Infra-Red (IR) regime. However, for the visible

range and higher energy bands, the experimental values differ from the numerical calculated results, as shown in Figure 1.4 for a gold bulk sample [39].

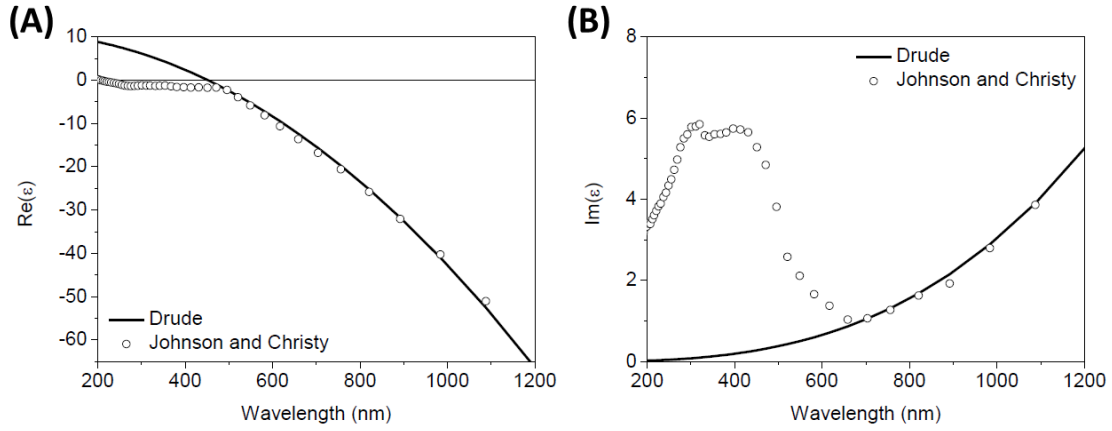


FIGURE 1.4: Theoretical results concerning the Drude model and experimental results (Johnson and Christy) of the real (A) and imaginary (B) part of the dielectric function of gold. From ref [40]

This deviation is due to how bound electrons answer to higher intensity radiation. These higher energy photons can excite the lower band's electrons into the conduction band, which is what will add a contribution to the dielectric function in the visible regime. In order to take into account these effects, the dielectric function can be adapted [39], as shown in equation 1.10, with γ being the damping constant of the metal and $\tilde{\omega}_p = \sqrt{\tilde{n}e^2/m^*\epsilon_0}$ being the plasma frequency with the inter-band transition's correction, in which $\tilde{n}$ is the electronic density of the bound electrons in the considered energy level and m^* is the effective mass.

$$\epsilon_r(\omega) = 1 + \frac{\tilde{\omega}_p^2(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2} + i\frac{\gamma\tilde{\omega}_p^2\omega}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2} \quad (1.10)$$

This theoretical model fits better the experimental data up to a wavelength of the incident light of 425 nm, as it can be seen in Figure 1.5 (A) and (B).

As the incident light used within the experimental studies discussed in this dissertation is characterized by a wavelength (λ) bigger than this limit, we will restrict our discussion to this correction. Drude's model opened the way for the proposal of the theoretical model describing the electron's collective coherent oscillations in the metal's band conduction in 1950, where plasmonic excitation modes arise [41]. When an electromagnetic wave is applied to a metal, longitudinal modes of electron collective oscillations, whose quanta are bosonic quasiparticles known as volume plasmons, can be excited when the condition $\epsilon(\vec{K}, \omega) = 0$ is met.

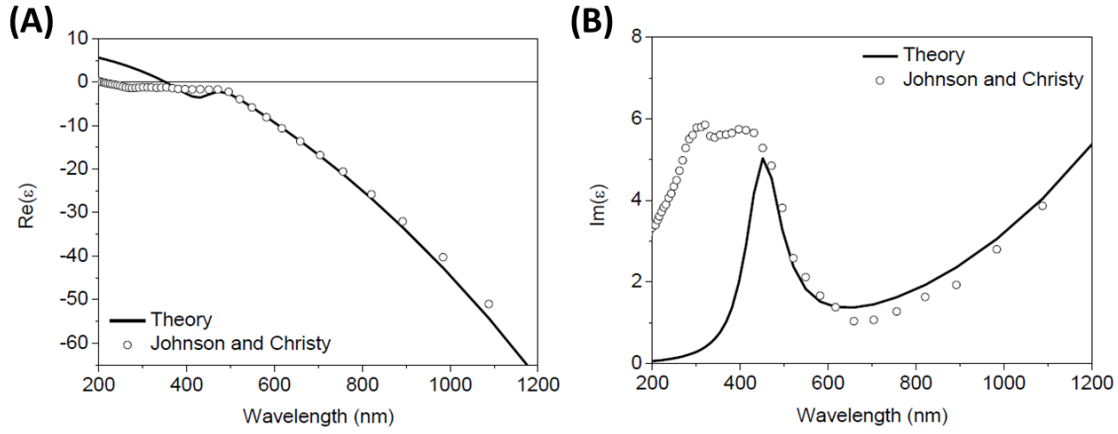


FIGURE 1.5: Theoretical results concerning the Drude's model and experimental results (Johnson and Christy) of the real (A) and imaginary (B) part of the dielectric function of gold by accounting the inter-band transitions. From ref [40]

These modes can only be excited by particle impact [6] and, once excited, remain localized and oscillate at the plasma frequency until their decay, either through Landau damping or collisional damping [42]. However, plasmonic modes can be excited at the surface by light when an incident photon couples with a plasmon and propagates throughout the surface of the material, which is the case for surface plasmon polaritons (SPP). This type of plasmon mode is illustrated in Figure 1.6 (A) and (B).

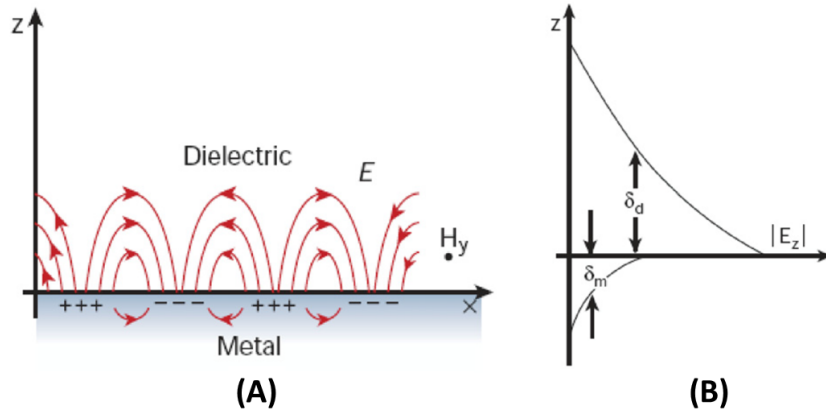


FIGURE 1.6: (A) In a SPP, the plasmon couples with an incident photon and propagates throughout the surface of the material until both decouple from each other; (B) The evanescent SPP wave exponentially decays into the metal and the dielectric medium. From ref. [43]

These are plasmonic modes excited by transverse magnetic (TM) polarized light that propagate in the perpendicular direction, at the interface of a dielectric and a metal [6], and are gaining popularity in the R&D community [44]. When considering the simplest plasmonic system, a metal and a dielectric, the dispersion relation of a SPP is given by equation 1.11, in which $\vec{k}_0$ is the wave vector of incident light, ϵ_m is the dielectric function for the metal, and ϵ_d is the dielectric function for the dielectric material, with graphical representation

schemed in Figure 1.7.

$$\vec{k}_{SPP} = \vec{k}_0 \sqrt{\frac{\epsilon_m \epsilon_d}{\epsilon_m + \epsilon_d}} \quad (1.11)$$

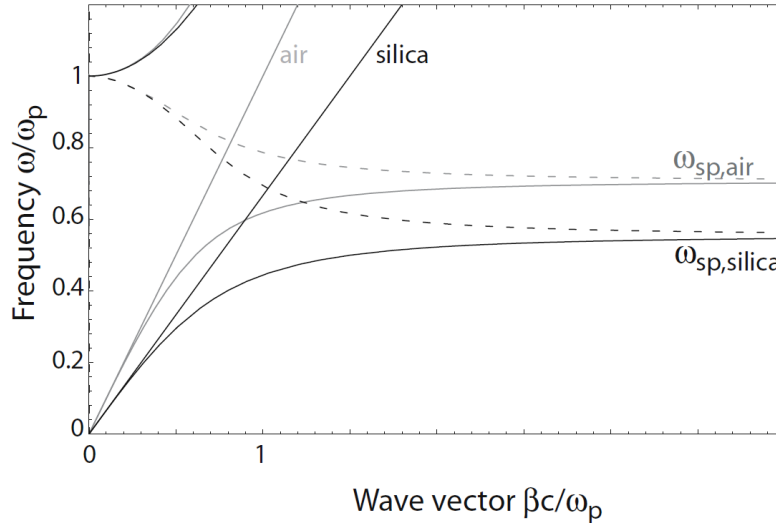


FIGURE 1.7: Graphical representation of the SPP's dispersion relation at the interface between a metal and a dielectric (silica or air) considering Drude's model. The dashed lines signalize the frequency of the surface plasmon for air and silica. From ref. [6]

For small wave vectors, thus for small frequencies (mid-IR or even lower), the dispersion relation of the SPP is close to the vector of the incident light at the light line. However, for large wave vectors and thus big frequencies, the frequency of the SPP reaches the surface plasmon frequency, as illustrated in Figure 1.7. This frequency is defined in equation 1.12.

$$\omega_{SPP} = \frac{\omega_p}{\sqrt{1 + \epsilon_d}} \quad (1.12)$$

This is true for the simplest systems. However, when considering multilayer systems of alternating dielectric and metallic layers, the conclusions are slightly different, since for every interface it is possible to have a bound SPP mode. If the separation between layers is of the same magnitude or even smaller than the decay length of the plasmon, the excited modes can couple. For example, by considering a dielectric-metal-dielectric system, the frequency of the SPP will depend on whether we are treating an odd or even mode, as stated in equations 1.13 and 1.14, in which $2a$ is the thickness of the metal layer, ω_+ is the frequency of the odd modes, and ω_- the frequency of the even modes.

$$\omega_+ = \frac{\omega_p}{\sqrt{1 + \epsilon_d}} \sqrt{1 + \frac{2\epsilon_d e^{-2ak_{SPP}}}{1 + \epsilon_2}} \quad (1.13)$$

$$\omega_- = \frac{\omega_p}{\sqrt{1 + \epsilon_d}} \sqrt{1 - \frac{2\epsilon_d e^{-2ak_{SPP}}}{1 + \epsilon_2}} \quad (1.14)$$

Plasmons can be used in a wide range of applications, ranging from biosensors, surface-enhanced Raman spectroscopy (SERS), solar cells, subwavelength optics (waveguides for example) and data storage [43]. Within these modes, the longitudinal oscillations of electrons interact with incident photons. Consequently, the resultant excited Surface Plasmon Polariton (SPP) waves exhibit an evanescent component extending into both the metal and the dielectric material. This behaviour is characterized by two distinct penetration depths: δ_d in the dielectric and δ_m in the metal. For example, for an interface of silver/silicon oxide, when exciting a SPP wave with a wavelength of 620 nm, $\delta_d=167$ nm and $\delta_m=23.6$ nm [43]. Another important parameter for characterizing these waves is the SPP propagation length δ_{SPP} , which is the distance beyond which the SPP wave intensity drops to 1/e of its starting value, which exhibits values of $2\ \mu\text{m}$ for aluminium when excited by a wavelength of 500 nm, for example [43]. These parameters are extremely important since they provide an estimate of the extent of plasmonic phenomena in the material.

The SPP will be propagated with the same momentum as the x-component of the incident light $k_x^{light} = k \sin \theta$, where θ is the angle of the incident light with the sample's surface. In order to excite these modes, the $k_x^{light} = k_{SPP}$ condition needs to be met. As schemed in Figure 1.8, the dispersion relations of both light and SPP do not cross, so it's crucial to use phase-matching techniques in order to excite these modes.

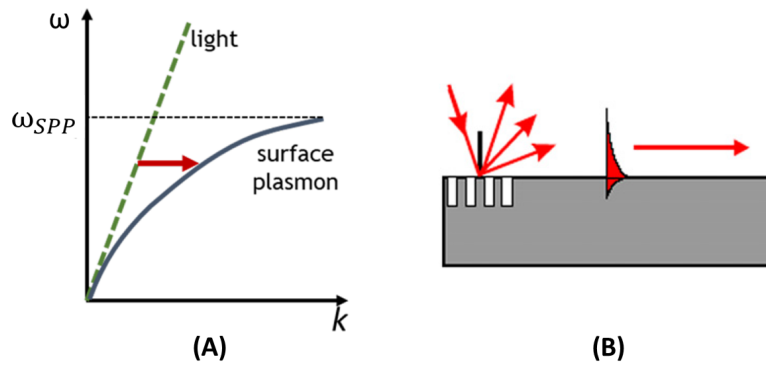


FIGURE 1.8: (A) The dispersion relations for a SPP and for light are dephased, so it is necessary to employ phase-matching techniques, such as grating coupling (B) to excite these modes. From ref. [45] and [43].

There are several ways of doing so, such as using the prism coupling method [46]. Nevertheless, this work will be focused on one method called grating coupling, which consists of fabricating grating metal structures with grating periodicity p , that can also be called the grating lattice constant [6]. In order to full-fill the condition $k_x^{light} = k_{SPP}$, the grating must obey the phase-matching condition, stated in equation 1.15, where $\nu = (1, 2, 3...)$,

$g = 2\pi/p$ is the reciprocal vector of the grating, and p is the grating periodicity.

$$k_{SPP} = k \sin \theta \pm \nu g \quad (1.15)$$

Equation 1.15, derived from Bragg's Law applied to this system, exemplifies what happens when light interacts with the grating structures. Several diffraction patterns are produced when light interacts with them, and these patterns are propagated away from the interface, creating evanescent modes at the interface. Therefore, these will couple with the surface plasmon [47]. This is an effective method [48] with an advantage in its implementation since it can be fabricated by relying on a very reproducible, low-cost, and scalable technique called Nanoimprint Lithography [49]. Manipulating the geometric properties of these structures, such as the period of the grating, is useful since it can change SPP properties, such as the damping of the plasmon mode Γ_p and its frequency ω_{SPP} [8], and it is easily accomplished with this technique, by using stamps differing in those parameters. Some examples of several of these structures are displayed in Figure 1.9.

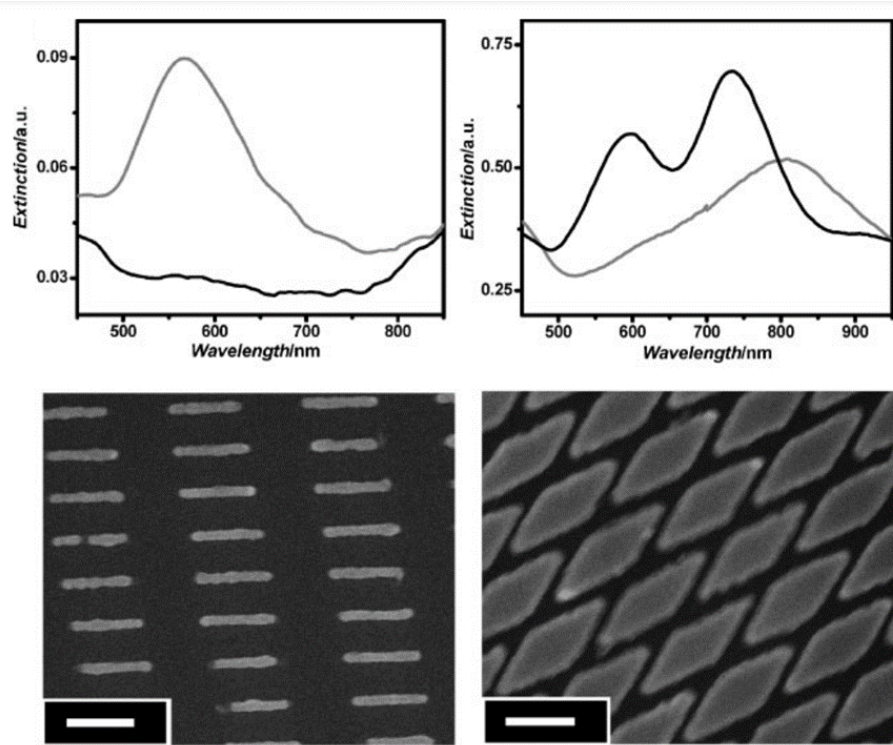


FIGURE 1.9: Stripe array (left) and diamond array (right) with respective extinction spectra for two orthogonal light polarizations (grey and black plots). Changing the structure's geometry changes the excited plasmonic modes. From ref. [50].

Exciting a Surface Plasmon Polariton (SPP) mode elicits a range of distinctive characteristics in the sample. These include the spatial confinement of electromagnetic fields and the emergence of resonant absorption peaks. Consequently, one can employ both direct and

indirect methods for detecting these excitation modes. These methods encompass optical spectrum measurements, Raman and Brillouin scattering spectroscopy, and electron energy loss spectroscopy. It's important to note that the manifestation of plasmonic phenomena is inherently linked to the choice of the metal in which these modes are induced. Noble metals, characterized by their low imaginary component in the dielectric function $\epsilon_2(\omega)$, traditionally occupy a prominent role in plasmonics applications. However, recent research efforts have extended beyond noble metals to explore materials such as Ga and Al [51]. Notably, there has been a surge in interest in Topological Insulator materials due to their intriguing and unconventional plasmonic properties [52].

1.2.1 Plasmonics in Topological Insulators

The recent interest in studying plasmonics in TI was motivated by the exotic behaviours theoretically predicted and experimentally observed in graphene [53]. Unlike the usual two-dimensional (2D) massive plasmon modes commonly observed in 2D electron gas (2DEG), 2D Dirac massless plasmon modes appeared in this 2D material. One way of distinguishing these two modes is by measuring their dispersion relation. In the case of massive plasmons $\omega_M(q)$ follows the dispersion relation as stated in the equation 1.16, with n_M being the electronic density of the massive carriers and m^* the effective mass. In the case of dirac massless plasmons, $\omega_D(q)$ follows the tendency of equation 1.17, with n_D being the electronic density of the dirac carriers, and g_s and g_v are respectively the spin and valley degeneracies (equal to 1 in Bi_2Se_3 [8]).

$$\omega_M(k) = \frac{1}{2\pi} \sqrt{k} \left(\frac{e^2}{2\epsilon_0\epsilon} n_M / m^* \right)^{1/2} \quad (1.16)$$

$$\omega_D(k) = \frac{1}{2\pi} \sqrt{k} \left(\frac{e^2}{4\pi\epsilon_0\epsilon\hbar} v_F \sqrt{2\pi n_D g_s g_v} \right)^{1/2} \quad (1.17)$$

This last mode was also theoretically [9] predicted and experimentally [8], observed for Bi_2Se_3 , an already known TI, as it is illustrated in Figure 1.10.

Despite exhibiting a playground of effects, plasmonics in TI have an advantage over graphene. Since the electrons from the TSS in TI exhibit spin-momentum locking, it's expected that their plasmons are spin-polarized and have longer propagation lengths [54], due to the absence of electrons' backscattering. Additionally, TI exhibits these interesting features in an unexplored frequency range within plasmonics: the terahertz region.

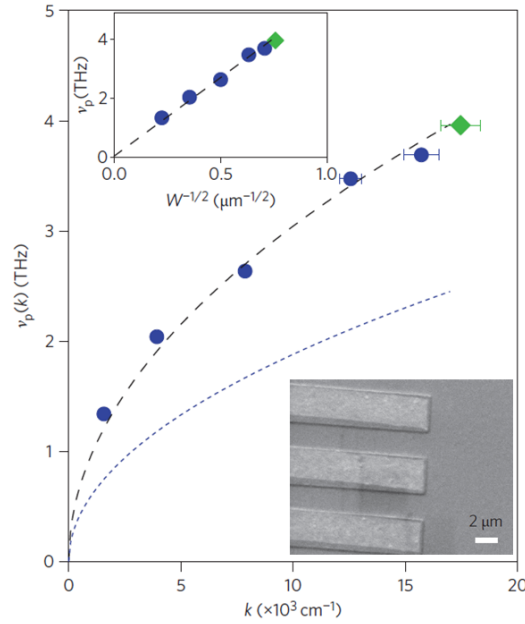


FIGURE 1.10: Theoretical dispersion relation of plasmons in micro-ribbon arrays of Bi_2Se_3 for Dirac (dashed black line) and massive (dashed blue line) plasmons. The experimental data (dots) follow the Dirac plasmon tendency. From ref. [8].

These resonances are tunable in frequency by changing geometrical properties such as the width of the nanostructures [8] and the film thickness [54]. However, we cannot conclude that the Dirac behavior of these plasmons is caused by the TSS because there has been no experimental evidence of TSS presence in micro-grating structures of topological insulators yet. Despite that, topological protection implies that the topological properties of the material should be maintained as long as there is no bandgap opening [55]. In addition, translational symmetry is not a necessary condition for the emergence of topological phases, and novel theories on amorphous topological insulators seem to confirm the idea that the limits of topological protection are much wider than initially thought [56]. There has been growing consensus that the topological surface states/properties may persist even in the absence of long-range order. As a matter of fact, it has already been studied that a topological phase emerged on Bi_2Se_3 amorphous systems [57]. Additionally, TI have already been fabricated onto various morphologies, such as nanoribbons, nanoplates, and nanowires [58], and their topological properties have been validated [59]. So, despite the lack of experimental confirmation of the presence of the TSS in TI grating structures, we have strong reasons to believe they are present and they should have an influence on this Dirac plasmon behaviour.

Furthermore, when considering thickness values typical of thin films, it is possible to excite plasmon modes both on the top and bottom of the film, which will electrostatically couple.

This will lead to the emergence of different behaviours in both optical and acoustic plasmon modes as schemed in Figure 1.11.

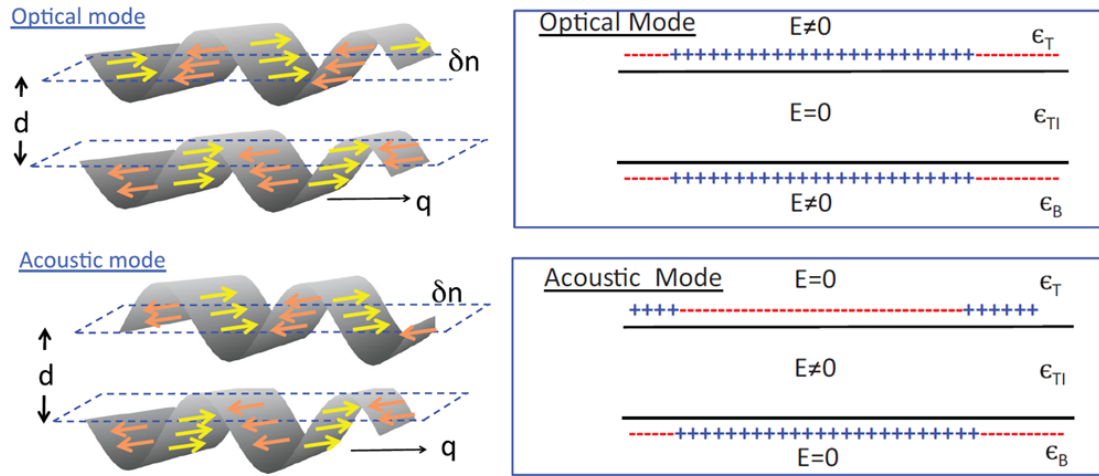


FIGURE 1.11: Schematics of the spin and charge dynamics in both optical and acoustic coupled plasmon modes, that will lead the emergence of pure "spin-like" and "charge-like" behaviours. From ref. [9]

An optical plasmon is a high energy mode with a wave vector dependence $\sqrt{k}$ at the long wavelength approximation, and it's the common mode found in most plasmonics studies and applications. Opposing this one, an acoustic plasmon is a low-energy mode that, instead of following the square root wave vector dependence, follows a linear tendency in its dispersion relation at the long wavelength approximation. These sound-like excitation modes are found in metal surfaces [60] and can affect phonon dynamics in the material [61]. It's predicted theoretically that in both of these modes, the charge density wave typical of the plasmon excitation occurs always with a transverse spin wave [9]. However, when considering the optical mode, these charge density waves are in phase and the transverse spin waves are out of phase. Due to these in-phase oscillations, the inside of the TI is not electrically polarized, but the surrounding environment is. Because of this, the optical modes are "charge-like", since these charge oscillations are observable and dominate over the spin ones. Opposing this mode, the acoustic one has charge density waves out of phase and in-phase spin oscillations. In this, due to the out of phase charge oscillations, there is a non-zero electric field in the inner of the TI, which will act just like a capacitor. This leads to a zero electric field outside the TI, which makes only the spin waves visible, which gives a "spin-like" behaviour to this mode [9]. The pure spin nature of this mode would completely change plasmonics by linking it to spintronics for the first time. These acoustic modes have already been observed in electron energy loss spectroscopy (EELS) measurements [62]. However, their pure spin nature has only been theoretically

confirmed. Concerning their experimental observations, there is not a concise conclusion as to whether these features were effectively observed or not. It has been suggested by ARPES measurement in Bi_2Se_3 a contribution of the so-called spin-plasmons [63] but this conclusion has been challenged by another study [64] which associates such contributions with bulk free carriers. Separating both of these components has been a challenge within plasmonics' research in TI. Furthermore, it has been reported that optical phonons tend to hybridise with plasmon modes in TI [65]. One way to tackle this problem is by changing the geometrical properties of the pattern, since the phonon mode's frequency stays unaltered but the plasmon ones don't, as the data in Figure 1.12 shows.

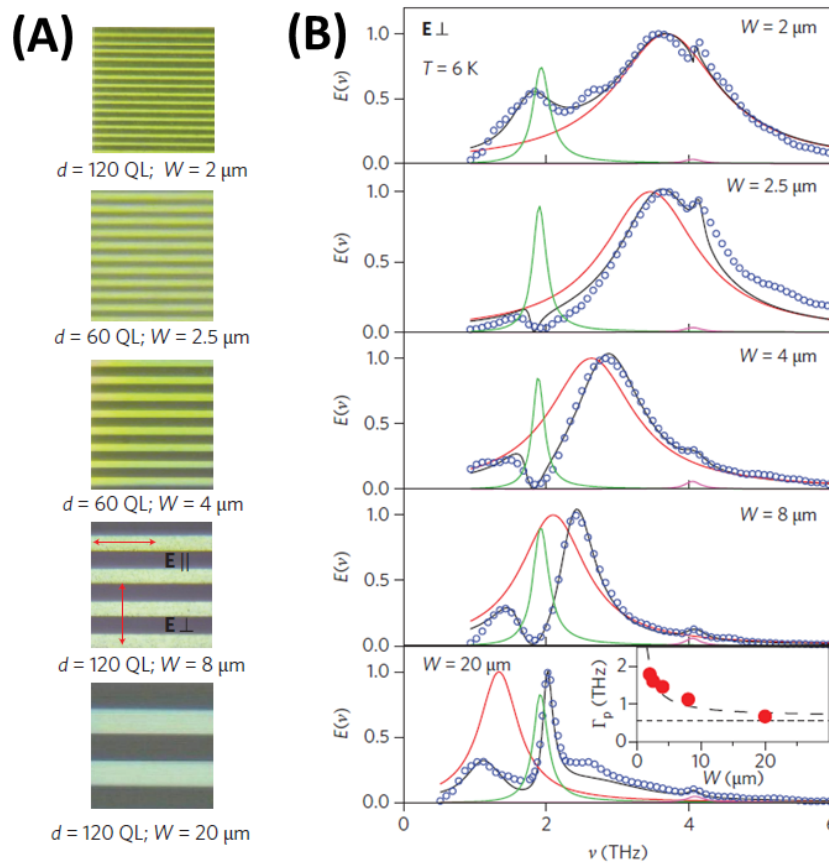


FIGURE 1.12: The phonon frequency remains unchanged with the change of period and thickness (A) of the TI pattern, while the plasmon linewidth and frequency (B). The plasmon damping increases with the decreasing of the grating period of the nanostructures. From ref. [8]

However, acoustic plasmons in TI usually have lower energies and are masked in the strongly coupled surface phonon-plasmon [62], which may add a difficulty in addressing these modes. Given all these drawbacks in addressing them, there is a special interest in having active control over these plasmonic resonances. This was proposed theoretically by K. W. Chiu and J. J. Quinn in 1972 [66] to metals and degenerate semiconductors, by

taking advantage of the behaviour of plasmons with an external magnetic field. This was the beginning of the magneto-plasmonics field.

1.2.2 Magneto-plasmonics

Magneto-plasmonics is the field where plasmon properties can be controlled by applying an external magnetic field to plasmonics structures, leading to the emergence of a new plasmon mode: the magneto-plasmon. When applying a magnetic field, the electrons will experience a Lorentz force, and this will induce closed elliptical loops in their trajectory, as illustrated in Figure 1.13.

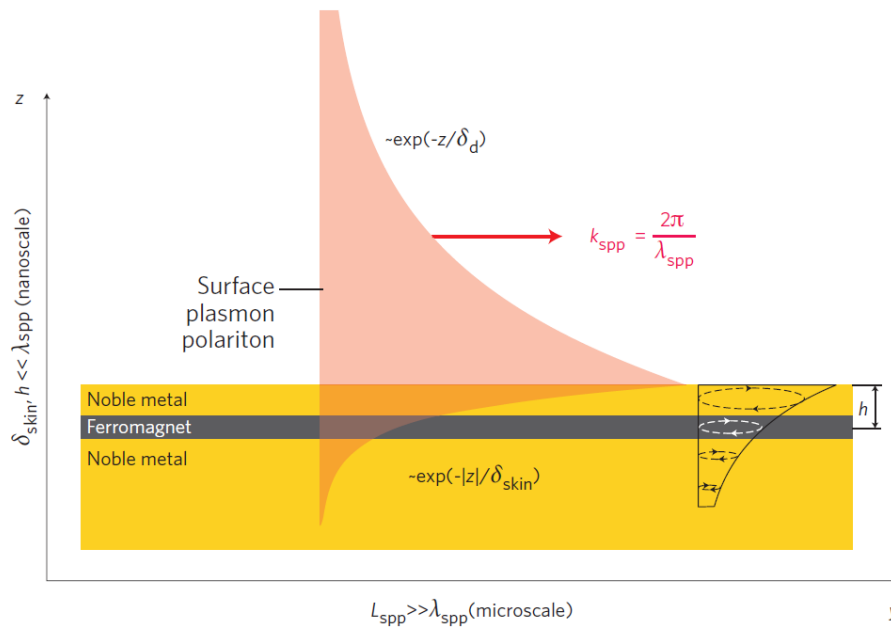


FIGURE 1.13: Magnetoplasmon in a metal-FM-metal heterostructure, in which the FM layer is comparable to the skin depth of the SPP. From ref. [67]

The action of this force in the electron's trajectories has been demonstrated to increase the SPP wave vector's magnitude [67], with the application of a magnetic field in an in-plane configuration parallel to the SPP wave vector providing the biggest change. In addition, the SPP frequency will also change, in a combination of both plasmonic ω_{SPP} and cyclotron frequency $\omega_c = eB/m^*$, described by equation 1.18, where $m^* = E_F/v_F^2$ is the dynamical mass [68].

$$\omega_{mp} = \sqrt{\omega_{SPP}^2 + \omega_c^2} \quad (1.18)$$

However, there is a fundamental limitation on studying magnetoplasmons in typical plasmonics materials, such as noble metals: their magneto-optic effect is too small $\epsilon_{yz}/\epsilon_{xx} \approx$

$10^{-7}B_x$ [67], concerning that practical applications usually use low magnetic fields in order to operate. In the other hand, ferromagnetic materials hold the biggest magneto-optic effect $\varepsilon_{yz}/\varepsilon_{xx} \approx 10^{-2}B_x$ [67], so they should be the ideal candidates to exhibit this type of phenomenon. However, ferromagnets hold a crucial drawback: they display poor plasmonic properties. Their imaginary component of the dielectric function (Equation 1.10) is bigger, which is linked to their big absorption losses. However, the real part of these two types of materials is considered comparable [39], which will lead to a smaller ferromagnet's quality factor, defined in equation 1.19. This parameter describes the damping in a given oscillatory system.

$$Q = \frac{\omega \frac{d\text{Re}(\varepsilon)}{d\omega}}{2\text{Im}(\varepsilon)} \quad (1.19)$$

This will lead to broad resonance plasmonic's peaks and shorter plasmon's lifetimes [39], which has as a consequence a smaller propagation length of the SPP in this type of material, as illustrated in Figure 1.14.

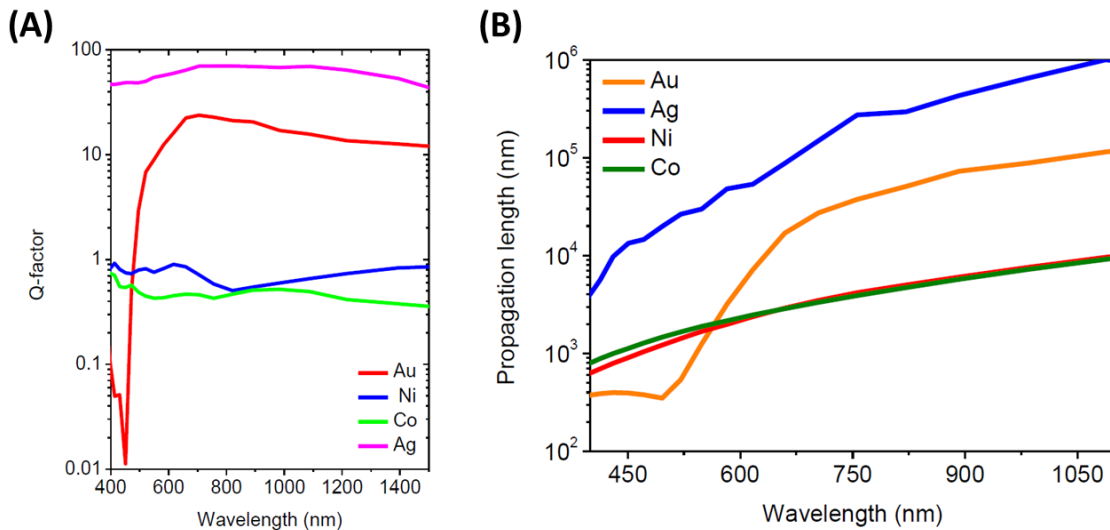


FIGURE 1.14: Due to their imaginary part of the dielectric function being bigger, noble metals hold a higher Q-factor (A) and a longer SPP propagation length (B) when compared with FM materials. From ref. [39]

Despite these occurrences being of interest in the context of magneto-optical applications, these materials are not suitable for plasmonics applications. Therefore, to study magneto-plasmonics, it is necessary to have noble and ferromagnetic metal hybrid devices, since noble metals help to bring equilibrium to these heterostructures and enhance the plasmonic signal. However, to verify this behaviour in the system, there are certain conditions that must be met: the thickness of the ferromagnetic layer needs to be thin enough to allow some overlap between the ferromagnet and the SPP evanescent wave. Therefore, this thickness should not exceed the SPP skin depth into the ferromagnet,

which is in the order of 10 nm [67].

The study of magnetoplasmons is not restricted only to noble-FM metal heterostructures, being already studied in the frame of topological insulators. It has been reported by Marta Autore *et al* [69] that these modes were observed in Bi_2Se_3 . In this material, the cyclotronic term appeared to dominate at high fields, as it is shown in Figure 1.15, whereas at low fields the magnetoplasmon mode is not fully hybridized with the latter. Besides that, the applied magnetic field breaks the time-reversal symmetry, producing chiral plasmonic modes [41].

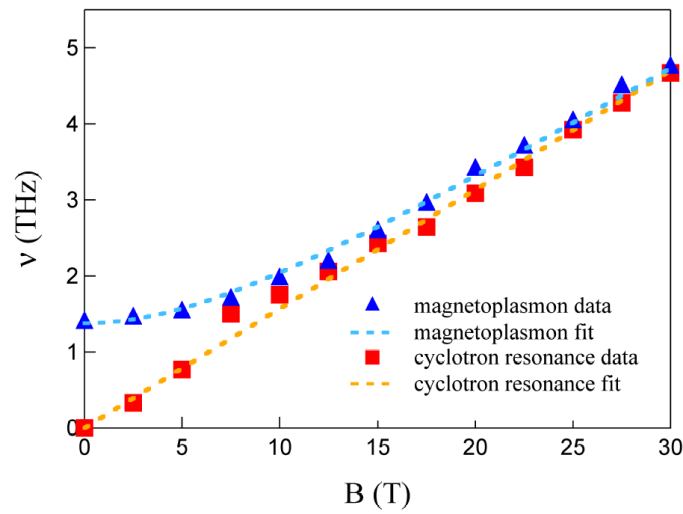


FIGURE 1.15: Comparison between the experimental magnetoplasmon and cyclotron resonances frequencies for Bi_2Se_3 ribbon arrays. The cyclotron resonance term dominates the magnetoplasmon mode at high magnetic field. From ref. [69]

Therefore, topological insulators have another advantage within the plasmonics area, since they can offer a platform to study several magneto-plasmonics phenomena while adding a lot of unexplored playground worth studying due to their electronic properties. Still, they are restricted to terahertz plasmon excitation, which may make them unsuitable for most current plasmonic applications. However, it is possible to implement the same strategy used with FM materials, by fabricating a hybrid structure of plasmonic metal and TI. In this way, it is possible to excite plasmons in TI outside of the already discussed THz range. This has already been done by Li *et al.* [70], in which they excited acoustic plasmons in a grating heterostructure of Bi_2Te_3 covered by Au in the range from 1800 to 4300 cm^{-1} .

2. Fabrication of Plasmonics Devices

Plasmonics systems are rising in popularity in R&D as they serve for a platform to a variety of effects worth applying in modern technology. These range from light confinement over sub-wavelength volumes, which can lead to enhancement of sensor's performance [71] and to thermal applications [72]. To excite plasmonic's modes and get these effects, the grating coupling method can be employed, as discussed in Chapter 1. This method is usually linked to lithography techniques, as they offer great resolution and are suitable for industrial production [73]. Between all the variety of techniques that lithography encompasses, Nanoimprint Lithography (NIL) is quickly emerging since it was first been proposed in 1995 [74]. This technique consists in pressing a stamp, containing the inverse of the intended structures, into a proper resist. Then, by applying a proper resist treatment, it is possible to transfer the associated pattern through. This treatment is different depending on the type of NIL we are dealing with: Ultra-Violet NIL (UV-NIL) or Thermal NIL (T-NIL). In UV-NIL, UV light is used to cure the photo-resist. In T-NIL, we use a thermoplastic polymer as substrate, and heat it up above its glass transition. Because it does not use light and is therefore free from the restrictions imposed by the diffraction limit, T-NIL is a noteworthy lithography technique, exhibiting better resolution than common photo-lithography experimental setups (sub-10 nm structures). In addition, it is an eco-friendlier process than its counterpart, the UV Nanoimprint Lithography [75]. In this chapter, we describe the fabrication of the plasmonic device by using this method to tailor the desired micro-grating structures. Furthermore, as DC-magneto-sputtering is the most widely used fabrication technique for the industrial production of magnetic memory devices [76], in this chapter it will also be discussed the deposition of Ag and Bi_2Te_3 onto the plasmonic structures, so that plasmon modes excitation in the samples can be possible.

2.1. Micro-grating Structures Manufacturing

2.1.1 Master Fabrication

Nanoimprint lithography relies on a stamp with the inverted of the desired nanostructures to pass them into a given thermoplastic substrate. Within the R&D community, it's

frequently used laser direct writing and electron beam lithography to produce the nanostructures onto the stamp [73]. However, as these two techniques are extremely time consuming, they are not suitable to use on an industry scale. As we intend to apply this study to future technology, we produced polydimethylsiloxane (PDMS) molds by using CDs and DVDs, as both these optical storage media already hold periodic grating microstructures tailored to them, as illustrated in Figure 2.1 (A) and (B).

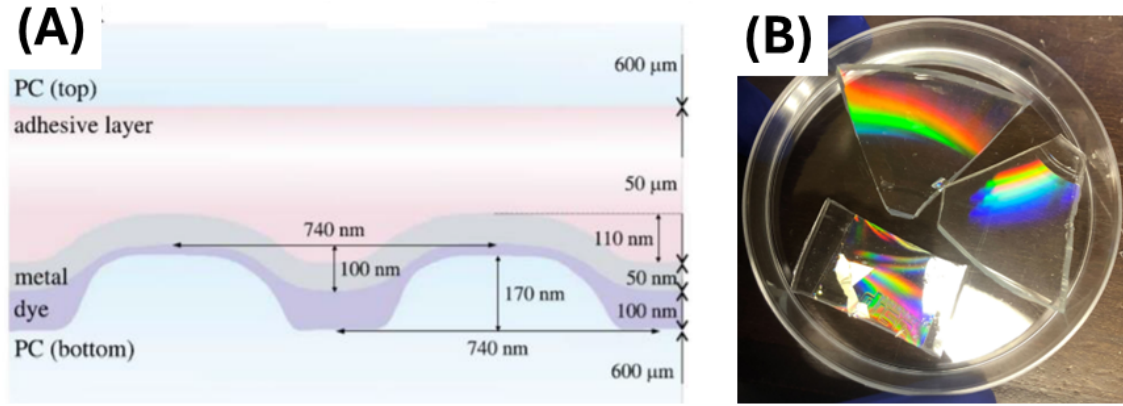


FIGURE 2.1: Schematics of the cross-section of a DVD (A) and picture of already fabricated stamps (B). By properly opening the DVD longitudinally, it is possible to access to the intended grating and use it to produce the PDMS molds

Experimental details: The PDMS stamps were prepared by using DVD-R from Verbatim and CD-R from Silver Circle as molds. To prepare the PDMS solution, it was used SYLGARDTM 184 Silicone Elastomer with a base and curing agent ratio of 7.5:1. This mixture was cured in an oven for 2 hours at a temperature of 85°C.

To verify whether the PDMS stamp held the micro-structures without any defect, a Scanning Electron Microscopy (SEM) measurement was performed in a FlexSEM 1000 from HITACHI in the secondary electrons measuring mode to better see the topography of the sample's surface. The results are displayed in Figure 2.2 (A) and (B), for both CD and DVD, respectively, covered by 15 nm of Ta. The images show that the pattern of the CD and DVD is present in the stamp perfectly without any defect. The particles present in the DVD SEM image result from the normal sputter deposition. In addition, the periodicity of the micro-structures present in the stamp was measured, $p=(1.424 \pm 0.001) \mu m$ for the CD and $p=(0.713 \pm 0.001) \mu m$ for the DVD, and compared with the values reported in the literature, approximately $1.600 \mu m$ and $0.800 \mu m$, respectively [77]. These results are summarized in table 2.1.

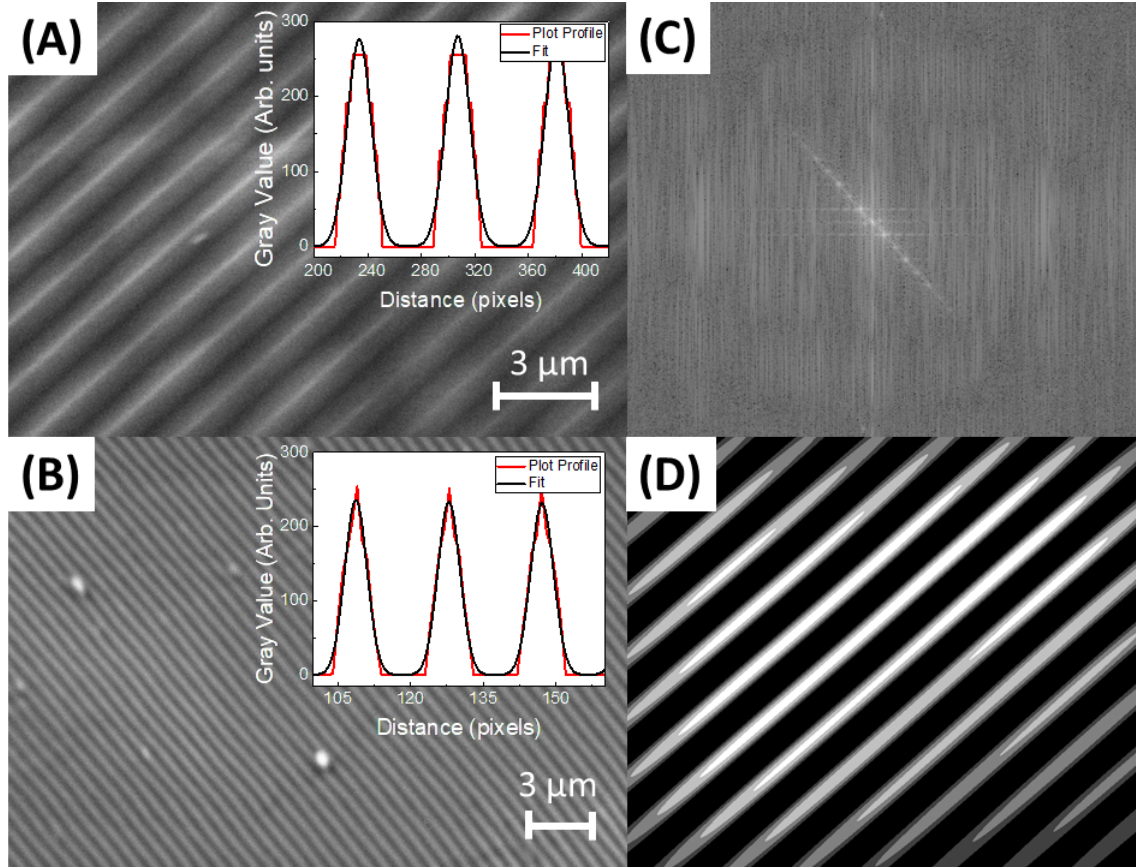


FIGURE 2.2: SEM images of the CD stamp (A), that exhibits a grating period of $p=(1.424 \pm 0.001) \mu m$, and the DVD stamp (B), which exhibits a grating period of $p=(0.713 \pm 0.001) \mu m$, both covered by 15 nm of Ta. The plot profiles of the SEM images are shown as insets, where the fit was made using a Gaussian function to obtain the maximum of the gray value. FFT of the SEM image of the CD stamp (C) with the inverse FFT after the frequency filtering (D).

TABLE 2.1: Period measured in the SEM images for the CD and DVD stamp, compared with the literature values

Stamp	Period (measured) / μm	Period (Literature) / μm [77]	Error (%)
CD	1.424 ± 0.001	≈ 1.600	11
DVD	0.713 ± 0.001	≈ 0.800	11

Data Analysis: The software used to measure the periodicity of the microstructures was ImageJ, by applying a Fast Fourier Transform (FFT) to each SEM image after enhancing contrast and brightness of pictures.

The structures without any material deposited on top were not imaged by SEM since PDMS is non-conductive and, therefore, needs a conductive layer to be imaged. When applying the FFT to the SEM image of the microstructures, the pattern present in Figure 2.2 (C) is obtained. As it can be seen, the FFT shows more than one frequency. Since

the frequency of the grating structure is the one that we are interested in, we must filter the other frequencies. The way to do that is to fill the center white pixel and the adjacent one, which are the frequencies of the gratings, and then apply an inverse FFT. This will generate the image displayed on Figure 2.2 (D). Then, we just need to make a plot profile on ImageJ, which is shown as insets in Figure 2.2. This functionality will analyse the gray value, which is the brightness of each pixel, and by doing this the periodicity is more accurately measured. One aspect that justifies the discrepancy between the measured and theoretical values is the fact that the period of the micro-structures could change with the optical storage media's producer. In addition, the values found in the literature are not exact, they consist only of an approximation. All these factors contribute to the error of 11 % compared with the literature values [77]. Despite being substantial, it doesn't represent any threat to this study since the pattern has high quality and, for the physical phenomena planned to be excited in the device, this condition is the only one that should matter most. Limitations due to software analysis are negligible, since the selection of the wrong pixel leads to an extra contribution of only 1.2 nm, consisting in a error percentage of only 0.2 %.

Furthermore, it is essential to have consistency in the period values throughout all the NIL processes employed in the fabrication of the device, so it is necessary to assure that after imprinting the devices the same periodicity is obtained. For that, a study of how the periodicity changes with the experimental conditions of the NIL process, namely temperature, pressure and reproducibility, was employed, in order to fully optimize this fabrication step.

2.1.2 Structures Fabrication

In this work, the plasmonic devices use PMMA as substrate since it is one of the most popular thermoplastic polymers used for T-NIL. The glass temperature T_g , the temperature at which the polymer suffers a phase transition into a viscous-elastic phase, of PPMA ranges from 95 °C [78] and 110 °C [79]. In order to perform the parameters' optimization to obtain perfect periodic structures in the PMMA substrate, the imprinting temperature T_i could be swept, as could the demoulding temperature T_d , the imprinting time, and the applied pressure to the stamp. The demoulding temperature, which is the temperature at which both stamp and substrate are separated, is a very important experimental parameter since its proper choice is responsible for minimizing internal stresses that can lead to fractures. Park *et al* [80] studied the best demoulding temperature for PMMA through

both experimental studies and numerical simulation, and they concluded that the best imprinting temperature was at $T_d=70^\circ\text{C}$. The imprinting temperature T_i is the temperature that must be applied to pass the pattern of the used stamp. This experimental variable must be superior to the polymer's glass temperature [79], since at this point the polymer does not display enough visco-elastic behaviour, since its storage modulus displays high enough values, to adapt itself to the micro-cavities of the stamp's pattern [81]. Figure 2.3 illustrates the transition, delimited by the glass temperature, between hard elastic to viscoelastic properties for a thermoplastic polymer.

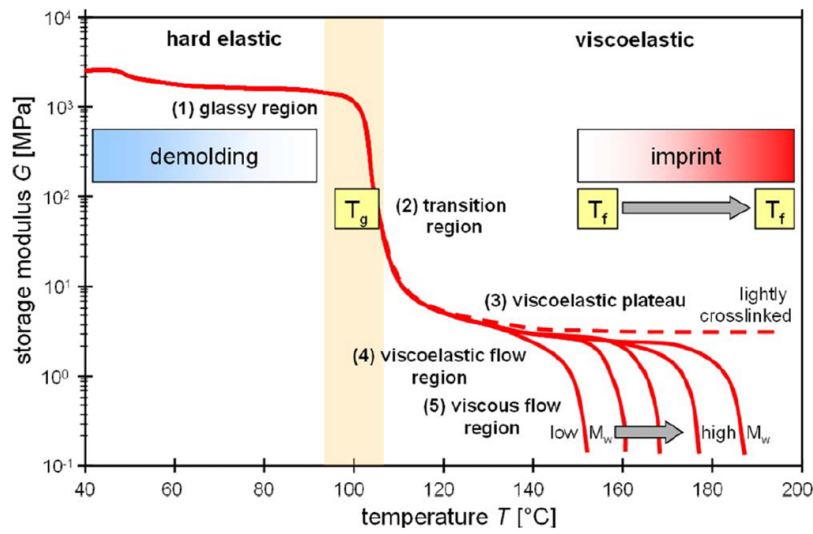


FIGURE 2.3: Storage Modulus behaviour with the temperature applied to the resist; The storage modulus is high enough at the glass temperature T_g such that the polymer is not too viscous to fill the nano- and micro-cavities of the stamp. From ref [81]

First, as a proof of concept, NIL was performed on both CD and DVD PDMS stamps, to evaluate the pattern quality after the transfer and respective periodicity on the PMMA substrate. For this step, the imprinting time was fixed at 90 seconds and the imprinting temperature at 155°C , since it has been studied that the working temperature for the T-NIL procedure should be 50°C above the glass transition of the polymer used [79]. The results are presented in Figure 2.4 (A) and (B) for a DVD and CD, respectively.

Experimental details: The NIL experimental set-up consists of a P8100 Heat Press Machine from FreeSUB. The commercial PMMA substrate, fabricated by Goodfellow, had a thickness of $250\ \mu\text{m}$ and the PMDS stamp a thickness of 1 mm.

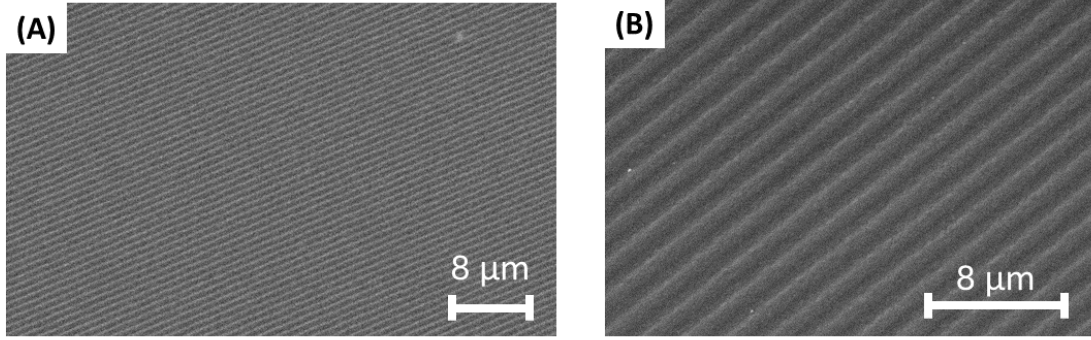


FIGURE 2.4: SEM images of both samples imprinted by both (A) DVD, periodicity of $p=0.725 \mu m$, deposited with 90 nm of Bi_2Te_2 15 nm of Ag, and (B) CD stamp, periodicity of $p=1.496 \mu m$, deposited with 15 nm of Ta.

Similarly with the previous case, the structures without material deposited on top were not imaged by SEM since PMMA is non-conductive. Concerning the pattern transfer, there is not any visible defect, and the periodicity, $p=(0.725 \pm 0.001) \mu m$ for the DVD-imprinted sample and $p=(1.496 \pm 0.001) \mu m$ for the CD-imprinted one, is within the expectable range of values for these stamps. Following this, an optimization study of the process was employed using the CD stamp in order to find the best conditions to imprint our samples. To find the best imprinting temperature to apply in this setup, the pattern quality and periodicity dependence with T_i was studied.

Experimental details: The imprinting temperature was varied from 135°C to 195°C for the aim of studying the consistency of the periodicity during the NIL procedure. The imprinting time of 90 seconds and the stamp area of 4.34 cm² was fixed for all samples.

The results obtained are displayed in Figure 2.5 (A), (B), (C), and (D). The square observed in Figures 2.5 (A), (C), and (D) encloses the electron beam produced by the SEM equipment, which has energy enough to damage the sample's pattern. This creates a serious challenge to the structures' observation because PMMA has a low glass temperature, making it simple to damage the structures. Moreover, PMMA is often used as resist in e-beam lithography [82], as such, it reacts easily with the electron beam. A quick image acquisition combined with low intensity and a low vacuum state is needed for observing the pattern. Furthermore, it is very challenging to capture a well-focused picture because the SEM technician's attempts to increase resolution resulted in the grating structures being completely destroyed. Furthermore, picture 2.5 (A) shows a discontinuity because the machine occasionally slides the detector to get a better focus during picture capturing.

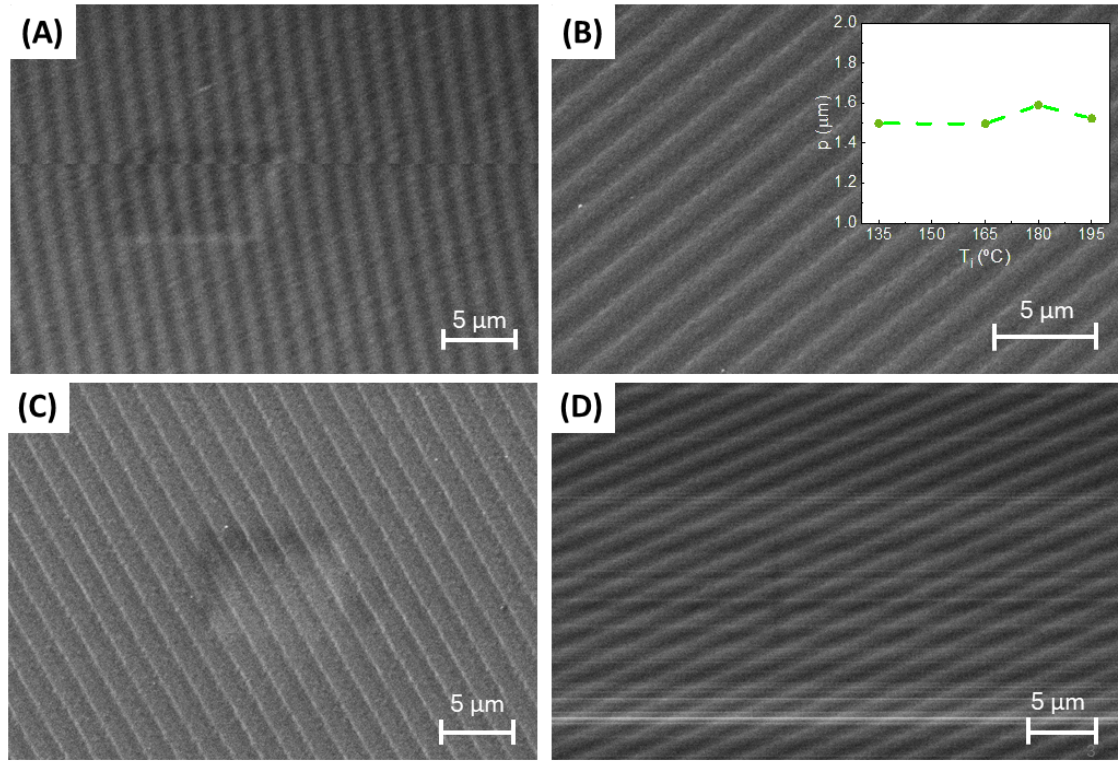


FIGURE 2.5: Dependence of the grating periodicity p with the imprinting temperature T_i (inset) together with the SEM images of the structures deposited with 10 nm of gold on top of the pattern, submitted to different imprinting temperatures of (A) 135 °C, (B) 165 °C, (C) 180 °C and (D) 195 °C;

Despite these factors, we are able to extract some information. Defects, such as areas where imprinting didn't occur (which is pretty typical due to air bubbles that are commonly trapped in NIL set-ups [83]), are not observable. Moreover, it is possible to extract the periodicity for each SEM image using the same data analysis tool as used with the stamp's structures. The data collected is presented in the inset of Figure 2.5 (B). The maximum variance of the grating period throughout the range of the imprinting temperatures was of $\Delta p = 93$ nm, which corresponds to 6% of the measured periodicity. The most likely hypothesis to justify this discrepancy is that the periodicity can vary for each CD made by the same producer since we used a fresh CD stamp for every sample. Since the pattern transfers without any noticeable defects and the variance falls within an acceptable range, there is no issue with this step of the device's manufacture and we can consider the periodicity to be constant.

The next experimental parameter that could potentially change this fabrication procedure is the applied pressure. Therefore, a similar study was employed, where the applied pressure was varied in order to see whether the pattern exhibits less quality, after being transferred, or to study where the period of the grating could change, in order to check

whether this fabrication step has constancy with this experimental parameter sweeping.

Experimental details: Within this experimental study, since FreeSUB does not reveal the heat press machine's pressure output, the pressure values are unknown. Hence, pressure data will be displayed as the area of the stamp. The stamp's area was changed from 1.06 cm^2 to 8.52 cm^2 to vary the pressure while maintaining a fixed imprinting temperature of 155°C . The time of imprinting remained the same. In this study, we used PMMA with a thickness of 2 mm as a proof of concept to test whether we could implement different thermoplastic thickness' values in our device.

The results gathered from this study are presented in Figure 2.6 (A), (B), (C) and (D). Several things can be concluded from this data.

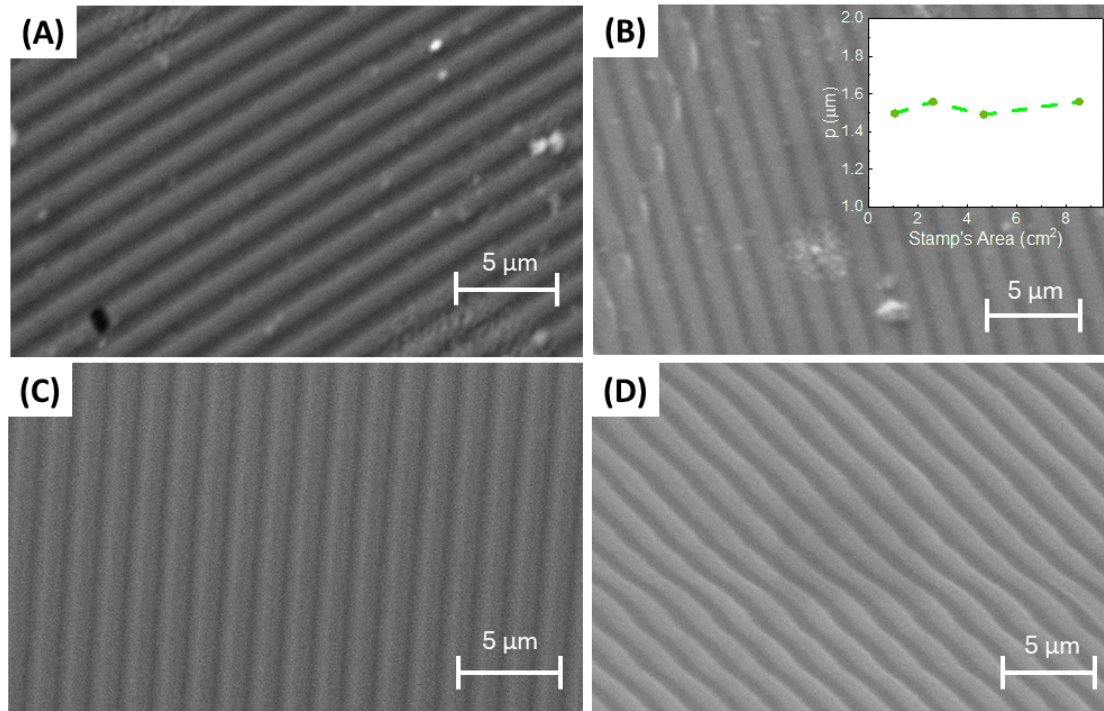


FIGURE 2.6: Dependence of the grating periodicity p with the stamp area (inset) together with the respective SEM images of the pattern with 15 nm of silver on top, associated with different stamp areas (A) 1.06 cm^2 , (B) 2.60 cm^2 , (C) 4.66 cm^2 and with 17 nm of Ta with a stamp area of (D) 8.52 cm^2 .

The first is that most of the SEM images exhibit bad quality, as several undesirable particles are on top of the grating and there are some scratches, due to substrate preparation when cutting the thick-PMMA substrate.

To obtain a better image, we could repeat the fabrication procedure with more caution, and after the cutting step, an ultra-sound treatment could be employed to the PMMA substrate.

However, when under ultra-sounds treatment, the internal cracks could grow, damaging even more the substrate. Despite the bad quality, we can observe that for most samples, the pattern was transferred without any fail related to NIL errors', such as air bubble trapping, with the expected periodicity. The same cannot be concluded regarding the sample of Figure 2.6 (D). This SEM image appears to exhibit a "zigzag" pattern instead of the common appearance seen in the previous images. This type of pattern was already studied by Ding *et al* [84] in polystyrene (PS) samples, where they submitted similar grating structures to a thermal annealing and studied their decay. This "zigzag" was attributed to relaxation mechanisms of the polymer. Similarly, this serves as a foundation to justify our result: as this defect only appears for the biggest area value, we think that the correspondent pressure is not enough to fully fill optimally the micro cavities of the stamp. This could be explained by thinking of both pressure and temperature as allies to induce a more viscous-elastic behaviour for the polymer to adapt itself to the irregularities of the stamp. Although the polymer undergoes a phase transition mostly due to temperature, the pressure that is applied can also impact the polymer's mobility and relaxation dynamics. This can lead to molecular rearrangements and better chain mobility, which increase flow and help the filing of the stamp features. If the pressure is not enough, all these mechanisms cannot be sufficient to obtain a well defined pattern transfer. For stamp areas enclosed within 1.00 to 4.66 cm², these features are not present in the samples and, moreover, the periodicity values remained coherent with the stamp ones with a maximum variance of $\Delta p = 69$ nm, which corresponds to only 5% of the measured periodicity. This variance is attributed to the same factors that influenced the previous study, since for each sample was used a new CD stamp. Therefore, upon this variation, we can consider the periodicity to be constant.

Another parameter calibrated in the T-NIL set up was the number of imprinting procedures applied to the same stamp. Considering that a significant amount of resources would not be wasted in the creation of the PDMS stamps, this is extremely significant for the implementation of industrial operations.

Experimental details: The only variable within this experimental study was the number of imprinting procedures the CD-stamp was exposed to. This was 1 mm thick, and the PMMA substrate was 250 μm thick. The imprinting temperature was fixed at 155°C, the stamp's area at 4.34 cm², and the imprinting time at 90 seconds.

The results gathered from this study are displayed in Figure 2.7 (A), (B) and (C).

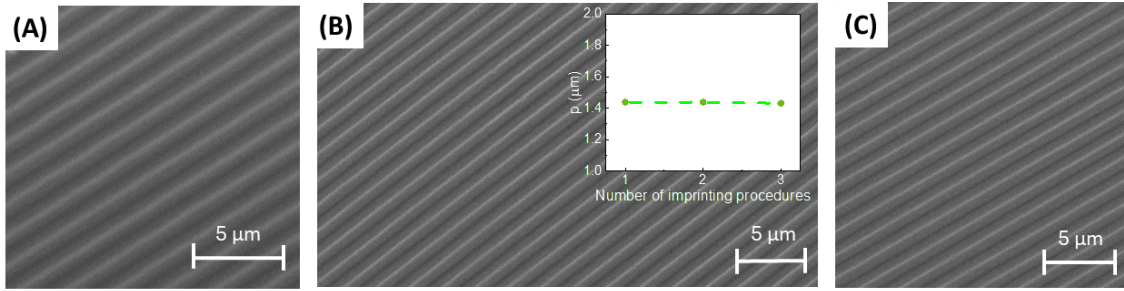


FIGURE 2.7: Dependence of the grating periodicity p with the number of imprinting procedures (inset) together with the respective SEM images of the pattern with 15 nm of Ta on top, exposed to 1 (A), 2 (B) and 3 (C) imprinting procedures.

The SEM images show a high quality pattern transfer with no defects visible, which indicates that the repetition of this process is within this range. A fourth attempt was made with no success since the pattern was visibly not fully passed to the substrate. This is easily observed by looking at the non-homogeneous diffraction pattern of the light, as certain regions of the substrate lack this feature since a diffraction pattern is associated with the presence of the micro-structures. Thus, the limit of repeatability of our set-up was fixed to be 3. This limit could be further extended with certain set-up adjustments, including implementing vacuum during the application of heat and pressure, as doing so would allow us to decrease both the difficulties associated with air trapping between the substrate and the stamp and contamination of the sample with undesirable particles that could interfere with the pattern transfer. This could also be solved by buying a proper NIL experimental set-up, that would encompass all these features. In addition, concerning the periodicity of each SEM image, the variance of the periodicity is much smaller than for the previous studies. We obtained a maximum variance of $\Delta p = 7$ nm, which corresponds to 0.5% of the measured periodicity. The fact that we used the same stamp in this study enhances the main source of error present: each CD has a slightly different periodicity, even when the CDs are from the same producer. Moreover, in the already mentioned range, the periodicity is also constant.

2.2. Material Deposition

The next step toward the fabrication of the intended devices is to proceed with material deposition on top of the structures, enabling the excitation of plasmon modes. Figure 2.8 (A) and (B) illustrate the cross-section and top of the desired structures.

Since exciting plasmon modes in topological insulators is commonly linked with THz excitation, to observe the phenomena planned to be excited, it is needed to incorporate these wavelength sources into the laboratory.

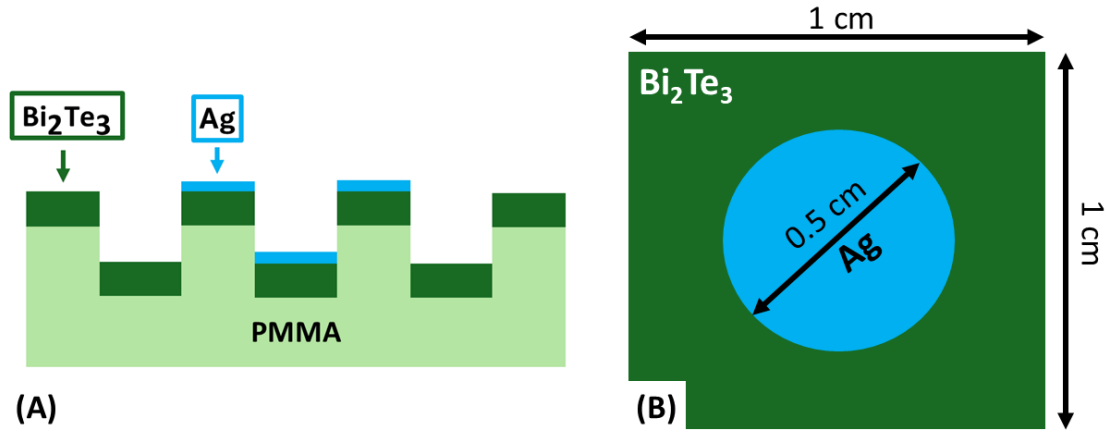


FIGURE 2.8: Schematics of the cross section (A) and top view (B) of the plasmonic devices' ideal structure. Ag (blue) is deposited on top of Bi₂Te₃ (green) only in the center for the plasmon's excitation.

However, due to equipment limitations, this is not possible. In addition, today's technology is more adapted to visible and near-infra-red (NIR) light rather than middle-IR. With this in mind, we planned on studying plasmonics in topological insulators by exciting these modes on a noble metal and couple it with the TI, as reported in [70] and already mentioned in Chapter 1. For this, the chosen noble metal was silver, as it belongs to the noble metals family and as it exhibits a smaller damping constant than gold within the visible spectrum [42]. To study the influences these phenomena have in Bi₂Te₃, this material was not deposited uniformly throughout the device but just in its centre. As a result, there will be space to connect the contacts to the TI when performing magnetoconductance measurements, instead of connecting it to the silver layer. For that, a mask was fabricated to cover the sample with a cut 0.5 cm diameter circle in the middle, in order to enclose the laser spot area. The technique of deposition used was a DC-magneto-sputtering deposition, as it consists of both a cost-efficiency and good-quality sample fabrication deposition technique [20], which is becoming widely popular in the fabrication of spintronics' applications in the frame of the industrial world [76].

Experimental details: The equipment used was a CY Scientific Instrument DC-Sputtering device. For the Bi₂Te₃ layer, the parameters used were 20 mA of applied current, table rotation speed of 60 rpm, and constant Ar flux of 50 sccm. For the Ag layer, the same conditions were applied. The vacuum was broken between depositions.

2.2.1 X-Ray Diffraction

In order to address the crystal properties of the materials in the device, X-Ray diffraction measurements were performed. This characterization technique is extremely important since a polymer substrate, which is amorphous, is being used and therefore monitoring the crystal parameters of the sample gives a better insight to understand the physical phenomena to be observed in the up-coming results. The first analyzed sample was a heterostructure of 17nm of Ag and 70 nm of Bi_2Te_3 . The XRD spectra are presented in Figure 2.9 (A) and (B) for both samples deposited onto PMMA and glass, respectively.

Experimental details: The equipment used was a Rigaku SmartLab Diffractometer in the Bragg-Bentano (θ - θ) geometry, in an angular range from 5° to 100° . The X-Rays used for the measurements were $K\alpha$ radiation obtained from a Cu target, with a wavelength of $\lambda = 1.5404 \text{ \AA}$. The diffractometer works at 9 kW, 45 kV, and 200 mA.

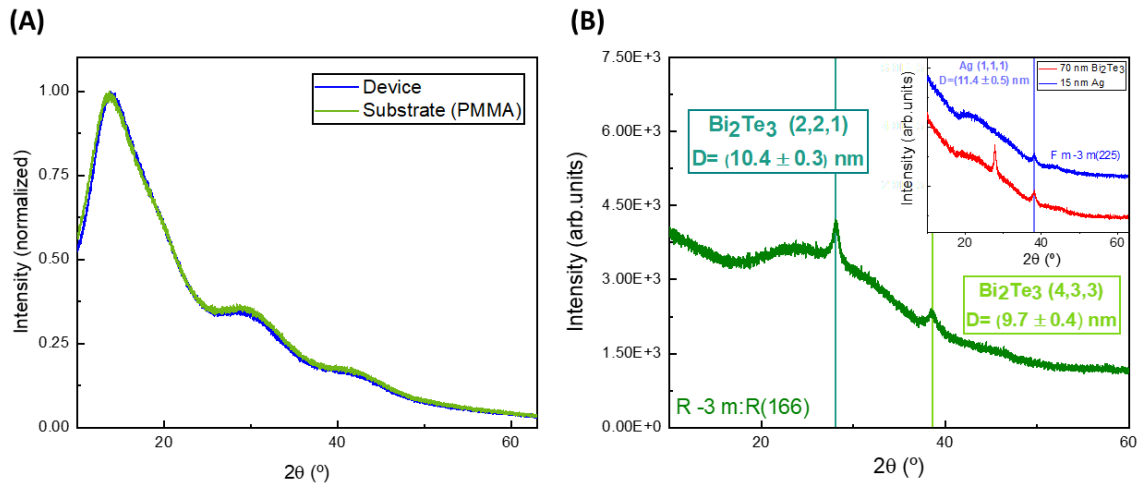


FIGURE 2.9: XRD spectra for a thin film heterostructure of 17 nm of silver and 70 nm of Bi_2Te_3 deposited on PMMA (A) and glass (B). The materials deposited on PMMA are highly amorphous and, therefore, the signal from the substrate masks the sample's signal. The inset on figure (B) shows the XRD of a 15 nm thin film of Ag (blue) and of 70 nm thin film of Bi_2Te_3 , both measured on the same XRD diffractometer but integrated in 6-sample holder rotative system. The D parameter quantifies the crystallite size for each peak.

Concerning the signal from the sample deposited onto PMMA, the background signal from the substrate completely masks the Bragg peaks from the sample, therefore, we cannot conclude anything from this sample. Possibly, since PMMA is a polymer and therefore highly amorphous, there is not any preferential orientation of the crystal growth,

and there are multiple diffraction peaks, which are not well defined throughout the spectrum, masked by the background signal. The same measurements were performed with a glass substrate, that is also amorphous, where two Bragg peaks that are associated with Bi_2Te_3 were identified. To treat this data, the Rigaku SmartLab Software installed in the computer associated with the XRD measurements was used. The software uses a Pseudo-Voigt function to fit the peaks of the spectrum and associate them to a given crystallographic plane. The Pseudo-Voigt function is a Voigt function's approximation, which by the hand is a convolution of a Guassian $G(x', \sigma)$ and Lorentzian $L(x - x', \gamma)$ function, and is defined in equation 2.1.

$$V(x, \sigma, \gamma) = \int_{-\infty}^{\infty} G(x', \sigma) L(x - x', \gamma) dx' \quad (2.1)$$

Reflections at 28.1° and at 38.6° were identified, respectively to the (2,2,1) and (4,3,3) planes of the R -3 m space group of Bi_2Te_3 . With this data, we could conclude that the Bi_2Te_3 layer has likely a low crystallinity degree, which was already expected due to the amorphous character of the substrate. Furthermore, there were no peaks that could be identified as coming from the Ag layer. In addition, this could indicate that the Ag layer has also a low percentage of crystallinity. To probe that, a thin film of 15 nm of Ag also deposited on glass was measured by the same XRD diffractometer, along with a thin film of 70 nm of Bi_2Te_3 deposited in the same substrate, and the respective results are shown in the inset of Figure 2.9 (B). Concerning the Ag samples, the F m -3 m (225) space group was identified, and there is only one peak identified at 38.1° , which coincides with the already identified Bi_2Te_3 peak at 38.6° on the XRD spetcrum of the Bi_2Te_3 / Ag thin film. Therefore, we believe that this Bi_2Te_3 peak, despite being attributed to a Bi_2Te_3 phase, could also display a contribution coming from the Ag layer.

The crystallite size D was also computed for each observable peak by using the Debye-Scherrer equation, stated in Equation 2.2, where k is the the Scherrer constant ($k=0.9$), $\lambda=1.5414\text{\AA}$ is the the incident X-ray wavelength and β represents the broadening of the Bragg peaks.

$$D = \frac{k\lambda}{\beta \cos \theta} \quad (2.2)$$

From equation 2.2, the crystallite size for the Bi_2Te_3 (2,2,1) peak was calculated to have a 10.4 nm diameter and the (4,3,3) one 9.7 nm, which is comparable with the lattice parameters of the identified phase (10.45 nm). Therefore, the samples are polycrystalline with very small grain sizes. Concerning the Ag thin film, the crystallite size was also calculated being 11.4 nm for the (1,1,1) Ag peak. This could have some influence on the

overall performance of the device, as it shall later be discussed in the Chapter 5.

2.2.2 Thickness Calibration

When fabricating plasmonic structures, one of the most important geometrical parameters to study is the thickness of the plasmonic material layer. By varying the thickness, one can change the optical behaviour and, consequently, the plasmonic phenomena, since the dielectric function and plasma frequency can both differ from bulk values as one approaches the nanoscale [50]. So, it is crucial to get control over deposition rates when dealing with this type of technology. We deposited both Ag and Bi₂Te₃ thin films onto glass substrate during a specific time in order to calibrate the sputtering equipment. We then used X-Ray Reflectometry (XRR) to measure the thickness of the thin films associated with that period of time. The equipment used was a Rigaku SmartLab X-Ray Diffractometer in the low-angle configuration. For this type of configuration, oscillations are observed in the XRR spectrum, which are known as Kiessig fringes [85]. These fringes, first observed by H. Von Kiessig in 1931 [86], arise from both constructive and destructive interference of the incident x-rays that are reflected in the different interfaces between layers of the sample. It does not depend on the crystalline parameters of the material, being more sensitive to surface parameters. The thickness of the layer and the roughness of the surface and interface can both be determined from these oscillations. The layer thickness is computed using Equation 2.3, being derived from the periodicity of the Kiessig oscillations.

$$m = -\frac{1}{2} + \frac{2t}{\lambda} F(\theta_m) \quad (2.3)$$

In Equation 2.3, m is the order index of the maximum of the fringe, t is the thickness of the sample, λ the x-ray wavelength, and $F(\theta_m) = \sqrt{\cos^2(\theta_m) - \cos^2(\theta_c)}$, where θ_m is the angle correspondent to the fringe maximum and θ_c is the critical angle at which the fringes become visible in the pattern. The roughness of both substrate and interface can be deduced by the decay of the XRR signal. The density of the material can also affect the pattern, being related to the critical angle at which the fringes start to become visible. The XRR spectra of both Ag (A) and Bi₂Te₃ samples (B) are presented in Figure 2.10.

Data Analysis: The software used to analyse the XRR data was GenX 3. The parameters chosen to be accounted to the fit were the the thickness of the film t , the roughness σ of both the film and the substrate and the density of the film. The Levenberg–Marquardt algorithm was used to fit the data.

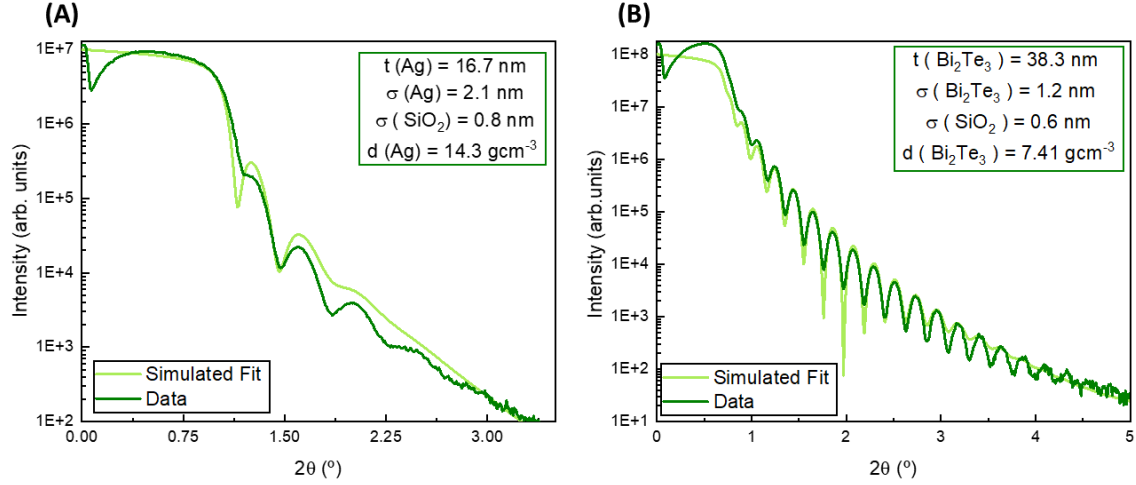


FIGURE 2.10: XRR spectrum for a 3 minute silver deposition (A) and a 5 minute Bi_2Te_3 deposition. The simulated parameters are also presented in both graphs.

As the goal was to determine the thickness of the material, which was $t=16.7 \text{ nm}$ for a 3 minute silver deposition and $t=38.3 \text{ nm}$ for a 5 minute Bi_2Te_3 deposition, it was given main focus to this parameter when fitting the experimental data. The density of the material was also deviated from the theoretical value $d_{\text{theoretical}}(\text{Ag}) = 10.36 \text{ gcm}^{-3}$ [87]. Density variations are possible since theoretical values are calculated for bulk samples, and when fabricating samples with spatial confinement at the nanoscale, we can induce stress and alter the packing factor, which will modify the density of the material as a consequence. The thickness value calculated through the GenX 3 software coincided with the one calculated through Equation 2.3.

2.3. Main Conclusions

Micro-grating structures were fabricated on polymethyl methacrylate substrates through nano-imprint lithography, with periodicities of approximately $p=(0.725 \pm 0.001) \mu\text{m}$ and $p=(1.496 \pm 0.001) \text{ nm}$. The periodicity of the samples is in agreement with the literature reported values [77] and exhibits good quality. The fabrication process was also optimized, where we verified that varying some parameters will not change the periodicity of the samples. These parameters include the pressure applied to the stamp, the number of procedures a given stamp was exposed to, and the imprinting temperature applied to the setup. This does not imply that these parameters do not influence the lithography procedure, but it does mean that above the recommended values, the periodicity is not altered and remains constant. Additionally, through x-ray diffraction measurements, an amorphous structure was identified for a thin film heterostructure of 17 nm of silver and 70 nm of Bi_2Te_3 deposited on PMMA.

3. Numerical Simulations

One of the ways to probe plasmonic modes and to gain a deeper understanding of the system's physics is to numerically simulate the behaviour of our devices and sweep external and internal parameters. To get a better understanding of the system's quantum influence, one can also compare the classical simulation to the experimental results. This is extremely useful for plasmonics analysis since these events are usually addressed by the classic motion's equations, which may be considered to oversimplify their quantum nature. Nonetheless, good qualitative and quantitative results usually come from these calculations [88]. However, when dealing with quantum materials, it is good to compare the already expected classic results with the quantum-related experimental results to analyse to what extent the quantum properties of the topological insulators alter the optical spectrum of our devices.

For that, the COMSOL multiphysics 5.6 software was used. It is a numerical simulation's software widely used within the physics' community, with a vast number of advantages. This software carries several modules that treat most physics phenomena individually by solving the equations related to the system intended to be studied via Finite Element Methods (FEM). Besides that, the software offers a platform to connect modules and explore the interaction of many physics phenomena in any system, which is its biggest advantage compared with other numerical simulations software. The Finite Element Method divides the overall geometry into a mesh of smaller elements at a user-defined size. This division can be distributed homogeneously throughout the overall geometry or be more refined at some domains or interfaces than others in order to better capture smaller local physical effects. Then it will solve the partial differential equations of the system for each one of the elements by numerical methods, which represent an approximation to the real solution of the equations. This numerical solving is characterized by a tolerance that is also user-defined.

The module used to simulate the plasmonic devices was the wave optics module, which can be used to solve plasmonics, gratings, and metamaterials problems beyond many others [89]. In particular, the Electromagnetic Waves, Frequency Domain (EWFD) analysis package was chosen, to employ frequency-dependent studies of the 2D electric field plot, which quantifies the modulus of the electric field over the built-geometry of the device, and the device's optical spectra calculation (absorbance, reflectance and transmittance).

This may raise some questions for the reader, as usually wave optics simulations use Finite-Difference Time-Domain (FDTD) methods instead of FEM to numerically simulate photonic systems. Despite being the most obvious choice when it comes to this type of numerical study, FDTD is only better than FEM for time-dependent problems. When the system doesn't have any time dependence, FEM is a suitable method. In addition, it has been studied that for some plasmonic systems, FEM is a more suitable numerical simulation method than FDTD [90], since FDTD simulations require a longer time iteration to display numerical stability and accuracy, which for nanostructured interfaces can turn the simulations too heavy to process. Therefore, since in the system proposed for this work there is no time-dependent component, a FEM numerical solving simulation is the best option for simulating our devices.

3.1. Model Building

A structure similar to the one shown in Figure 2.8 was initially simulated, focusing only on the Bi_2Te_3 and Ag layers. The periodicity was set to $p=0.700 \mu\text{m}$ and the grating height to $h=170 \text{ nm}$. An air layer was also introduced above the Ag layer to ensure accurate reflectance calculations. The input and output ports, as illustrated in Figure 3.1, were defined as periodic ports, the most suitable approach for simulating periodic structures and applying the periodic boundary condition, which will be further discussed.

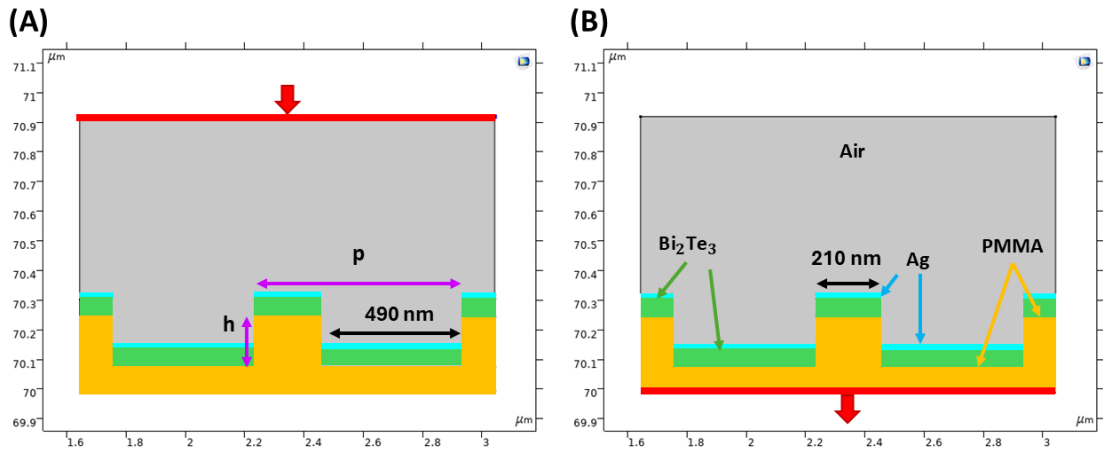


FIGURE 3.1: Device structure built in the geometry builder, with periodicity p , height of the grating $h=170 \text{ nm}$. The ports are illustrated by the red line, and their excitation direction signaled by the red arrow.

For both of them, we defined the magnetic field vector to be out of plane, $\vec{H} = H_0(0, 0, 1)$, so that the light used is within the TM state of polarization, and we also fixed the mode phase at $\theta_m=0$. For the input port, the wave excitation condition was activated, the angle

of incidence set to 0, and the real part of the refractive index fixed to 1, which is the respective value for air. For the output port, the refractive index was defined using the refractive index data of PMMA. Then, other boundary conditions were defined. Periodic boundary conditions were applied to the lateral limits of the geometry in order to repeat the unit cell. This is extremely important because it will spare a lot of simulation time and will lead to more accurate results rather than simulating a fixed number of repetitions of the unit cell. In addition, scattering boundary conditions were also applied to the same lateral limits of the geometry of the device. This is important for the outgoing waves to be absorbed when they encounter this limit. In this way, these waves can be considered as being propagated away from the domain without being reflected into the built geometry, which ensures that the simulation only considers the scattering components of interest to the physical system in study. These types of conditions were also used for other periodic plasmonic grating systems, such as the gold-grating structures simulated by Iqbal *et al.* [47]. A far field domain, which can be applied to a domain in the built-geometry to evaluate the electromagnetic field at a large distance from the simulated structure, was also applied to the air domain and bottom layer of the PMMA substrate, allowing to apply smaller values for the thickness of both these layers, simplifying the model and avoiding simulation cost. Concerning the numerical solving, the software will solve equations 3.1 and 3.2 where $\vec{E}$ is the electric field and μ_r is the relative permeability.

$$\vec{\nabla} \times \mu_r^{-1}(\vec{\nabla} \times \vec{E}) - k_0^2 \epsilon_r = \vec{0} \quad (3.1)$$

$$\vec{E}(x, y, z) = \vec{E}(x, y) \exp(-ik_z z) \quad (3.2)$$

The software requires an electric displacement field input in order to relate these equations to the material in the device by giving an input to the relative electric permittivity. The refractive index model was chosen, in which the software sets that $\epsilon_r = (n' - in'')^2$ and $\mu_r = 1$, where n' is the real part of the refractive index and n'' the respective imaginary part, often addressed as the extinction coefficient of the material κ . The reader might think that by choosing this model we are inducing error in our simulation results, since when it comes to simulating metals, the conductivity has an influence on the optical behaviour. However, the conductivity is being taken into account with this approach because it is dependent on the dielectric function. Since $n = n' + in'' = \sqrt{\epsilon_r(\omega)}$, $\epsilon_r(\omega)$ depends on the plasma frequency (as stated in equation 1.10), and the plasma frequency depends on

the conductivity, as shown in equation 3.3.

$$\omega_p = \sqrt{\frac{ne^2}{\epsilon_0 m}} = \sqrt{\frac{\sigma(\omega)}{\epsilon_0 \tau}} \quad (3.3)$$

The values of the real and imaginary parts of the refractive index will be given as an input for each material used in the simulations. For that, it was given as input the experimental data from bulk samples of both these optical variables from P.B. Johnson [91], N. Sultanova [92] and H. Swaha Krishnamoorthy [93]. The extinction coefficient of PMMA was fixed at 0.004, since the values are mainly constant and very near zero for the frequency range considered in this work [94]. Then, the software will compute the scattering parameter (S-parameter), which is a complex-valued matrix that correlates the transmission and reflection of electromagnetic waves at different defined ports. Later, the reflectance and transmission can be calculated using these quantities [95], as stated in equations 3.4 and 3.5, in which $\vec{E}_c$ is the computed electric field, $\vec{E}_1$ is the fundamental mode of the electric field in the first port, $\vec{E}_2$ the fundamental mode of the electric field in the second port and A_1 and A_2 are the cross-sectional areas of the input and output ports respectively.

$$R = |S_{11}|^2 = \left| \frac{\int_{port1} ((\vec{E}_c - \vec{E}_1) \cdot \vec{E}_1^*) dA_1}{\int_{port1} (\vec{E}_1 \cdot \vec{E}_1^*) dA_1} \right|^2 \quad (3.4)$$

$$T = |S_{21}|^2 = \left| \frac{\int_{port2} ((\vec{E}_c - \vec{E}_2) \cdot \vec{E}_2^*) dA_2}{\int_{port2} (\vec{E}_2 \cdot \vec{E}_2^*) dA_2} \right|^2 \quad (3.5)$$

Then, the absorbance is simply computed by remembering the relation $A+T+R=1$.

With all these parameters defined and all the physics of the system well-established, the model is solidly built.

3.2. Device with DVD periodicity

A device with all the already mentioned parameters and a periodicity of $p=0.700 \mu\text{m}$ was simulated, where the top part of the grating had a width of $0.210 \mu\text{m}$ and the lower part had a $0.490 \mu\text{m}$ width.

3.2.1 Ag thickness sweep

As mentioned in Chapter 2, changing the thickness of the material on top of the grating structures could alter the plasmonic resonant frequency. Therefore, this study was focused on sweeping the thickness of both Bi_2Te_3 and Ag. The wavelength of the incoming light was varied from 400 to 1600 nm in the perpendicular direction to the device. The

mesh was defined with a free triangular pattern, which is simply a non-structured triangular mesh. The overall size restrictions to the mesh throughout the whole device's structure were a minimum element size of 10 nm and a maximum one of 50 nm. However, as it can be seen in Figure 3.2, the mesh was defined to smaller element sizes in the ports and in the borders of the Ag and Bi_2Te_3 layers.

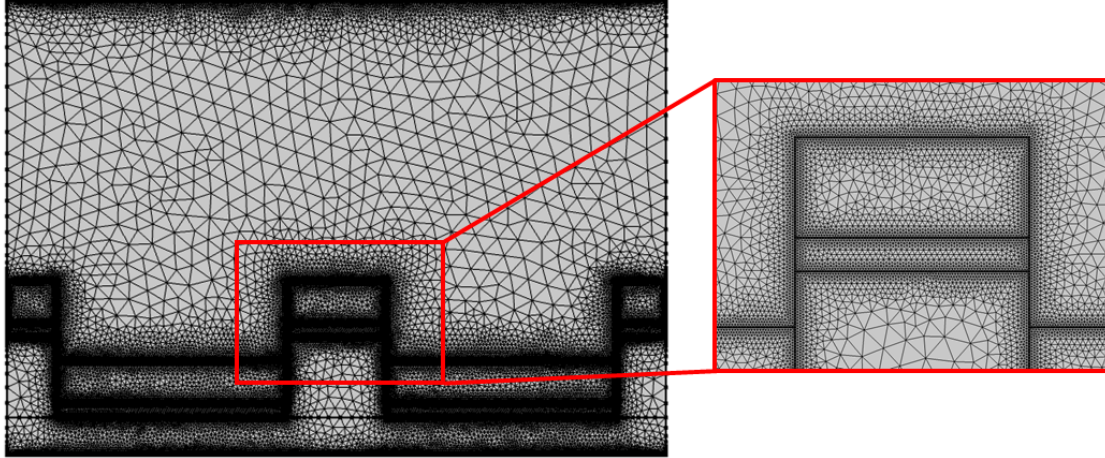


FIGURE 3.2: Mesh configuration of the simulated device. The mesh is finer in the limits of the material layers and in the ports for a more accurate calculation.

This takes into account the fact that the material layer thickness will be smaller than 10 nm for some cases, therefore, smaller meshes have to be defined. In addition, in order to take into account a better approximation to the plane wave, these must have an element size smaller than 20 nm. Finally, a finer mesh leads generally to better calculation, despite the longer calculation time. For all these reasons, the mesh in these limits was defined to have a minimum element size of 2 nm and a maximum one of 2.5 nm.

It was first simulated a thin film with 30 nm of Bi_2Te_3 and 15 nm of Ag on a PMMA substrate together with a grating sample with the same before-mentioned parameters, as displayed in Figure 3.3. This is important to distinguish the origin of different contributions to the spectra. It is possible to observe that the numerical simulations' data from the film does not exhibit any peak, whereas the grating structures exhibited three peaks that could potentially be linked to plasmonic resonances, located between 442 nm and 480 nm, 555 nm and 573 nm, and finally 680 and 695 nm. So it will be assumed from now on that the peaks are indeed coming from the grating structures and are linked with plasmonic phenomena. Additionally, the spectra coming from the grating exhibit a vertical shift relatively to the thin film one, highlighting the increment of absorbance that the tailoring of structures to the sample can enhance.

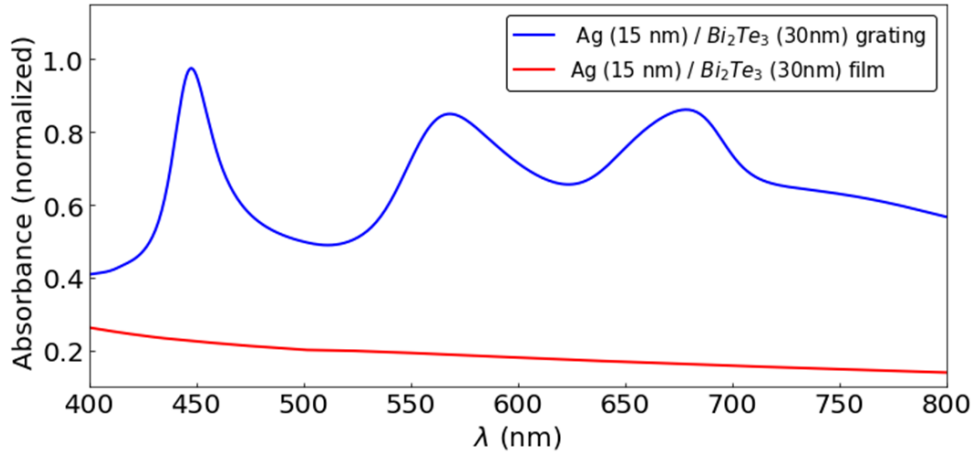


FIGURE 3.3: Comparison of the absorbance spectra of a device with and without gratings for a Bi_2Te_3 thickness of 30 nm and an Ag thickness of 15 nm. For the film, we observed no peaks.

Following this, the thickness of the Ag layer was swept from 5 nm to 90 nm while maintaining the Bi_2Te_3 one fixed at 30 nm. The absorbance spectra resulted from this sweeping are shown in Figure 3.4.

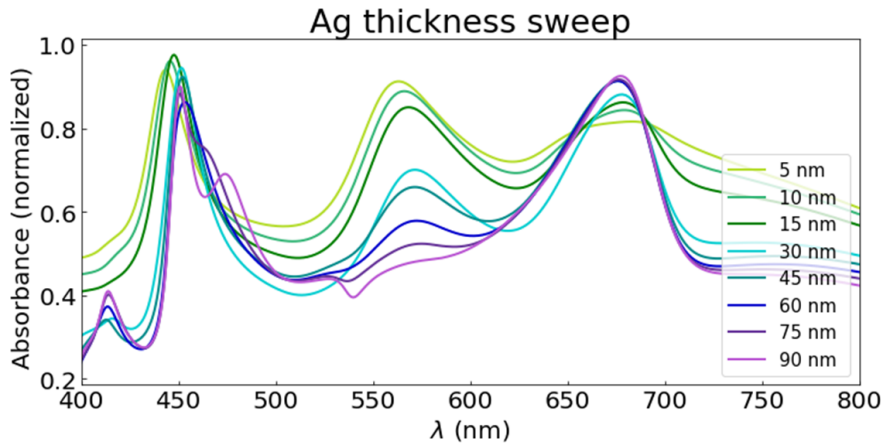


FIGURE 3.4: Absorbance spectra of the device while varying the Ag layer thickness from 5 to 90 nm. Three possible plasmon modes were observed.

These peaks exhibit a shift in frequency upon Ag thickness increase. Therefore, these shifts were analyzed to observe whether they follow any tendency. In Figure 3.5 (A), the frequency shift for the first peak, which is signaled in the inset, is plotted measured in terms of its center wavelength. The line-shape of the peak exhibits some degree of asymmetry, which has been observed in some plasmonic systems due to Fano interference [96]. However, the fano description consists of a quantum interference phenomenon [97], and quantum phenomena are not included in this COMSOL package, so we cannot attribute this asymmetry to such a contribution and, therefore, we cannot use a fano fit to analyse our data.

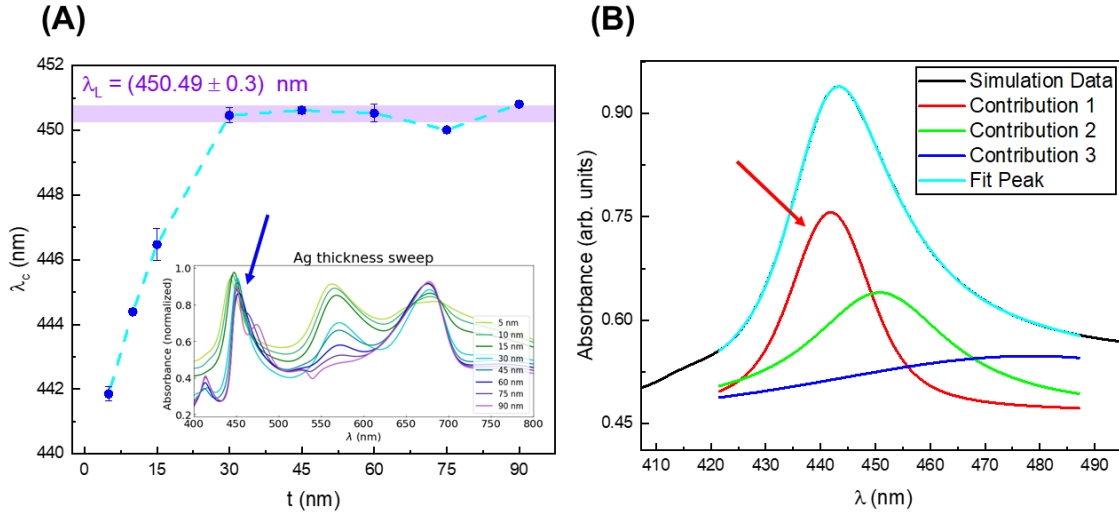


FIGURE 3.5: Frequency shift of contribution 1 of the first peak (A), signaled on the inset, and respective peak deconvolution for 5 nm of Ag (B). The mode linked with contribution 1 increased its frequency when increasing the thickness up to 30 nm. The violet-shaded region encloses the λ and respective uncertainty.

The only thing we can conclude is that there is a coupling of two modes that leads to an asymmetric peak. To fit this peak, we chose to use a voigt function since it encompasses a convolution of both Gaussian and Lorentzian functions, and since it was the one who better fitted the simulation data. We identified three contributions, as shown in Figure 3.5 (B). The contribution 3 was identified to take into account the background signal from the spectrum. Then, we analyzed the behaviour of the contribution 1. This mode increases its plasmon excitation's wavelength as the thickness is increased up to 30 nm. Afterwards, it doesn't shift in frequency anymore, having a fixed wavelength of plasmon excitation at 450 nm. This has also been observed previously with silver thin films [98] and gratings [99]. This can be interpreted by recalling that, as previously mentioned in the chapter 1, the penetration depth of silver is $\delta_m = 23.6 \text{ nm}$ for a 600 nm wavelength excitation in a silver/glass interface [43]. Therefore, based on our results, the penetration depth of silver for a 450 nm plasmon wavelength excitation at an silver/air or silver/ Bi_2Te_3 is overcome for 15-30 nm of silver layer thickness. After overcoming this limit, the plasmon cannot couple evanescently with the neighbour layer, and there is no overlap of the plasmon wavefunction with the neighbour dielectric layer. However, the behaviour of these results does not match with the already existing data for Ag thin films [98] and gratings [99]. Instead of the plasmon shifting to a smaller wavelength of excitation upon thickness increase, the plasmon shifts to bigger wavelengths, thus to a smaller plasmon frequency. The fact that the data from both these studies from the literature are associated with structures only of

Ag deposited on a substrate, instead of being a heterostructure of Ag and a semiconductor, could possibly be the origin of this difference in behaviour. Subsequently, the shift in frequency for contribution 2 was studied, which is shown in Figure 3.6 (A) and identified in Figure 3.6 (B).

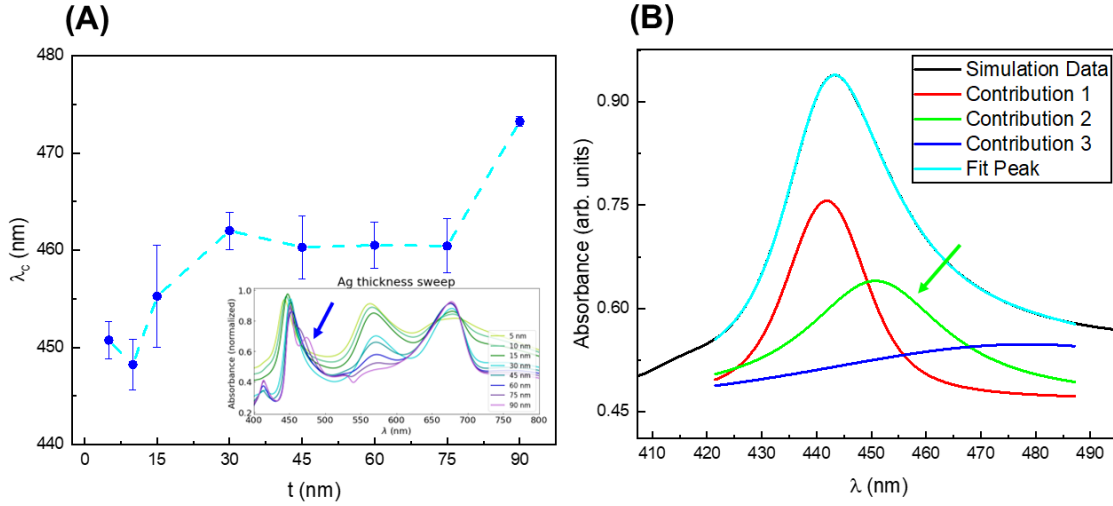


FIGURE 3.6: Frequency shift of contribution 2 of the first peak (A), signaled on the inset, and respective peak deconvolution (B) for 5 nm of Ag.

As it can be seen, for thickness values between 15 and 75 nm, the contribution 2 mode remained with a constant center wavelength λ_c . For smaller thickness values, the λ_c is slightly inferior, and for 90 nm it is relatively superior, inducing the emergence of a new peak on the absorbance spectrum. If this contribution would be from plasmonics events, we should observe a thickness dependent frequency shift for thickness below the silver penetration depth. Given that this effect is not observed in this contribution, it is unlikely that its origin is related to plasmonic phenomena. Instead, as the only significant shift occurs at the highest thickness, which approaches the boundary between the nano- and microscale, it is more plausible that this is a result of the intrinsic properties of the material. While simulating a greater silver thickness could help trace the shift of this mode and provide further insights, this approach would offer limited advantage due to the inherent constraints of silver's δ_m . For these reasons, the primary plasmonic contribution will be considered as Contribution 1. The same analysis was conducted for the second peak, with key results presented in Figure 3.7 (A). As shown in Figure 3.4, the second peak is coupled with the third, and as the thickness increases, the intensity of the second peak decreases, eventually becoming almost masked out by the third peak. Therefore, both peaks were fitted together to obtain more accurate results. In Figure 3.7 (B) it is possible to see that the second peak is made out of two contributions.

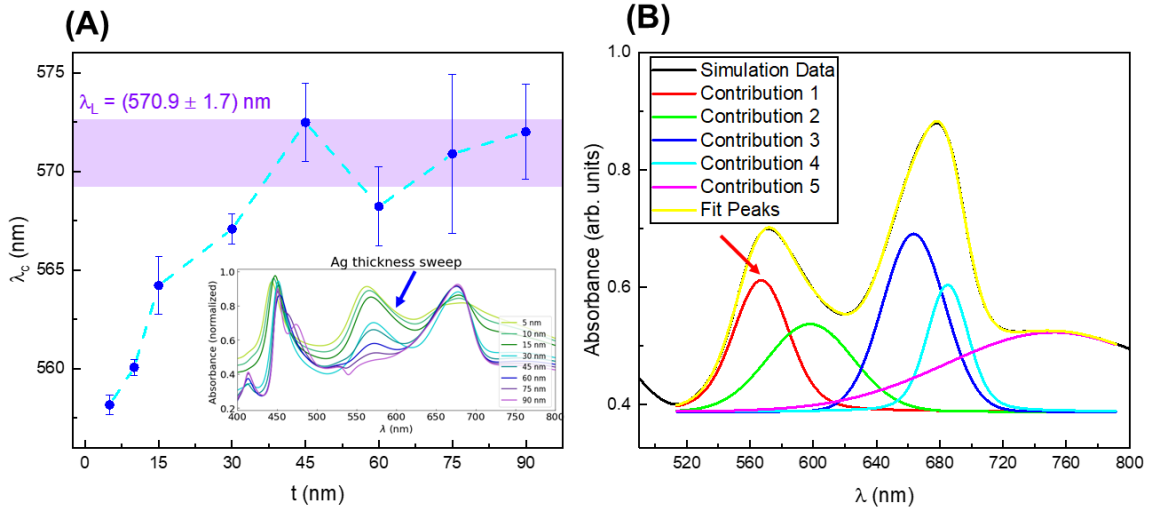


FIGURE 3.7: Frequency shift of the second peak (A), signaled on the inset. The only contribution for the second peak is the contribution 1 (B). The violet-shaded region encloses the λ and respective uncertainty.

Contribution 2 is only added to the fit in order to intermediate the coupling between the two peaks. Therefore, we will consider contribution 1 as the main mode.

This mode exhibits a similar behaviour to the first peak. Both shift to bigger wavelengths, thus smaller frequencies, as the Ag thickness is increased. However, while for the first one it seems it stops at a limiting wavelength for values up to 30 nm, the second one stops for values up to 45 nm at a wavelength $\lambda_L = 570.9$ nm. Thus, based on our results, for an excitation wavelength of 571 nm, the δ_m for silver is between 30 and 45 nm. Moreover, the data correspondent to thickness values above 30 nm have a high uncertainty associated, which makes the determination of the limiting wavelength λ_L more difficult. This is due to the fact that as the intensity of the peak starts to decrease, adding to the fact that it becomes more asymmetric, it becomes harder for the software to recognize the peaks, and therefore it adds more uncertainty while performing the fit.

Finally, the same analysis was performed on the third peak, whose results are displayed in Figure 3.8 (A). As seen in Figure 3.8 (B), three contributions are added to this peak. Contribution 5 is just to take into account the background signal of the spectrum, therefore, it will not be taken into account within this analysis. Contribution 3 does not exhibit any shift in frequency, therefore only exists to add asymmetry to the third peak. For that reason, only contribution 4 was considered as the main mode for this peak. The behaviour of this mode is different from the previous two. It follows the tendency of the already existing works of plasmonic frequency shifting with silver films [98] and gratings [99], shifting to smaller wavelengths and therefore bigger frequencies with increasing thickness.

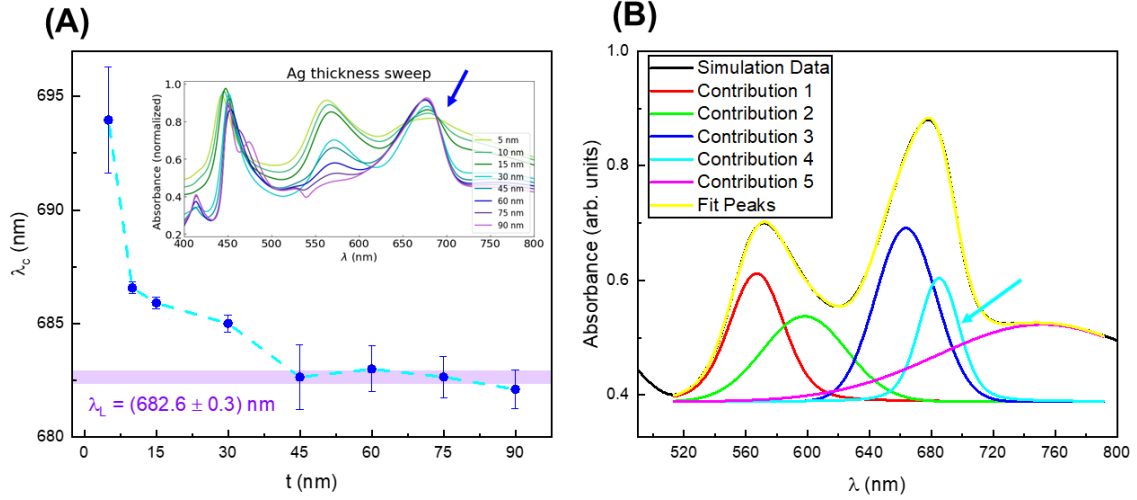


FIGURE 3.8: Frequency shift of the third peak (A), signaled on the inset. The only contribution for the second peak is the contribution 4 (B). The violet-shaded region encloses the λ and respective uncertainty.

However, similarly as the previous peak, it stops to shift at a silver thickness of 45 nm, at a limiting wavelength of $\lambda_L=683$ nm. This enhances the fact that these two peaks are coupled, and then their physics phenomena are correlated. If the opposite behaviour (shifting to bigger wavelengths) happens due to the fact that the dielectric medium in which the plasmon couples is the Bi_2Te_3 instead of the surrounding air, this could indicate that the third peak is associated with the generation of a plasmon at the interface of silver and air, whereas the previous two are associated with the interface between silver and Bi_2Te_3 . This is plausible, by recalling that the dielectric function for Bi_2Te_3 is way bigger than the one for air (close to 1), and therefore could lead to distinctive behaviours. Even if, for the already mentioned studies, the plasmon couples to their substrate (PMMA and quartz), the dielectric constant is still very different from Bi_2Te_3 , as shown in Figure 3.9.

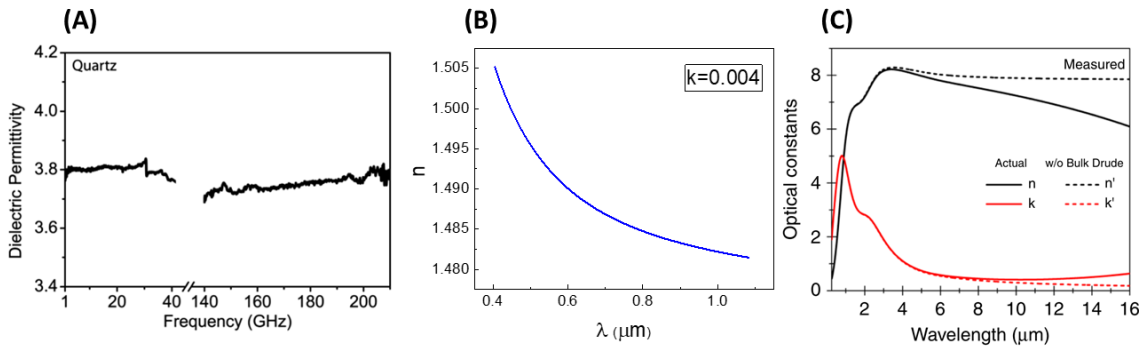


FIGURE 3.9: Dielectric function $\epsilon_r = (n - i\kappa)$ for quartz glass (A), PMMA (B) and Bi_2Te_3 (C). The imaginary part of ϵ_r for quartz can be considered near zero since it is a highly transparent material. From ref. [100], [92], and [93], respectively.

Despite supporting the previous argument, some caution is needed, as this could represent a leap of faith in the conclusions for this step. For example, the shift in wavelength for this study, approximately 10 nm, is smaller than for these studies, which was approximately 100-200 nm. This may be due to the fact that the input data consists of bulk values for the dielectric function, whereas the results from both these studies used not only input experimental values but also were compared with experimental data of the absorbance spectrum of these structures. To obtain more solid conclusions, a comparison with experimental data is needed, which will be made in the next chapter.

3.2.2 Bi_2Te_3 thickness sweep

In this step of the proposed study, focus was given on sweeping the thickness of Bi_2Te_3 between 5 and 90 nm while maintaining the Ag thickness fixed at 15 nm. The wavelength of the incoming light was also varied from 400 to 1600 nm in the perpendicular direction to the device, like in the previous sweep, and the same mesh configuration was maintained. The absorbance spectra for this sweep are shown in Figure 3.10.

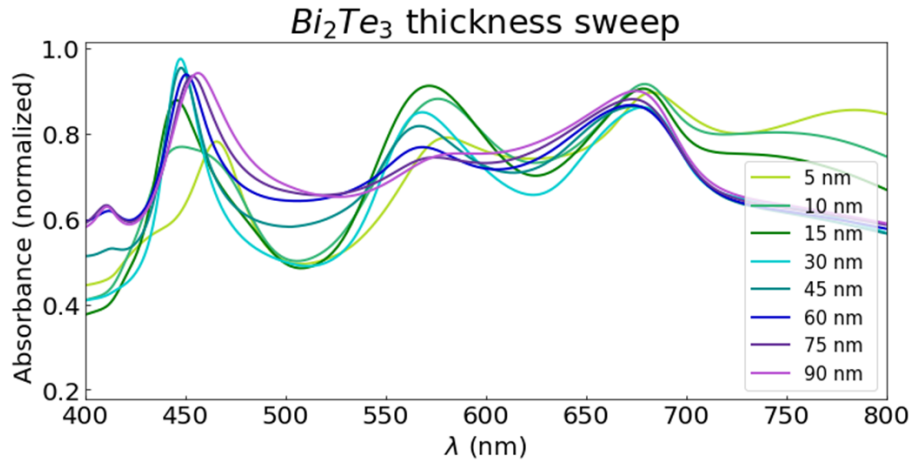


FIGURE 3.10: Absorbance spectra of the device while varying the Bi_2Te_3 layer thickness from 5 to 90 nm. As expected from the previous sweeping, three possible plasmon modes were observed.

The same three peaks were present on the simulated spectra. Therefore, the same analysis was performed for all of them. The behaviour of the frequency shift for the mode linked to contribution 1 is displayed in Figure 3.11. This mode shifts to bigger wavelengths when increasing the thickness of the Bi_2Te_3 , such as the case when increasing the thickness of the Ag layer. However, there is no critical thickness at which the frequency shifting stops. In addition, the maximum frequency shift is bigger for this case, 22 nm in wavelength-shift, rather than the other, 9 nm. Since the plasmon is excited on the silver layer, and, at this wavelength, the Bi_2Te_3 layer only contributes as the dielectric surrounding medium

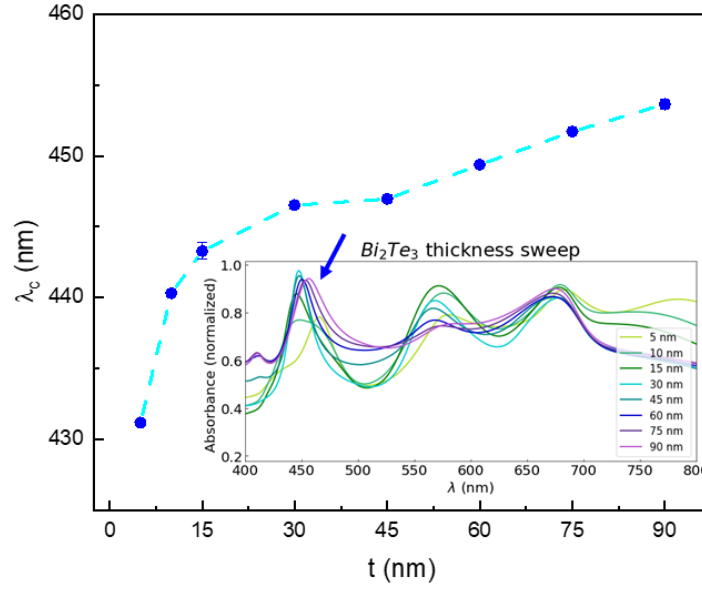


FIGURE 3.11: Frequency shift of contribution 1 of the first peak. The mode linked with contribution 1 increased its excitation wavelength when increasing the thickness.

that serves as the interface for the plasmon to be excited, the same behaviour was not expected for this sweeping like we obtained with the Ag thickness one.

To further understand the influence of Bi_2Te_3 in the absorbance spectra and in the frequency shifting of these peaks, the rest of the peaks were analyzed in order to get better conclusions. The second one is displayed in Figure 3.12.

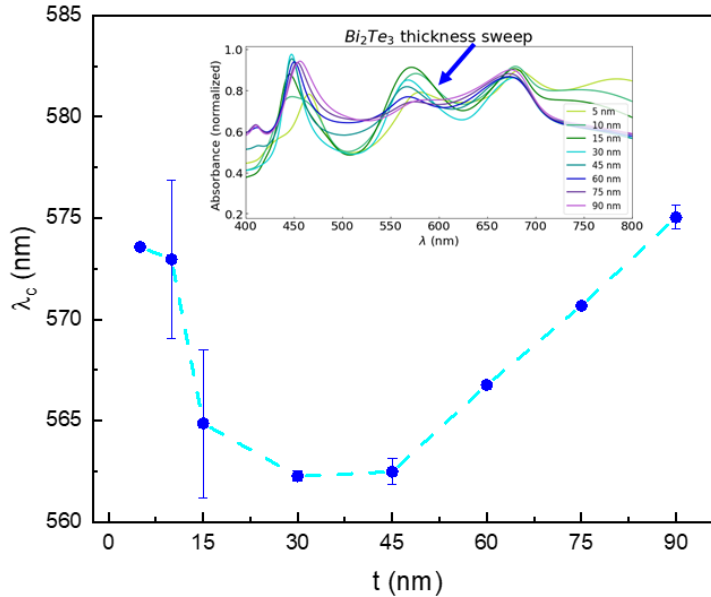


FIGURE 3.12: Frequency shift of contribution 1 of the second peak. The mode linked with contribution 1 decreased its excitation wavelength when increasing the thickness up to 30 nm and then increased.

The excitation's wavelength for this mode decreases when increasing the thickness of

Bi_2Te_3 up to 30 nm, and then increases. With the previous results in mind, this behaviour does not make sense. This shifting may come from the Bi_2Te_3 , which is a dielectric material with poor plasmonic properties, and therefore the frequency-shift would not be due to plasmonic phenomena, instead being attributed to the optical properties of the material. Subsequently, the same analysis was employed for the third peak, as shown in Figure 3.13.

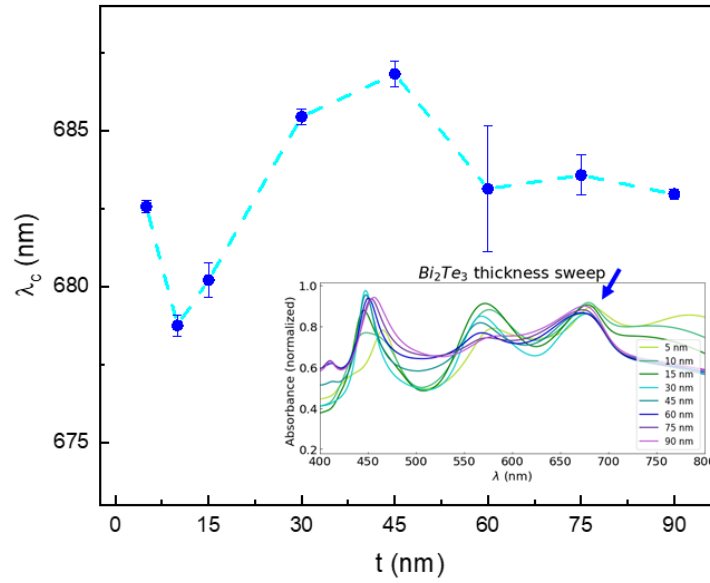


FIGURE 3.13: Frequency shift of contribution 4 of the third peak. The mode linked with contribution 4 does not exhibit any worth mentioning frequency dependent tendency with t .

No special tendency was observed concerning the frequency shifting. It seems that the frequency remains fixed for bigger Bi_2Te_3 thickness values, but it does not exhibit any tendency worth highlighting for smaller thicknesses. These results show that the effects of adding a silver layer are substantially different than the ones when adding a Bi_2Te_3 one. The Ag layer is responsible for a plasmonic frequency shifting, while the Bi_2Te_3 does not display any significant role for the absorbance peak displacement besides interface phenomena that could lead to peak deviation. A comparison with experimental data will be made in the next chapter to build a more solid basis for the knowledge and understanding of the physics within this model.

3.2.3 Grating geometries' variation

As it was already mentioned in Chapter 1, plasmonic resonances tend to shift in frequency by altering geometric parameters. The influence of sweeping the thickness of the material on top of the grating structures was already studied. Another important parameter to

shift is the grating pattern geometry. By recalling Figure 2.1, the reader can notice that the fabricated structures are not the ideal square-shaped grating previously simulated. Instead, there is the relaxation of the grating walls. For that reason, a ϕ angle was defined, which describes the angle between the square-shaped grating wall and the relaxed one, as schemed in the inset of Figure 3.14. It was assumed that, by increasing the ϕ angle, material would be deposited on the walls. The same was not assumed for an ideal square-shaped grating since it was admitted a conformal deposition would take place in the fabrication process. This angle was swept from 0 to 50 degrees while maintaining a periodicity of $p = 700$ nm, a silver layer thickness of 15 nm, and a Bi_2Te_3 one of 30 nm. The result of this sweeping is displayed in Figure 3.14.

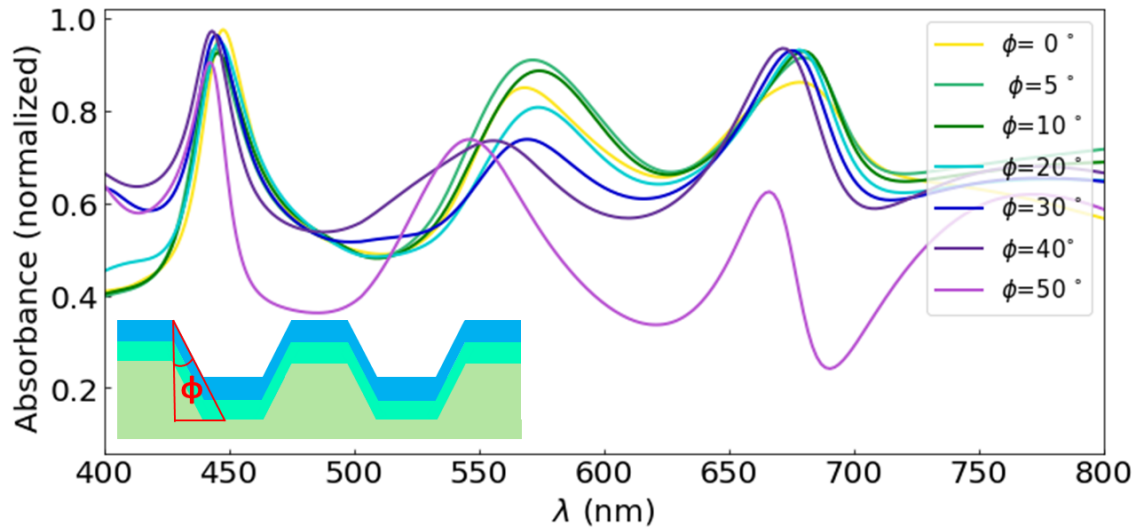


FIGURE 3.14: Absorbance spectra of the device while varying the ϕ angle from 0 to 50° nm. The ϕ angle is defined in the inset scheme.

By changing the geometry of the pattern, the peaks remained present in the spectra. Therefore, changing the geometry while maintaining the periodicity of the structures will not nullify the plasmonics' phenomena. The first peak remained almost unaltered by increasing ϕ . The shifting in frequency was analyzed, and it is displayed in Figure 3.15. The maximum wavelength shift for this mode is 4 nm, and there is not any special tendency worth mentioning. So it can be concluded that this peak is not dependent on the pattern geometry of the structures. Concerning the second peak, whose behaviour in frequency is represented in Figure 3.16, the conclusions are slightly different. This mode displays a maximum shift of 24 nm, so it is geometry-dependent. From $\phi = 10^\circ$ to 50° , this mode shifts to smaller wavelengths. However, between 0° and 10° , the wavelength for this peak increases with increasing ϕ .

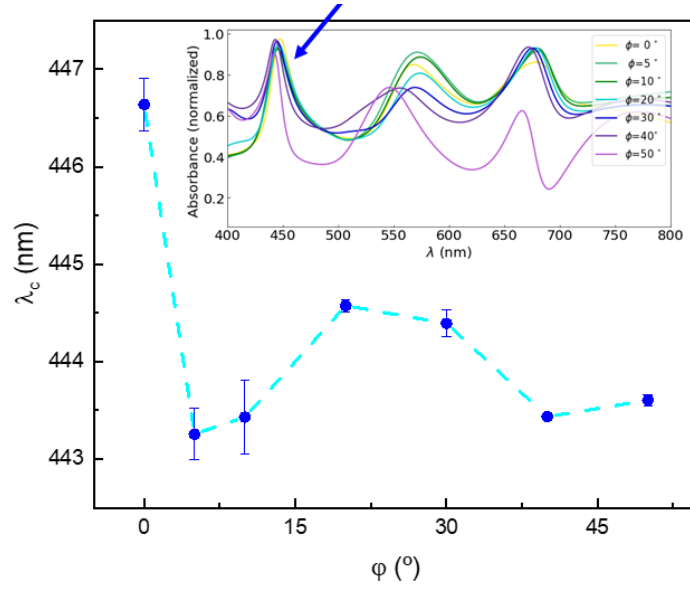


FIGURE 3.15: Frequency shift of contribution 2 of the first peak, signaled on the inset, when increasing ϕ . This mode does not exhibit any frequency-dependent behaviour .

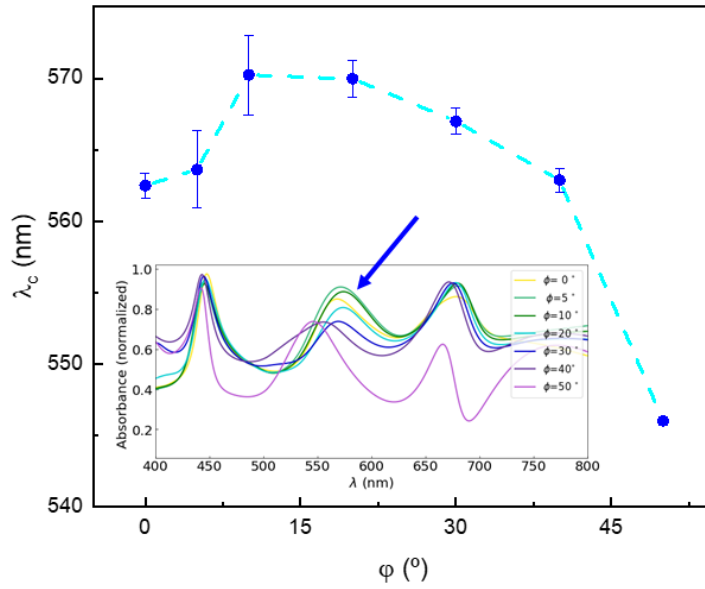


FIGURE 3.16: Frequency shift of contribution 1 of the second peak, signaled on the inset, when increasing ϕ (A). The mode linked with contribution 4 shifts to smaller λ_c when increasing ϕ from 10° to 50°

This shifting may not be only due to the increment of ϕ , but also due to the increment of thickness of the material deposited in the walls of the grating, which can be calculated by simple trigonometry. The thickness of the material deposited on the grating walls t' dependence with the ϕ angle and the thickness of material deposited on the top of the structures t is stated in equation 3.6.

$$t' = t \sin \phi \quad (3.6)$$

Additionally, for this peak, the absorbance also decreased from 0.91 to 0.73 when increasing ϕ from 5° to 50° . This does not affect the intensity of the phenomena on a large scale, so this does not represent a threat to the observation of the phenomena planned to be observed. Regarding the last peak, the same analysis is presented in Figure 3.17.

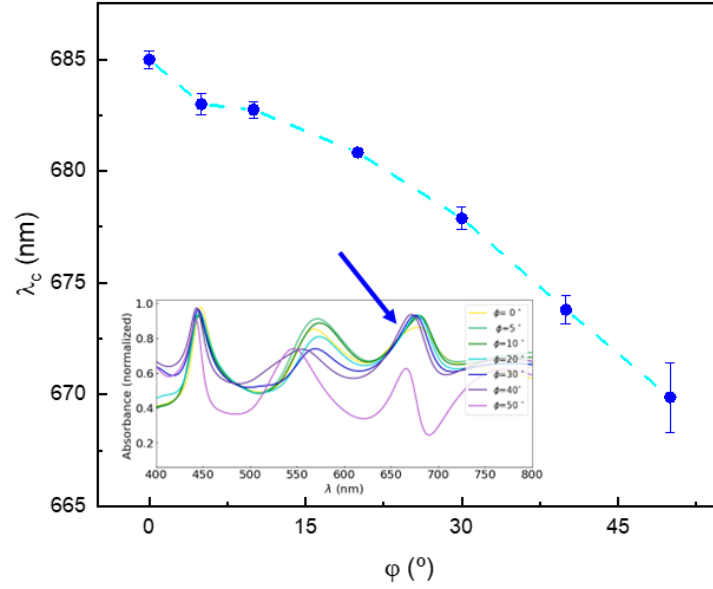


FIGURE 3.17: Frequency shift of contribution 4 of the third peak, signalized on the inset, when increasing ϕ . The mode linked with contribution 4 shifts to smaller λ_c when increasing ϕ .

The intensity of the peak remains constant for most ϕ values, excluding for $\phi=50^\circ$, where it decreases from 0.94 for $\phi=40^\circ$ to 0.62. Despite being superior to 0.5, it is not the most suitable value when dealing with plasmonic phenomena, given the fact that plasmonic events are resonance effects. Therefore, for this ϕ value there is the deterioration of the plasmonics effect. Concerning the excitation's wavelength shifting, when increasing ϕ , smaller λ_c are obtained for the main mode, thus bigger excitation frequency, with a maximum shift of 15 nm. Therefore, this mode is also geometry-dependent. The coupling between the second and third peak may be influencing the frequency shifting of this last one.

3.3. Main Conclusions

Micro-grating plasmonic structures were numerically simulated using PMMA as a substrate and covered by both Bi_2Te_3 and Ag. Three possible plasmonic peaks were identified, at the excitation's wavelength of 450 nm, 571 nm, and 683 nm. Their frequency shift was also analyzed along with the increment of material thickness. When increasing the silver thickness, the frequency stayed fixed at a limiting excitation's wavelength λ_L . It was

concluded that this is due to the overcoming of the penetration depth of Ag for the wavelength used. The third peak exhibits a different behaviour than the previous two, that likely must be due to different interfaces excitation of the plasmon. Moreover, the frequency-shifting behaviour when increasing the Bi_2Te_3 layer is not the same when increasing the Ag one. This is due to the fact that Ag is a noble metal and, therefore, has better plasmonic properties than Bi_2Te_3 , leading to different frequency-dependent behaviours. Later, the grating-pattern geometry was changed, being verified that the plasmonic peaks don't get nullified upon geometry variation. The first peak is geometry-independent, while the others two have a certain degree of dependence when increasing the ϕ angle. These results do not match 100% with the literature, so, a comparison of these results with experimental results is needed to gain a better insight into the physics of this model. This will be the next step of the present study, fully discussed in the next chapter.

4. UV-VIS-NIR Spectroscopy

When conducting numerical simulations, it is essential to include an experimental section to validate the model. This approach has been employed by other researchers in studies involving plasmonics, such as those addressing the plasmon dispersion relation of a material [99], and in the study of other physical phenomena [101]. Additionally, by comparing experimental data with theoretical predictions, it is possible to identify novel phenomena that were not considered in earlier models [8]. With this in mind, samples were fabricated following the procedure outlined in Chapter 2, with a periodicity of 725 nm, a fixed Ag thickness of 15 nm, and varying Bi_2Te_3 thickness from 5 nm to 107 nm, in order to measure their optical spectra using UV-VIS-NIR spectroscopy.

Experimental details: The equipment used was a UV-3600 Plus UV-VIS-NIR spectrophotometer from SHIMADZU in CF-UM-UP at the University of Minho. This equipment uses non-polarized light emitted from a lamp. The wavelength of the emitted light ranges from 400 to 2000 nm. To measure the samples, it was attached to the equipment an ISR-603 Integrating Sphere and placed a sample adaptor to only measure the signal coming from them.

However, some remarks must be made concerning the differences between the model where the numerical simulations took place and the experimental conditions at which the measurements were performed. First of all, in the simulations, the light used to excite the system was defined at the TM polarization state, whereas the spectrophotometer worked with non-polarized light emitted by a lamp. This itself already weakens the effect expected to be observed, as well as potentially altering some characteristics of the spectra. Moreover, the bulk real and imaginary components of the refractive index were given as input, whereas all measured samples have thicknesses in the order of some nanometers. Since the dielectric function $\epsilon_r(\omega)$ is altered from bulk samples to thin films [102], it is expected that the absorbance spectra may differ from bulk samples to thin films. Moreover, the model assumes conformal deposition. Since all samples were deposited using DC-magneto sputtering, which provides a non-conformal deposition, it is expected some extra-deviation between experimental and simulation results. In addition, the simulation assumes the material to be single-crystal, with perfect interfaces between adjacent materials and substrate. Given that, as discussed in Chapter 2, the material deposited on the

samples exhibits an amorphous structure, and as polymers do not exhibit perfect interfaces [103], it is also expected an extra contribution that may differ both simulation and experimental spectra.

4.1. Spectral Contribution Analysis

Firstly, it was identified from where it came all spectrum contributions. For that mean, it was measured the optical spectra from the substrate with no material deposited on top, one sample holding grating structures and covered only with Bi_2Te_3 , another covered with only Ag, another covered with both, and a sample without grating structures covered with both materials, as schemed in the Table 4.1.

TABLE 4.1: Samples measured under UV-VIS-NIR spectroscopy for the spectral contribution analysis

Sample	Structure	Deposited Material
1	Grating	Bi_2Te_3 and Ag
2	Grating	Bi_2Te_3
3	Grating	Ag
4	Film	Bi_2Te_3 and Ag
5	Film	None

The results for those measurements are displayed in Figure 4.1. As seen, the substrate had no contribution to the spectra. This is due to the fact that PMMA is a highly transparent polymer [104], therefore, it was expected such an absence of contribution. Then, the film with 38 nm of Bi_2Te_3 and 15 nm of Ag had an increasing absorbance with increasing frequency, exhibiting no peaks. Therefore, the heterostructure is intrinsically more absorptive for higher frequencies rather than for smaller ones. Then, concerning the grating samples' spectra, it was identified as the same film contribution as the background signal for all the grating samples. However, this signature had less expression for the Bi_2Te_3 grating sample, so, the presence of the silver on the structures has a bigger contribution in this feature. Furthermore, these spectra also highlight one common feature with the numerical simulations employed in Chapter 3. The grating samples' spectrum is shifted vertically to bigger values, relatively to the film spectra. Therefore, this highlights that the tailoring of structures to the sample enhances the absorbance. Additionally, for the mixed Bi_2Te_3 / Ag grating sample, bigger absorbance values than the Bi_2Te_3 are displayed, so the addition of the silver layers shifts the absorbance to bigger values. Following that, the peaks present in the grating samples were analyzed.

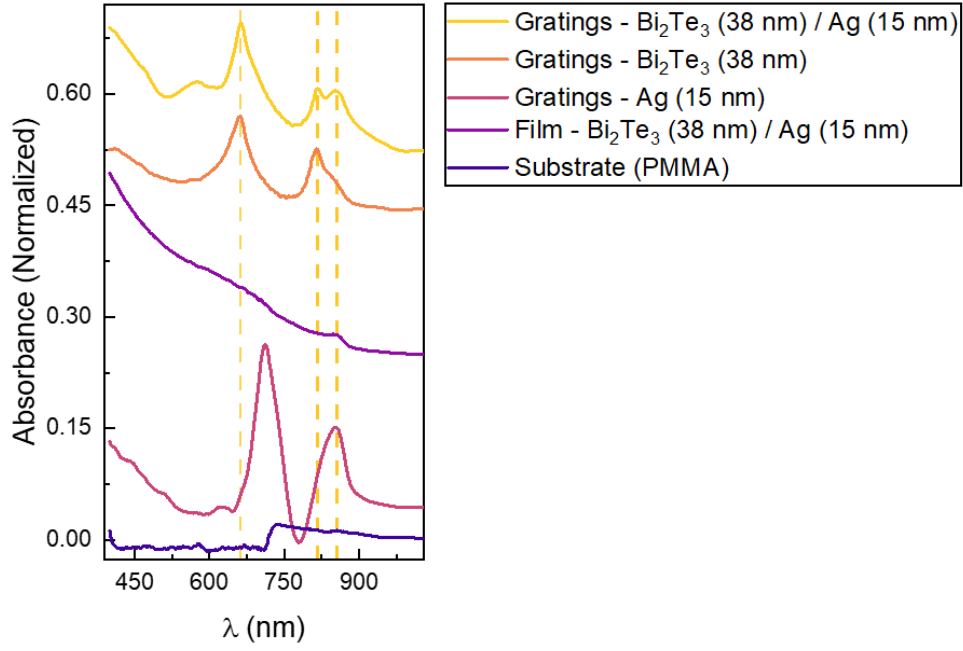


FIGURE 4.1: Absorbance spectra for grating samples covered by 15 nm of Ag, 38 nm of Bi_2Te_3 and both these materials, for a film covered by both these materials and the PMMA substrate.

A proper peak identification was made, and the same peak function to fit the data was used in order to have some consistency when comparing the experimental data with the simulation ones. The results are depicted in Figure 4.2 (A) for the Bi_2Te_3 / Ag and Bi_2Te_3 samples and in Figure 4.2 (B) for the Ag one.

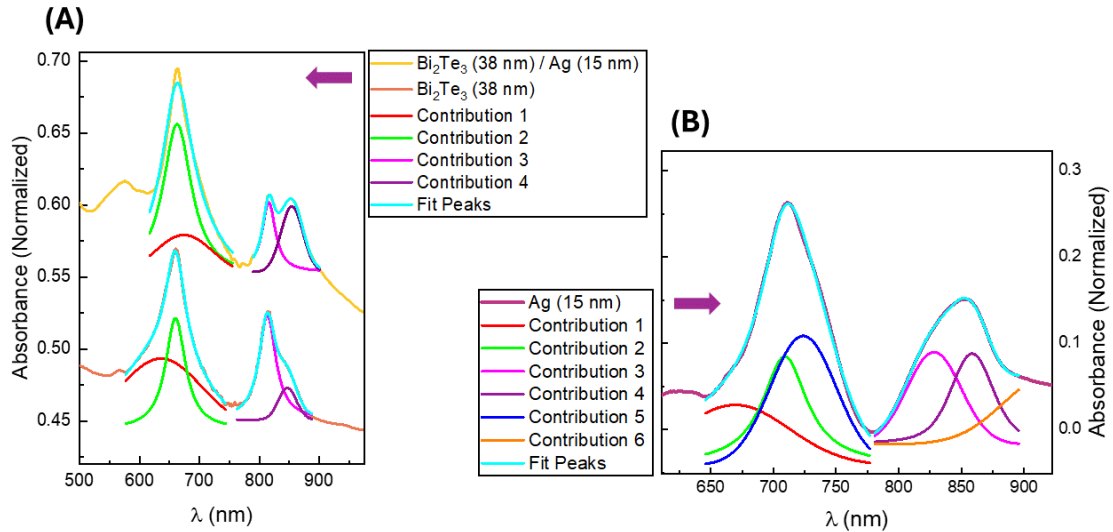


FIGURE 4.2: Peak identification for the grating sample covered only by 38 nm of Bi_2Te_3 , both Bi_2Te_3 and 15 nm of Ag (A) and only Ag (B). Contributions 2,3 and 4 are likely linked with plasmon modes.

In Figure 4.2 (A), contribution 1 takes only into account the background signal, therefore, it shall not be taken into consideration as a plasmonic contribution. The remaining identified

contributions will be considered to be linked to plasmonic modes. We identified three in both spectra. However, contribution 4 is more enhanced for the Bi_2Te_3 / Ag rather for the one only composed by Bi_2Te_3 . Concerning the spectra from the sample covered solely with 15 nm Ag, whose peak identification is made in Figure 4.2 (B), an extra contribution (5) was added to take into account the asymmetry of the first peak, and another extra contribution to the second peak was also added, to take into account the background signal. The mode linked to contribution 2 identified for the Bi_2Te_3 / Ag sample at a wavelength of $\lambda_c = (662.9 \pm 0.6)$ nm coincides with the one covered only with Bi_2Te_3 , identified at $\lambda_c = (660.1 \pm 0.1)$ nm. This one does not coincide with the one identified for the Ag sample, excited at a wavelength of $\lambda_c = (709.2 \pm 0.6)$ nm. We believe this means that, for our Bi_2Te_3 / Ag samples, the first peak must be due to the Bi_2Te_3 layer rather than the Ag. Subsequently, we see that the following peak is a convolution of two others. The one linked with contribution 3 was identified at a wavelength of $\lambda_c = (815.9 \pm 0.1)$ nm for Bi_2Te_3 / Ag and of $\lambda_c = (813.4 \pm 0.2)$ nm for the Bi_2Te_3 samples. For the Ag one, this contribution was identified at $\lambda_c = (828.3 \pm 17.9)$ nm, and we consider it to take into account the asymmetry of the peak rather than a fundamental mode. So, the emergence of the mode is linked with contribution 3 to the Bi_2Te_3 layer. Concerning the mode linked with contribution 4, it was identified at a wavelength of $\lambda_c = (854.4 \pm 0.1)$ nm for the Bi_2Te_3 / Ag and $\lambda_c = (858.6 \pm 1.7)$ nm for the Ag one. For the Bi_2Te_3 is not the main mode of the peak, being masked out by the previous contribution, and is identified at $\lambda_c = (846.4 \pm 0.5)$ nm. We believe this mode comes from the silver despite being present also in the Bi_2Te_3 one with smaller intensity.

4.2. Bi_2Te_3 thickness sweep

Then, the absorbance spectra was measured while varying the thickness of Bi_2Te_3 between 5 and 107 nm, while maintaining the Ag one fixed at 15 nm. The main results are displayed in Figure 4.3. Just by looking at the spectra, there is only one peak that remains present in all spectra, which is the second one in Figure 4.2 (A), which is identified in the range between $\lambda = 810$ nm and $\lambda = 880$ nm. The one excited for lower wavelengths, which is the first peak in Figure 4.2 (A) identified in the range between $\lambda = 660$ nm and $\lambda = 760$ nm, is present for all spectra except the one for a Bi_2Te_3 thickness of 70 nm. For this thickness, the spectrum seems to have a small bump near the same wavelength range that could represent this mode, but it is too masked out by the background signal and, therefore, difficult to identify.

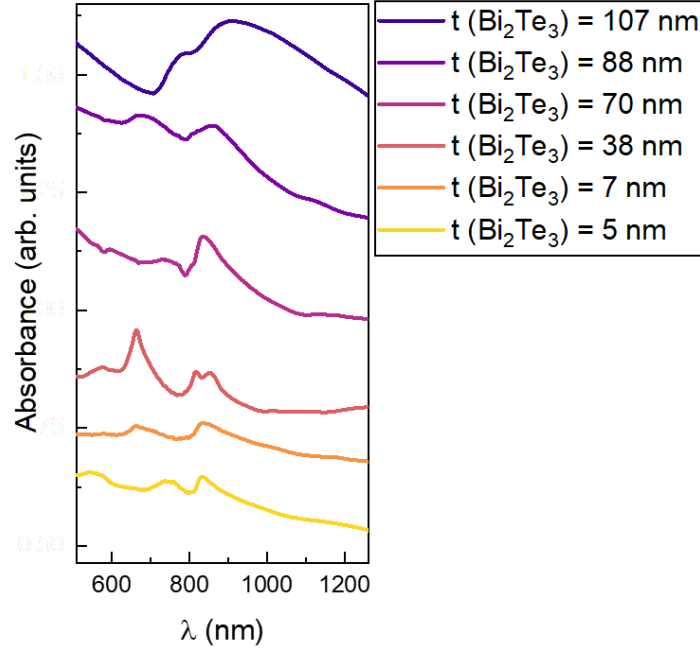


FIGURE 4.3: Absorbance spectra for several grating samples while sweeping the Bi_2Te_3 thickness from 5 to 107 nm. The second peak is present in all spectra, while the first one is present for all spectra besides the 70 nm one.

In addition, the spectra for Bi_2Te_3 thickness of 38 nm and 5 nm exhibit small bumps, located at $\lambda=573$ nm and $\lambda=553$ nm, respectively. However, since these are not present in the rest of the spectra, we will not focus on those. Besides that, another thing worth highlighting is that the intensity of the first peak does not respect any particular tendency, while the intensity of the second increases with increasing thickness and gets wider too. Plasmon modes tend to couple better when the resonances peak are more narrow [47], therefore, despite the resonance effect seems to be bigger for bigger thicknesses, the plasmon should be better coupled for the smaller ones.

Then, the experimental data was compared with the simulation data. A plot of the simulation and experimental data for the 38 nm Bi_2Te_3 sample is displayed in Figure 4.4. At first glance, the two data sets show some resemblance, as both display three peaks. However, the peaks in the experimental and simulation data are shifted. The first peak in the simulations, at $\lambda = (445.3 \pm 0.3)$ nm, is missing in the experimental data, while the last peak in the experimental data, formed by the combination of two modes, does not appear in the simulations. Furthermore, a small bump before the first peak in the experimental data aligns with the second peak in the simulations at $\lambda = (561.3 \pm 0.9)$ nm.

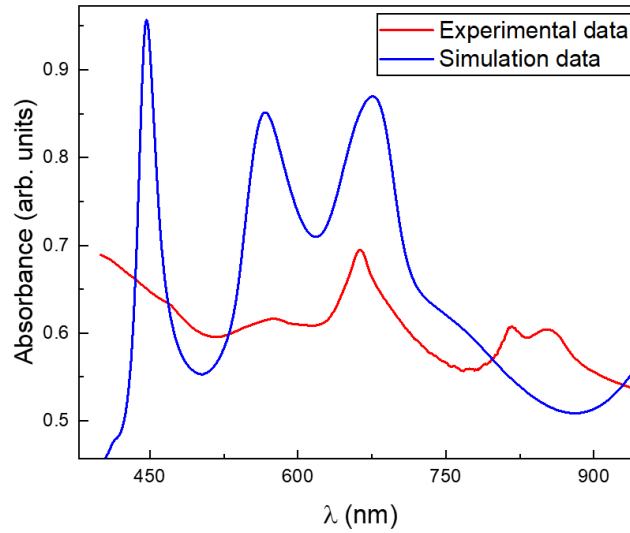


FIGURE 4.4: Comparison between the experimental and simulation spectra for a sample with 38 nm of Bi_2Te_3 . The experimental data does not follow the simulation predictions, since some peaks are unmatched.

The first peak in the experimental data corresponds to the last one in the simulations, at $\lambda = (680.6 \pm 2.0)$ nm. This raised some questions, as despite the apparent initial similarity, two peaks remain unmatched, and the small bump only appears in the spectra for samples with 5 and 38 nm of Bi_2Te_3 . Ultimately, the simulations and experimental results only fully align at one peak. Despite not being what it was ideally expected, it is not a surprise, given the fact that the simulation model and experimental conditions differ in many aspects. The simulation model uses bulk values for the refractive index, does not take into account the amorphous nature of the samples nor the irregularities of the interfaces between layers, and employs TM polarized light instead of the non-polarized light used in the UV-VIS-NIR spectrophotometer. All this factors altogether contribute to the considerable mismatch obtained.

Nevertheless, the plasmonic peaks still remain present, even though the experimental conditions were not the best for comparing both data. We just are not sure whether there is a big frequency deviation and the second simulation peak must match the first experimental peak, and so on with the following, or if in fact the small bump matches the $\lambda = (561.3 \pm 0.9)$ nm peak, the first experimental peak matches the $\lambda = (680.6 \pm 2.0)$ nm simulation one, and the last two peaks of the experimental data are peaks that come from the disorder of the system.

To probe that, we could repeat this measurements with a polarized light spectrophotometer. In that way, some error contribution could be minimized. Moreover, we also could measure the dielectric permittivity for the amorphous samples and use that value as the

input data in the numerical simulations. However, since these measurements were made in a late stage of this work, we did not have the time to fully take into place all these study steps.

4.3. Main Conclusions

All samples were successfully measured through UV-VIS-NIR spectroscopy. The substrate has no contribution to the absorption spectra and the samples are intrinsically more absorptive for higher frequencies. The grating samples exhibit peaks, however, only one of them was present in all measured samples' spectra. Despite that, the peak excited at $\lambda_c = (662.9 \pm 0.6)$ nm for the 38 nm Bi_2Te_3 thickness sample remained present in all spectra. There are some similarities between the simulation and experimental data, however, these do not match completely. This happens given the overall amount of extra parameters differing between the simulation model and the experimental conditions. Therefore, further characterization is needed to probe whether there is something wrong with the simulations or if it is not being taken into account any other contribution. Nevertheless, two possible plasmon modes were identified, one between 662 nm and 751 nm and the other between 817 nm and 879 nm.

5. Electronic Transport Measurements

While fabricating Topological Insulators, electronic transport measurements are one of the most important characterization steps since they give us a better insight into the transport behaviour happening within the samples and, therefore, help to better understand the quantum phenomena intended to be explored.

5.1. Resistivity Dependence on Temperature

The resistivity of the samples was firstly measured with varying temperature. This enables the identification of whether the sample exhibits either an insulating, semiconductor, or metallic behaviour [105]. Moreover, it is possible to extract several important information from these measurements concerning the conduction mechanisms within the material. To probe that, a Physical Properties Measuring System (PPMS) was used, since it not only allows the measurement of several physical phenomena, such as ferromagnetic resonance, FMR, photoconductivity, and heat capacity, but also displays a wide range of parameters available to sweep, such as temperature, ranging from 1.9 K to 400 K [106]. The samples were measured under a four-probe setup. Figure 5.1 shows a sample with no structures, deposited with both 18 nm of Bi_2Te_3 and 15 nm of Ag on PMMA already prepared for measurement under the four-probe setup, where the copper wires were fixed with a specialized silver ink made out of a mixture of two silver epoxies from Quantum Design.

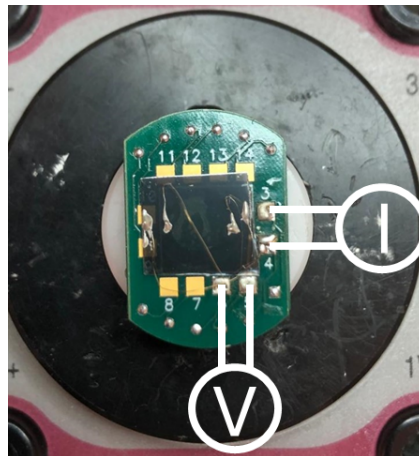


FIGURE 5.1: Picture of the 18 nm Bi_2Te_3 and 15 nm Ag film fixed on the PPMS puck. The outer contacts are applying current, while the inner ones measure voltage. The inner 0.5 cm diameter circle does not hold any contact since it is there where the silver is deposited, as before-mentioned in Figure 2.8 (B).

Experimental details: To measure the resistance over temperature a Quantum Design Physical Property Measurement System (PPMS) Model 6000 was used. The resistance of the samples was measured in a range of temperatures between 3 and 300 K in response to an AC electrical current of 0.1 mA amplitude and 0.4 Hz frequency. The samples were installed in an adequate puck, provided by Quantum Design, and inserted into the pressurized sample chamber.

Samples with 18 nm and 38 nm of Bi_2Te_3 , with and without grating structures were measured. For most of them, the thickness of Ag was fixed at 15 nm, just like in the previous chapter. Table 5.1 displays the measured samples alongside their features.

TABLE 5.1: Samples measured under the four-probe setup for the resistance over temperature characterization

Sample	Structure	Bi_2Te_3 thickness (nm)	Ag thickness (nm)
1	Grating	18	15
2	Film	18	15
3	Film	18	0
4	Grating	38	15
5	Film	38	15

Then, by using Equation 5.1, the 2D resistivity ρ was computed, where a geometrical correction $C=0.4292$ was applied [107] to calculate the sheet resistance R_s from the resistance R values, that together with the thickness t enabled us to get the final value.

$$\rho = R_s t = C \frac{\pi}{\ln 2} R t \quad (5.1)$$

Figure 5.2 shows the results for samples 1 and 2, together with an inset of the natural logarithm of the experimental data. One common feature to the data for both film and grating samples is that their resistivity increases abruptly for low temperatures, becoming highly resistive, just like in the freeze out zone of an extrinsic semiconductor [108]. However, to better understand the main mechanism of conduction, we fitted the present data to the Mott's Variable Range Hopping model [109]. This model typically is used for strongly disordered materials, describing the resistivity dependence on temperature, that arises from thermally assisted variable-range hopping between electrons' localized states differing in energy [110].

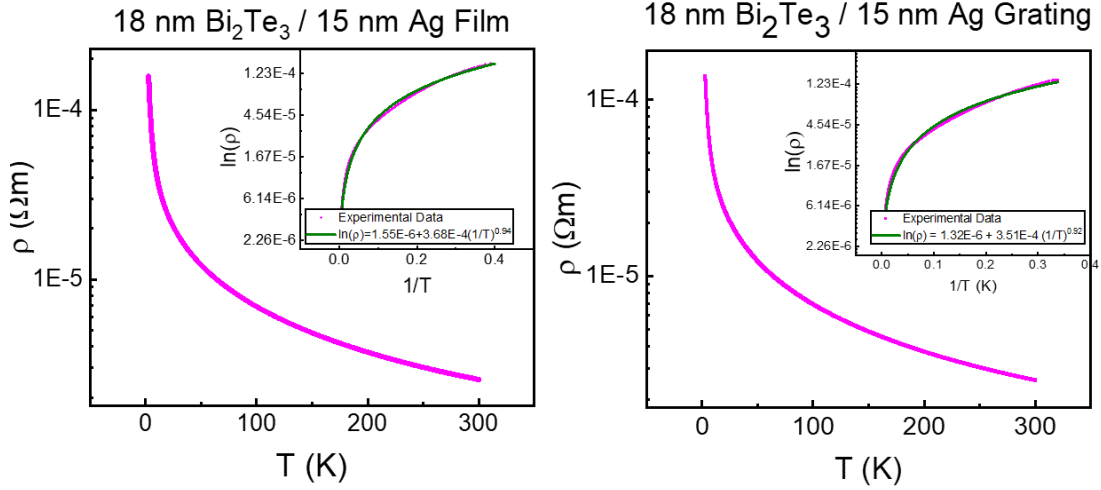


FIGURE 5.2: Resistivity dependence on temperature for a film (left) and grating (right) samples with 15 nm of Ag and 18 nm of Bi_2Te_3 . Both graphs exhibit their y-axis in the logarithmic scale and both inset in the natural logarithmic scale.

This dependence is stated in Equation 5.2, where ρ_0 is an pre-exponential factor, T_0 is defined in Equation 5.3, depending on b , which is a numerical factor, k_B the Boltzmann constant, $g(\mu)$ the density of states at the Fermi level, a the localization radius of states near the Fermi level [109] and γ is defined in Equation 5.4 depending on the d dimensionality of the system [110].

$$\rho(T) = \rho_0 \exp \left[\left(\frac{T_0}{T} \right)^\gamma \right] \quad (5.2)$$

$$T_0 = \frac{b}{k_B g(\mu) a^3} \quad (5.3)$$

$$\gamma = \frac{1}{1+d} \quad (5.4)$$

Therefore, by applying the natural logarithm to the data, the dimensionality of the main conduction mechanism happening within the samples is extracted, as shown in Equation 5.5.

$$\ln(\rho) = \ln(\rho_0) + \left(\frac{T_0}{T} \right)^{\frac{1}{1+d}} \quad (5.5)$$

Since the samples are amorphous, as mentioned in Chapter 2, this model is suitable to use. The main results for this fit are displayed in Table 5.2.

TABLE 5.2: Results obtained for the γ coefficient obtained through the Mott's Variable Range Hopping model for samples 1 and 2.

Structure	Bi_2Te_3 thickness (nm)	Ag thickness (nm)	$\gamma \pm u(\gamma)$
Film	18	15	0.940 ± 0.001
Grating	18	15	0.918 ± 0.001

Both these values are very close to 1, which means their dimensionality is $d = 0$. A system with zero dimensionality is the equivalent of a quantum dot, which exhibits quantum confinement in three dimensions [111] and their size range from 1 to 10 nm [112]. By recalling Chapter 2, when deposited on glass, the XRD measurements revealed that the sample has a crystallite size of 10 nm for the identified peaks, so with PMMA, despite no peaks being observed, the crystallite size should be smaller. Therefore, it is possible that the inter-grain resistance is so high that this system behaves as multiple quantum dots, and the transport properties are dominated by these localized structures. This would eventually add up as another contribution justifying the mismatch observed on the absorbance spectra between numerical simulations and UV-VIS-NIR spectroscopy. The simulation's model considers the material to be single-crystal. When entering this degree of quantum confinement, the optical properties of the materials are altered [113]. Therefore, a highly amorphous structure, that potentially is based on an ensemble of quantum dots, must not display the same absorbance spectra as a single crystal-layer structure. Additionally, the data also show that the existence of structures tailored to the samples does not alter their resistivity dependence on temperature, nor does it alter the dimensionality of the system. Then, to study whether the thickness has any influence on the dimensionality of this system, film and grating samples with 38 nm of Bi_2Te_3 and 15 nm of Ag were measured. Figure 5.3 shows the results for these samples, together with an inset of the natural logarithm of the experimental data.

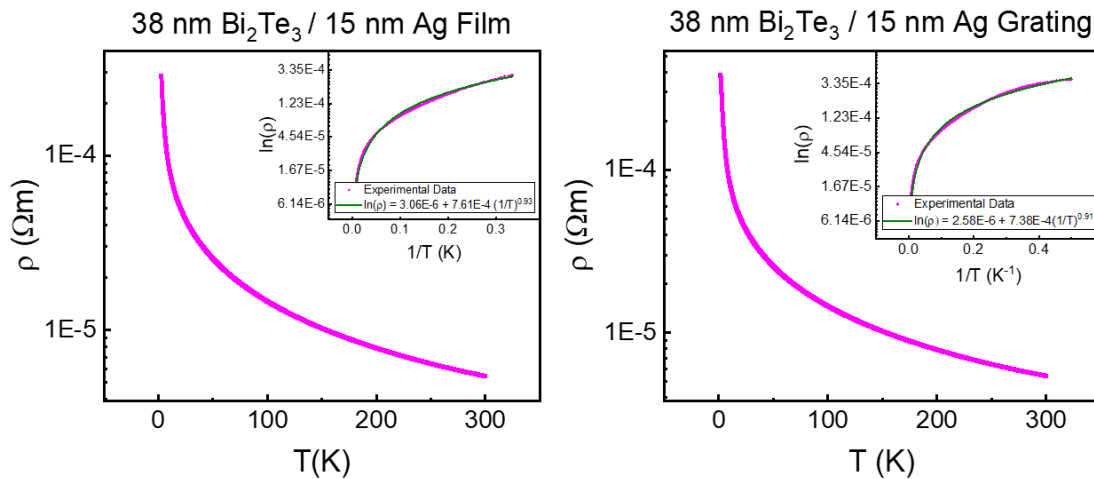


FIGURE 5.3: Resistivity dependence on temperature for a film (left) and grating (right) samples with 15 nm of Ag and 38 nm of Bi_2Te_3 . Both graphs exhibit their y-axis in the logarithmic scale and both inset in the natural logarithmic scale.

The main results of the data fitted with the Mott's Variable Range Hopping for these samples are depicted in Table 5.3.

TABLE 5.3: Results obtained for the γ coefficient obtained through the Mott's Variable Range Hopping model for samples 1 and 2.

Structure	Bi ₂ Te ₃ thickness (nm)	Ag thickness (nm)	$\gamma \pm u(\gamma)$
Film	38	15	0.930 ± 0.001
Grating	38	15	0.913 ± 0.001

The data for these samples follows the same tendency as for the previous ones, so concerning the 38 nm samples, the transport properties are dominated by the ensemble of quantum dots arising within the amorphousness of the material.

To address whether the Ag spot deposited on top of the samples had any influence on the previously analyzed samples, we also measured a thin film of Bi₂Te₃ with no Ag spot deposited on top. Figure 5.4 depicts the results for sample 3, a film with only 18 nm of Bi₂Te₃, together with an inset of the natural logarithm of the experimental data.

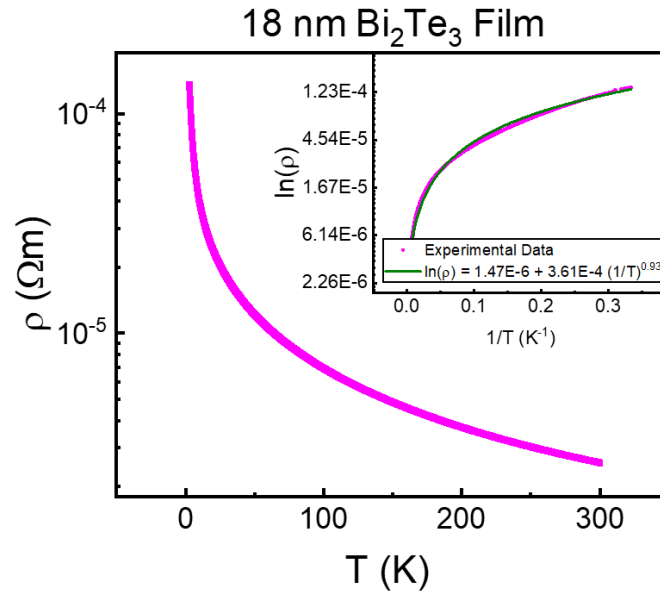


FIGURE 5.4: Resistivity dependence on temperature for a film with 18 nm of Bi₂Te₃. The main graph y-axis exhibits the resistivity in the logarithmic scale and the inset one in the natural logarithmic scale.

The γ coefficient for this sample was $\gamma=(0.932 \pm 0.001)$, which is also close to 1 and therefore this sample exhibits the same $d = 0$ dimensionality as the previous ones. Therefore, the existence of the Ag spot deposited on top, not only does not alter the resistivity dependence on temperature but also does not alter the dimensionality of the system.

5.2. Magnetoresistance Measurements

Characterizing these samples through magnetoconductance measurements not only helps to further extend the understanding of the transport properties of the samples, but also can provide signatures of the topological features displayed by them. As previously mentioned in Chapter 1, at cryogenic temperatures, a quantum correction arises on the magnetoresistance data of topological insulators, such as Bi_2Te_3 [114], from which several information can be extracted by using the HLN model, stated on Equation 1.3. With that in mind, the resistance of the samples listed in table 5.4 was measured while sweeping the applied longitudinal magnetic field from -8 T to 8 T, at temperatures ranging from 3 K to 20 K.

TABLE 5.4: Samples measured under the four-probe setup for the magnetoresistance characterization

Sample	Structure	Bi_2Te_3 thickness (nm)	Ag thickness (nm)
1	Grating	18	15
2	Film	18	15
3	Film	18	0
4	Grating	38	15
5	Film	38	15

Experimental details: The equipment used for the magnetoresistance measurements of the samples was a Quantum Design Physical Property Measurement System (PPMS) Model 6000. The resistance of the samples was measured under a four-probe setup in a range from -8 T to 8 T of the out of plane applied magnetic field in response to an AC electrical current of 0.1 mA amplitude and 0.4 Hz frequency. The samples were installed in an adequate puck, provided by Quantum Design, and inserted into the pressurized sample chamber.

Equation 5.6 was used to compute the magnetoconductance $\Delta\sigma_{2D}$ of the samples, where t is the thickness of the samples, $C=0.4292$ is the geometrical correction [107], R is the measured resistance, R_0 is the measured resistance at zero field, h is the Planck's constant, and e is the electron's charge.

$$\Delta\sigma_{2D} = \frac{h}{e^2} \frac{\ln(2)}{\pi t C} \cdot \left(\frac{1}{R} - \frac{1}{R_0} \right) \quad (5.6)$$

Figure 5.5 shows the results for the magnetoconductance of samples 2 and 5 from Table 5.4.

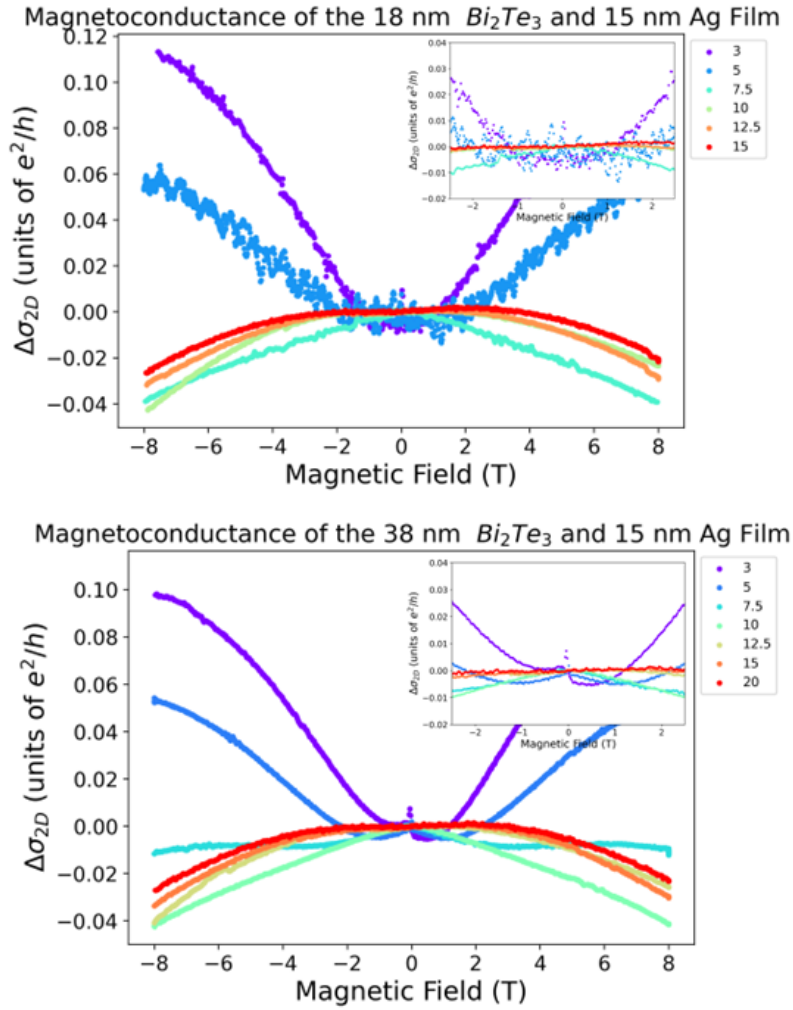


FIGURE 5.5: Magnetoconductance of the 18 nm (top) and 38 nm (bottom) Bi_2Te_3 with the 15 nm Ag spot deposited on top of the films. Both data exhibit an interplay between the WL and WAL, where the WL dominates for low temperatures.

From the work reported on [20], with Bi_2Te_3 deposited on kapton, the authors observed that the magnetoconductance suffered an increment at zero applied magnetic field due to the quantum correction that arises from the WAL effect. However, for the new results coming from these samples, there is the opposite correction for smaller temperatures, in which the magnetoconductance decreases at zero applied magnetic field. Then, the behaviour is inverted between 7.5 K and 10 K, where the conductance is increased for low applied magnetic fields. This inversion of behaviour was already observed for Bi_2Te_3 nanowires [115], Cr doped Bi_2Se_3 films on sapphire substrate deposited by molecular beam epitaxy (MBE) [116], and for carbon nanowalls [117]. In those studies, it was concluded that this inversion of behaviour was due to a competition between the WL and WAL

effect. In particular, for the case of the Bi_2Se_3 films doped with Cr, ARPES measurements were employed on the samples, showing that above a certain percentage of doping, the topological surface states were eliminated due to the breaking of the TRS by the magnetic impurities. For intermediate percentages of doping, the ARPES spectra showed that the surface states were weakened and the gap amplitude increased. Despite the measured samples not having this type of impurity incorporation, it could be that we are weakening their surface states and increasing as well their gap amplitude. However, this weakening may not be caused by doping, but by the macro-scale disorder of the samples, due to their amorphousness, as previously discussed in Chapter 2. This led to the emergence of three dimensional quantum spatial confinement typical of quantum dots, as already discussed in the previous section, localising some electrons with a definite spin. Due to energy considerations, these electrons have a spin antiparallel to the moving electrons [118]. When applying magnetic field, all spins start to align accordingly. Since the spin reversal of the conducting electrons is one type of scattering mechanism [119], by applying magnetic field and aligning their spins, this type of scattering is eliminated, which increases the conductance of the sample. This would lead to a competition between the two effects, so that the $\Delta\sigma_{2D}$ would be dominated by the WL effect at the lower temperatures (≤ 7.5 K) and by the WAL effect at higher temperatures (≥ 10 K). This is consistent with the fact that the WL effect is observable in disordered systems [17]. As a matter of fact, this amorphous character's was already reported to be the main mechanism behind the observation of a similar competition between the WL and WAL effects on carbon nanowalls structures [117].

As temperature is increased, this correction gets wider, transitioning to the opposite behaviour between 7.5 K and 10 K for both samples. Then, the curve at 10 K does not exhibit a typical WAL profile since an almost linear response is displayed with the magnetic field. This was already observed for non-magnetic silver chalcogenides [120], and a theoretical model was also proposed for heavily disordered semiconductors [121]. This linear behaviour was also observed in Bi_2Te_3 films [122], attributed to a classical MR contribution that should be taken into account together with the WAL effect [123]. For all these studies, this feature was destroyed when increasing the temperature, where the curves got wider and exhibited the quadratic behaviour our data also exhibit. This widening could indicate the phenomenon got weakened. However, these studies exhibited a resistivity dependence on temperature different than the present results and less resistive samples, so the underlying physical phenomena may not be exactly the same.

Then, to address whether this behaviour could be influenced by the silver layer on top, we measured the 18 nm Bi_2Te_3 film, correspondent to sample 3 from Table 5.4. Figure 5.6 depicts the results for these samples, depicting the same behaviour observed for the previous two samples.

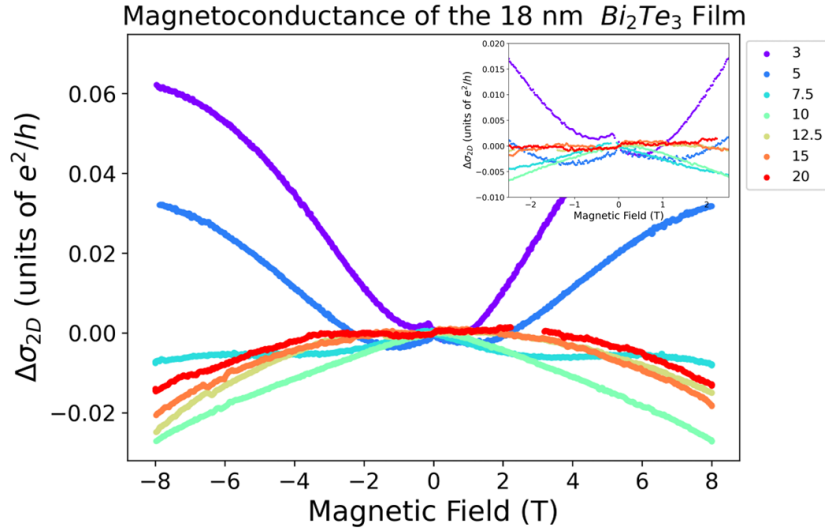


FIGURE 5.6: Magnetoconductance of the 18 nm Bi_2Te_3 sample. There is also an interplay between the WL and WAL, where the WL dominates for low temperatures.

This similarity enhances that this competition comes from the disorder of the samples instead of having any influence on the material deposited on top. Then samples 1 and 4 from Table 5.4, which are both grating samples with 18 nm and 38 nm of Bi_2Te_3 and with the 15 nm of Ag circular spot on top, were measured to probe whether the existence of structures tailored to the sample could have an influence on these effects, since it may increase the disorder of the system. Figure 5.7 shows the results for these samples, where it is evident the competition of the WL and WAL effects remains present in a similar way. These data seem to be noisier than the film ones. This could be due to the extra disorder induced by the non-flat substrate or just a contact issue. However, since the behaviour remains present and the transition occurs between the same temperatures (between 7.5 K and 10 K), the existence of extra disorder provided by tailored substrate does not alter the competition between the WL and WAL effects.

Subsequently, the HLN model was used to fit the data and extract several important information, such as the α coefficient and the phase coherent length L_ϕ . Equation 1.3 states the HLN model with a quadratic background. However, to take into account the classical linear MR effect our data exhibits, a linear contribution in the background was introduced, as previously done for previous results within the research group [20].

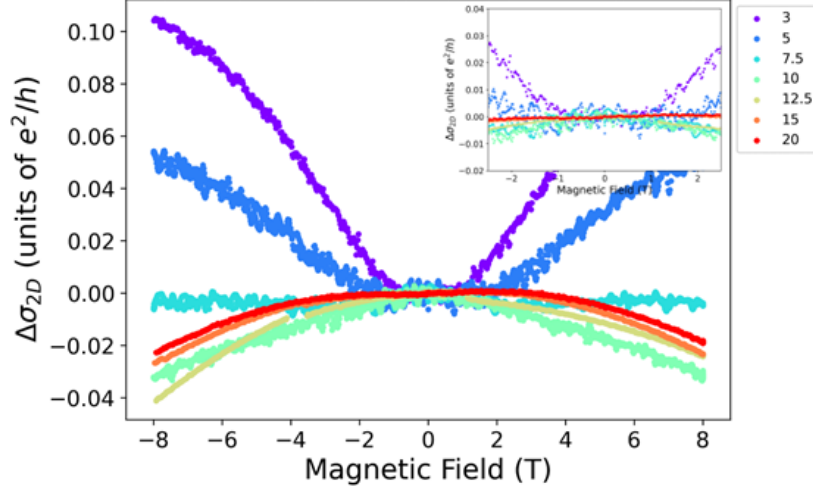
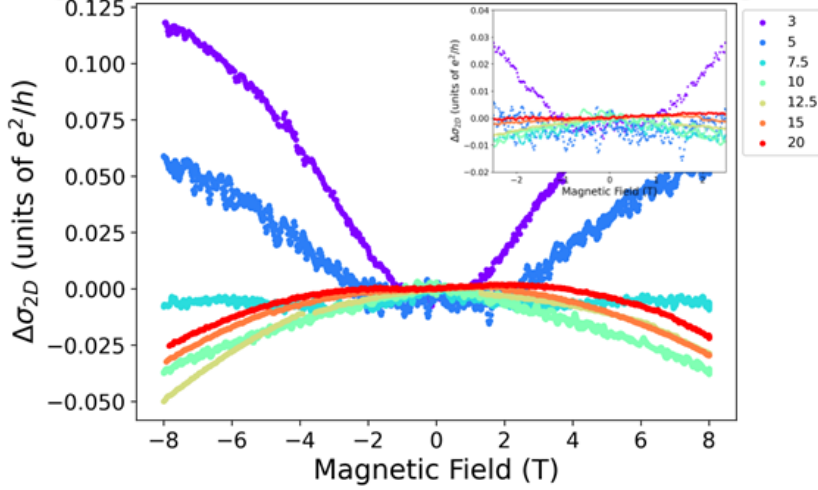
Magnetoeconductance of the 18 nm Bi_2Te_3 and 15 nm Ag GratingMagnetoeconductance of the 38 nm Bi_2Te_3 and 15 nm Ag Grating

FIGURE 5.7: Magnetoeconductance of the 18 nm (top) and 38 nm (bottom) Bi_2Te_3 with the 15 nm Ag spot on top grating samples. Both data are noisier than the latter results, however, they also exhibit an interplay between the WL and WAL, where the WL dominates for low temperatures.

In that way, the updated HLN model is described by Equation 5.7, where dH takes into account any offset displayed by our data, a is the linear background term, and c takes into account any vertical offset displayed by our data.

$$\Delta\sigma(H) = \frac{\alpha e^2}{\pi h} \left(\ln \left(\frac{B_\psi}{H + dH} \right) - \Psi \left(\frac{1}{2} + \frac{B_\psi}{H + dH} \right) \right) + \beta H^2 + aB + c \quad (5.7)$$

To take into account the competition between the WL and WAL effects, the WL contribution of the HLN model was summed to the WAL one. This is shown in Equation 5.8 [124], where $i = 0, 1$ refers to the WL and WAL contribution respectively, and $B_\psi(i)$ is defined in Equation 5.9.

$$\sum_{i=0,1} \Delta\sigma(H) = \frac{\alpha(i)e^2}{\pi h} \left(\ln \left(\frac{B_\psi(i)}{H + dH} \right) - \Psi \left(\frac{1}{2} + \frac{B_\psi(i)}{H + dH} \right) \right) + \beta H^2 + aB + c \quad (5.8)$$

$$B_\psi(i) = \frac{h}{8e\pi(L_\phi(i))^2} \quad (5.9)$$

However, by fitting this sum, there is no good convergence on the fit parameters due to over-parametrization of the $\Delta\sigma(H)$ function. Consequently, the data was fitted just by considering the WL term, assuming that it dominates the data between 3 K and 7.5 K, rather than adding up the two terms. For 10 K to 20 K, the WAL contribution was assumed to dominate and the data was fitted solely taking this one into account. The fitted data is all displayed on the Appendix B.3, together with all the fit parameters. The main results, enclosed to the $\alpha(0)$ and $l_\phi(0)$, are displayed in Figure 5.8, for the data between 3 K to 7.5 K.

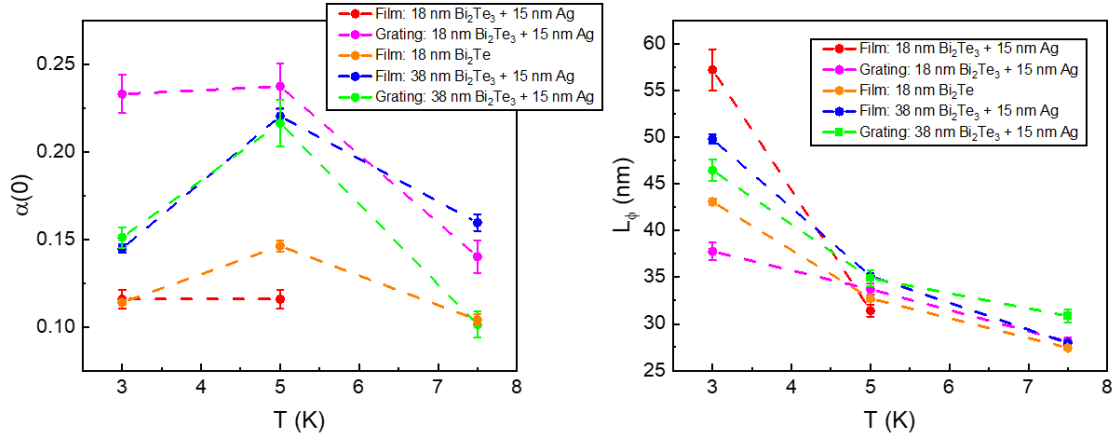


FIGURE 5.8: Dependence of the $\alpha(0)$ and $l_\phi(0)$ on temperature. The existence of structures tailored to the samples altered both coefficients.

Since there is a small amount of data for the $\alpha(0)$ and $l_\phi(0)$ coefficients (given the transition between the WL effect dominance and the WAL one occurs already at 7.5 K), it is not useful to plot them as a function of temperature and analyse their decay relations. At 3 K, the phenomenon is better protected. At this temperature we get the highest and most representative value for the $l_\phi(0)$, which decreases with increasing temperature. The values obtained for this coefficient are similar to the ones obtained for Bi_2Te_3 deposited on kapton by the research group [114]. In TI, large l_ϕ that significantly exceed both the mean free path and the sample thickness are associated with a 2D coherent transport process [125], since the electron can travel through the sample thickness without losing coherence. The fact that the $l_\phi(0)$ remains comparably bigger to the sample thickness for the 18 nm sample could be interpreted as the samples having a 2D coherent transport. The

same cannot be concluded regarding the 38 nm sample. The $\alpha(0)$ value also changes with increasing temperature, however, the highest value occurs at a temperature of 5 K, instead of occurring at the lowest temperature, as previously reported for previous results within the research team [114], and for other sputtered topological insulator thin films, like Bi_2Se_3 [125]. However, these values are within the range $[0, 1/2]$ expected for the WL effect [21]. Moreover, the tailoring of structures seems to increase the $\alpha(0)$ coefficient at 3 K. However, for bigger temperatures, the $\alpha(0)$ coefficient seems to be smaller for the grating samples than the one for the films. Therefore, we cannot conclude whether the localization mechanism is strengthened by the tailoring of structures. Concerning the $l_\phi(0)$ coefficient, the same is verified.

Table 5.5 displays the main results for the WAL term, observable for temperatures between 10 K and 20 K, correspondent to the samples 1, 2, 4, and 5 from Table 5.4.

TABLE 5.5: Results for the coefficients $\alpha(1)$ and $l_\phi(1)$ obtained through the HLN fit, stated in Equation 5.8 for samples 1, 2, 4, and 5, all with 15 nm of Ag deposited on top, in the range of temperatures between 10 K and 20 K

Structure	$t(\text{Bi}_2\text{Te}_3) / \text{nm}$	T / K	$\alpha(1) \pm u(\alpha(1))$	$l_\phi(1) \pm u(l_\phi(1)) / \text{nm}$
Film	18	10.0	-0.0026 ± 0.0011	104.219 ± 73.590
		12.5	-0.0081 ± 0.0012	36.211 ± 2.252
		15.0	-0.0001 ± 0.0781	5.817 ± 1100.160
		20.0	0.0002 ± 0.0995	5.843 ± 696.248
Grating	18	10.0	-0.0021 ± 0.0011	129.336 ± 131.255
		12.5	0.0002 ± 0.0132	14.154 ± 257.725
		15.0	-0.0204 ± 0.0021	24.014 ± 0.354
		20.0	-0.0205 ± 0.0050	18.279 ± 0.687
Film	38	10.0	-0.0242 ± 0.0192	10.303 ± 3.501
		12.5	-0.0230 ± 0.0020	27.940 ± 0.543
		15.0	-0.0230 ± 0.0051	24.523 ± 86.796
		20.0	-0.0229 ± 0.0062	19.772 ± 0.682
Grating	38	10.0	-0.0149 ± 0.0075	29.714 ± 3.654
		12.5	-0.0139 ± 0.0500	10.841 ± 17.144
		15.0	-0.0108 ± 0.0015	36.087 ± 2.070
		20.0	-0.0096 ± 0.01918	11.467 ± 8.996

A common feature of all samples is that above 10 K the effect seems to be already destroyed. Therefore, focus will be given to the 10 K curve. This one was already discussed, in which we highlighted the almost linear behaviour with the longitudinal applied magnetic field that was linked with a classical MR contribution. The linear background term will take

into account this classical contribution and dominate the fit, as depicted in the Appendix B.3. Therefore, the WAL contribution, represented by the $\alpha(1)$ and $l_\phi(1)$ parameters, will have a high uncertainty associated. This big uncertainty forbids the comparison of these two coefficients between the film and grating samples, so nothing can be concluded regarding that. The only thing worth highlighting is that, as in the latter cases, the grating samples seem to exhibit noisier data than the film ones, which is represented in bigger uncertainties for the $\alpha(1)$ and $l_\phi(1)$ parameters when compared to the film ones.

Then the same analysis was performed on the 18 nm Bi_2Te_3 with no Ag spot deposited on top, which is correspondent to sample 3 from Table 5.4, to address whether the silver layer has any influence on the $\alpha(0)$, $l_\phi(0)$, $\alpha(1)$ and $l_\phi(1)$ coefficients. Figure 5.8 exhibits the results for that sample concerning the temperatures between 3 K to 7.5 K and Table 5.6 displays the ones for temperatures between 10 K to 20 K.

TABLE 5.6: Results for the coefficients $\alpha(1)$ and $l_\phi(1)$ obtained through the HLN fit, stated in Equation 5.8 for sample 3, with no 15 nm Ag spot deposited on top, in the range of temperatures between 10 K and 20 K

Structure	$t(\text{Bi}_2\text{Te}_3) / \text{nm}$	T / K	$\alpha(1) \pm u(\alpha(1))$	$l_\phi(1) \pm u(l_\phi(1)) / \text{nm}$
Film	18	10.0	-0.0014 ± 0.0002	165.048 ± 81.208
		12.5	-0.0056 ± 0.0009	34.769 ± 2.223
		15.0	-0.0019 ± 0.0015	37.084 ± 12.332
		20.0	-0.0006 ± 0.0107	15.206 ± 63.939

Concerning the WL effect dominated range of temperatures, sample 3 exhibited an $\alpha(0)$ smaller than sample 2, which has the same thickness of Bi_2Te_3 but with the 15 nm Ag spot deposited on top. This means that the silver layer likely strengthen the weak-localization effect. The $l_\phi(0)$ values are also relatively smaller than the ones for sample 2, meaning that the silver layer increases the phase coherent length associated with the weak localization effect. Regarding the 10 K to 20 K range of temperatures, the same can be concluded as the previous data. The $\alpha(1)$ and $l_\phi(1)$ coefficients only display reliable values for the 10 K data. Despite that, the curve for this data also displays an almost linear behaviour, supporting our idea of the dominance of the classical linear MR effect. Therefore, the $\alpha(1)$ coefficient is really small for this curve. The $l_\phi(1)$ value is similar to sample 2, meaning that at this temperature the silver layer does not alter the phase coherent length.

All these results may induce the reader to believe that the samples are not what would be expected of them. This could have an impact regarding the phenomena planned to be

excited. Plasmonics on topological insulators have as basis the existence of the surface states to enhance the propagation of their spin-polarized plasmons [54]. By observing the competition between the WL and WAL effects, being both of them weak, raises questions about whether the excitation of the theoretically reported plasmon-spin waves [9] will be possible. It has been studied in past works from the research group that the annealing of the topological insulator samples improves their topological properties. However, for PMMA and most NIL-compatible substrates, annealing is not an option since it would destroy the structures fabricated. Despite that, the PMMA substrates improve disorder, given their polymeric amorphous nature, enabling us to observe phenomena not so common within this class of materials.

5.3. Main Conclusions

The resistivity dependence on temperature was measured together with the magnetoconductance of both film and grating samples with 18 nm and 38 nm of Bi_2Te_3 , all covered by a 15 nm of Ag circular centered spot of 0.5 cm in diameter. Concerning the resistivity dependence on temperature, the films and grating samples behaved similarly. For small temperatures all samples become highly resistive, increasing abruptly their resistivity like in the freeze out zone of an extrinsic semiconductor. The resistivity dependence on temperature was analyzed with Mott's Variable Range Hopping model, where it was concluded that the transport of the samples is dominated by an ensemble of quantum dots that arise from the amorphous matrix of the material. Regarding the magnetoresistance measurements, a competition between the WL and WAL effects was observed, possibly due to this amorphousness. The WL effect dominated for temperatures in the range of 3 to 7.5 K. For bigger temperatures, the positive correction typical of WAL was observed. However, a classical linear MR behaviour dominated for the 10 K data, and for bigger temperatures the effect was already highly weakened. The existence of structures tailored to the samples altered the $\alpha(0)$, $l_\phi(0)$ coefficients, however, as this data is concerned, it is unclear whether the existence of structures strengths or weakens the weak-localization effect. Concerning the $\alpha(1)$ and $l_\phi(1)$ coefficients, it is not possible to conclude if these are affected by the tailoring of microstructures to the samples.

Moreover, the deposition of a silver layer on top of the Bi_2Te_3 seems to strengthen the $\alpha(0)$ and $l_\phi(0)$ coefficients. Both $\alpha(1)$, and $l_\phi(1)$ coefficients remained unaltered for this condition. Based on these results, these samples may not display robust topological surface states. However, they enable the observation of distinctive features coming from the amorphousness of this complex system.

6. Conclusion and Future Work

This work had as its main goal the fabrication of micro-grating structures of Bi_2Te_3 to excite plasmon modes and, by taking advantage of the theoretically predicted spin-plasmon modes emerging on these materials, explore the quantum effects that typically arise on them, such as the emergence of the WAL effect, and explore possible applications within the spin injections with ferromagnetic materials. The work has not yet been completed, however, we already move forward in understanding the complex physics within this system.

Micro-grating structures were fabricated on polymethyl methacrylate substrates through nano-imprint lithography with two different periodicity values of approximately $p=(0.725 \pm 0.001) \mu\text{m}$ and $p=(1.496 \pm 0.001) \mu\text{m}$. Subsequently numerical simulations were performed on COMSOL multhyphysics 5.6 for grating structures with a periodicity of $0.700 \mu\text{m}$, covered by 30 nm of Bi_2Te_3 and 15 nm of Ag . Three possible plasmon modes were identified at excitation's wavelengths of 450 nm , 571 nm , and 683 nm . Then a parametric sweep was employed for the Ag and Bi_2Te_3 thicknesses and the consequent frequency shift of these peaks was analyzed. For the Ag thickness sweeping, there is a critical thickness value at which the frequency stops shifting and stays fixed, due to the overcoming of the penetration depth of the Ag for the respective incident wavelength. The third peak is likely excited in the interface between the air and the Ag layer, whereas the first and second peaks are possibly excited at the interface between the Ag and Bi_2Te_3 layers. Moreover, the observed simulation peaks do not get nullified when changing the grating geometry from the ideal square-shaped form. In addition, we concluded that the first peaks is geometry-independent.

Then, the samples, which were deposited by DC-magneto sputtering deposition with the same materials as the simulation ones, were measured through UV-VIS-NIR spectroscopy in the same range of thicknesses for the Bi_2Te_3 layer. There are some similarities between both experimental and simulation data, however, these are not sufficient to overcome the poor overlap between them. The differences between the numerical simulations' model and the experimental conditions, such as the differences between polarized and non-polarized light, single crystal and amorphous structures, perfect and irregular interfaces, among many others, all contribute to the mismatch between both data.

Nonetheless, possible plasmon peaks were identified, one of them localized for excitation's wavelength between 662 nm and 751 nm and the other one between 817 nm and 879 nm.

Afterwards, the transport properties of the samples were characterized, namely the resistivity dependence with temperature and the magnetoresistance. The samples are highly resistive at small temperatures. Through the analysis of the resistivity dependence on temperature, by using Mott's Variable Range Hopping model, the transport of the samples was determined to be dominated by the $d = 0$ dimensionality of the system. Additionally the magnetoconductance of the samples displayed a competition between the WL and WAL effects. This feature is likely due to the amorphousness of the samples. The WL effect disappears for temperatures bigger than 7.5 K. At 10 K, a classical linear MR behaviour emerges, and for bigger temperatures, the quantum correction typical of the WAL effect got highly suppressed. Furthermore, the existence of structures tailored to the samples altered the magnetoconductance of the samples, however, we cannot conclude whether it strengthens or weakens the localization mechanisms. All these results suggest that the samples may not display robust topologically protected surface states, which raises a question about the potentiality of observing the phenomena intended to be excited on these devices.

As future works, both numerical simulations and UV-VIS-NIR spectroscopy will be studied on grating samples with a periodicity of approximately 1500 nm, as samples were already fabricated with this feature by using a CD-PDMS stamp, as previously discussed in Chapter 2. Additionally, the transport properties for these samples with different thicknesses of Bi_2Te_3 will be measured. Following this, a fibre will be installed on the PPMS chamber connected with lasers with wavelengths that coincide with the excitation's wavelength identified in the UV-VIS-NIR spectra of the samples in Chapter 4. The setup will follow the application note made by Quantum Design for photoconductivity measurements [126]. With the setup fully operational, the magnetoresistance will be measured while directing the laser light to the 0.5 cm diameter circular Ag spot deposited on top of the samples, to observe whether the excitation of the possible plasmon modes that were identified alters the observation of the phenomena already discussed in Chapter 5. At a later stage of these future works, all these characterizations will be again performed, but including a Gd layer into the device. Finally, to study the spin dynamics of the samples, the FMR will be measured while exciting plasmon modes on them with laser light at the excitation's wavelength of the identified plasmon peaks.

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A. Gd deposition

A.1. XRD analysis

Since it is also planned to integrate a Gd layer, an XRD analysis was also performed on this material. We first deposited a thin film of Gd onto glass. However, when taking out the samples from the sputtering chamber, we noticed these exhibit a white colour instead of the typical silvery metal colour, which could be associated with the formation of gadolinium oxide. When performing XRD measurements, the results, which are shown in Figure A.1, show that the sample suffered from oxidation and exhibits a Gd_2O_3 phase.

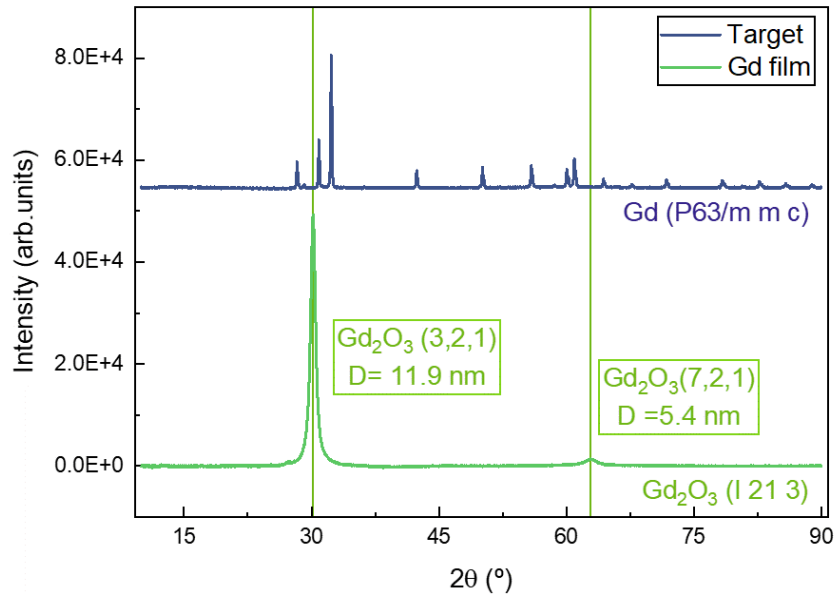


FIGURE A.1: XRD spectra for the Gd target used in the DC sputtering deposition and for a thin film of Gd deposited onto glass. The target was identified as Gd, but the thin film was identified as an oxide.

To tackle this problem, a Ta layer was deposited together with the Gd thin films. We used this layer as a buffer, capping, and both buffer and capping in order to address possible ways to avoid this oxidation and also to study from where this oxidation came from. All these samples were measured through XRD, and the Gd_2O_3 phase was identified to only be present in the sample where Ta was used as a capping layer, as it is shown in Figure A.2. This could indicate that the oxidation doesn't come from the contact of the Gd with the surrounding atmosphere. Instead, the oxidation is coming from the substrate. A possible reason that may justify this to happen is that, since the ions reach the substrate with high energy, they could lead to a reaction where the oxygen from the SiO_2 could attach with the Gd. Moreover, the oxide crystallizes in the $I 21 3$ (199) space group, which

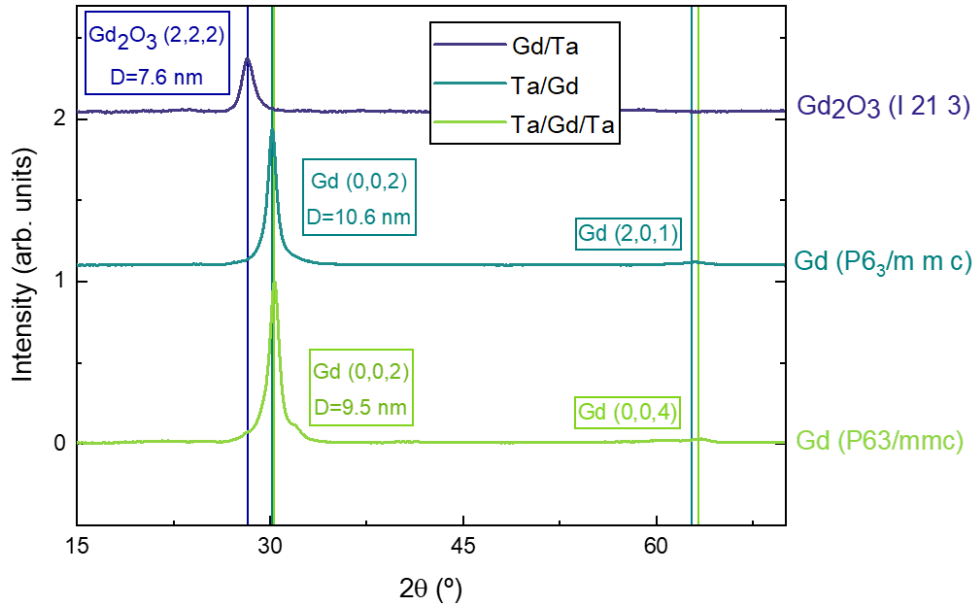


FIGURE A.2: XRD spectra for a thin film heterostructure of 57 nm of Gd and 15 nm of Ta used as only a buffer layer (Ta/Gd) , a capping layer (Gd/Ta) , and both within the same structure (Ta/Gd/Ta). The oxidation of Gd only comes for the structure where the Ta was used as capping layer.

exhibits cubic symmetry, and when incorporating Ta as buffer layer, the Gd crystallizes in the $P6_3/mmc$ (194) space group, which exhibits hexagonal symmetry. By adding the Ta buffer layer, a bigger crystallite size is obtained. However, despite this increment, it is still substantially small. Therefore, the sample is polycrystalline with small grain size. In addition, the sample where Ta was used both as a capping and buffer layer exhibits a small contribution that appears within the (0,0,2) peak. This contribution was not identified with the Rigaku software since it has a relatively low intensity when compared with the (0,0,2) reflection, and because of that, is masked out. Despite that, it is expectable that this peak is related to the Ta layer.

A.2. XRR analysis

The same analysis was performed with the Gd samples. However, since a Ta layer was included, the analysis is slightly different. The XRR measurements were performed on the sample which uses Ta as both a capping and buffer layer. As this technique is surface sensitive, the Ta capping layer should have a great contribution to the data. As can be seen in Figure A.3 (A) and (B).

Therefore, the main fringes are attributed to the Ta layer, whereas the small oscillations within the main fringes are linked with the Gd one. Since these oscillations display small amplitude, the program could not detect and fit them. Because of that, this data was analyzed by using on equation 2.3, which has already been done with good accuracy

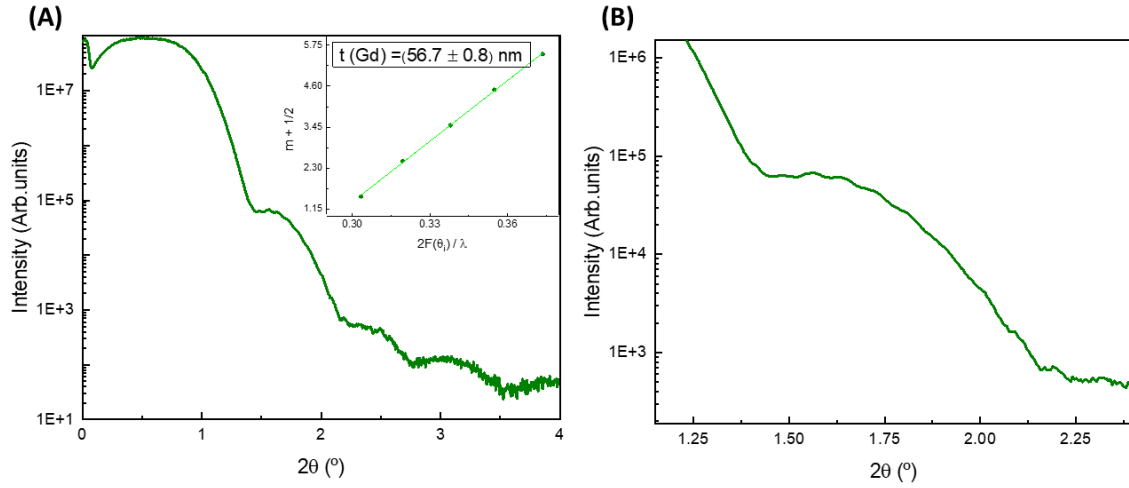


FIGURE A.3: XRR spectrum for a 10 minute Gd deposition and a 5 minute Ta deposition, together with the linear fit according to equation 2.3 (A). A zoom in is shown (B) to highlight the oscillations contribution from the Gd layer.

in other works [20]. This method has as a limitation getting only the thickness value, since there is not the sensibility to extract the attenuation of the signal. Despite that, there is a significant attenuation and, therefore, the surface roughness is also significant. Concerning the thickness value, it was determined a thickness value of 57 nm for a 10 minute deposition. Since Ta was used as a capping and buffer layer, a thin film of Ta was also measured through XRR, which was equivalent to a 10 min deposition. The XRR spectrum is presented in Figure A.4.

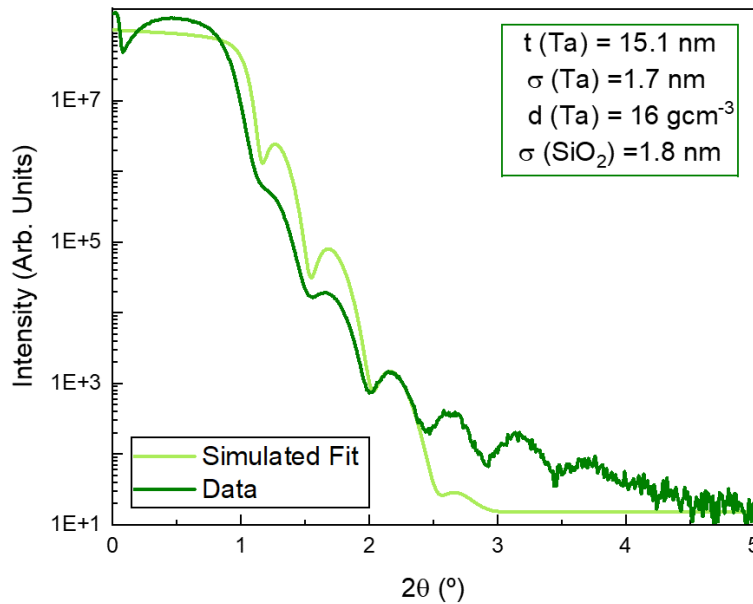


FIGURE A.4: XRR spectrum for a 10 minute Ta, which corresponds to a thickness value of 15.1 nm.

Similarly as before, a prior focus was given onto the Ta thickness, since it is the most

important parameter for this step of fabrication. As a result, the surface roughness and density are not entirely accurate. Instead, they are just a rough approximation of the real values for these parameters, but they are used here as an auxiliary fitting parameter so that it would be possible to obtain a thickness value closer to the real value.

A.3. Magnetic Characterization

A magnetization over temperature measurement was employed to the thin films of Gd in a MPMS3-SQUID magnetometer, so that we could probe the quality of our Gd samples. This is a crucial step in order to understand further results of the magnetoplasmonic device. Focus was given to the samples associated with the identified Gd phase on the XRD spectra presented on Figure A.2, the sample where the Ta layer is used as both capping and buffer layer and the one where the Ta layer is only used as buffer layer.

Experimental details: The equipment used was a MPMS3-SQUID magnetometer from Quantum Design. The samples were cut with 0.63 cm^2 for the Ta / Gd sample and 0.70 cm^2 for the Ta / Gd / Ta one. Both of them were measured in the in-plane configuration. An external magnetic field of 100 Oe was applied when measuring the samples.

The data for those two samples is shown in figure A.5.

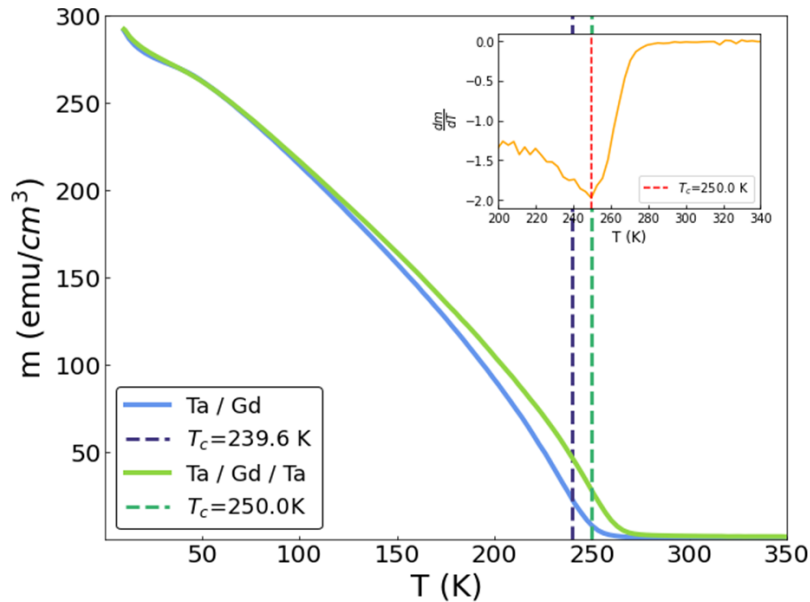


FIGURE A.5: Magnetization over temperature data of the Ta(15nm) / Gd(57nm) / Ta(15nm) and Ta(15nm) / Gd(57nm) samples. The critical temperature was determined for both samples, respectively $T_c = 250.2 \text{ K}$ (blue line) and $T_c = 262.0 \text{ K}$ (green line). To determine the Curie temperature, we performed the derivative of the data, as shown in the inset.

The critical temperature for the paramagnetic to ferromagnetic phase transition was determined by applying a second derivative to the experimental data. For the Ta / Gd / Ta sample, the Curie temperature was 250.0 K. Assuming the bulk Curie's temperature for this material to be 293 K [127], there is a relative error of 15% for this sample. However, it has already been reported [128] that the addition of a capping layer could alter the Curie temperature, and the same applies to the addition of a buffer layer. In addition, Scheunert *et al.* [129] also reported that smaller curie temperatures in Gd thin films are linked to small grain sizes, since this parameter got closer to the theoretical value after *in-situ* thermal annealing. Since the crystallite size was determined to be 9.5 nm, this potentially could be a contribution for the sample's lower Curie temperature. For the Ta / Gd layer, the Curie temperature was determined to be 239.6 K, which when compared with the bulk theoretical value, represents a relative error of 18%. The fact that the Curie temperature for this sample is smaller than the previous one is expected, since by not having a capping layer, it was expectable some oxidation, even though of small degree. There is also a small kink at smaller temperatures at 20 K for both samples, that may come from a substrate contribution.

B. Complementary Experimental Data

B.1. Chapter 3

All fit done for Bi_2Te_3 , Ag and grating angle sweep will be displayed in this appendix section, alongside the respective fit parameters.

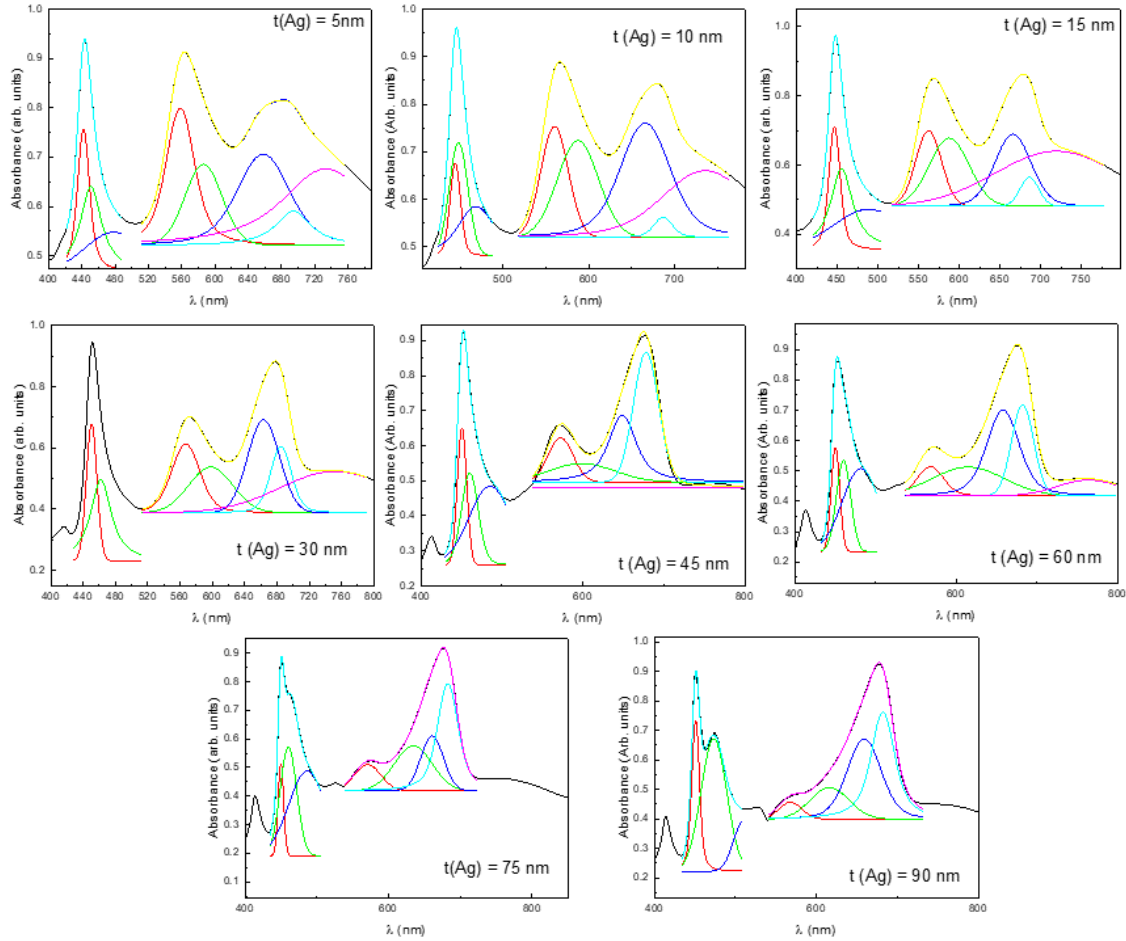


FIGURE B.1: Peak Identification for the Ag thickness sweep. The Bi_2Te_3 thickness was fixed at 30 nm.

t (nm)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
5	441.854	0.221	6.802	0.724	10.922	0.734	10.275	0.953	17.416	0.365
10	444.389	0.092	3.317	4.055	12.746	3.748	3.286	12.996	14.594	3.957
15	446.467	0.499	7.526	3.946	12.161	0.706	7.373	3.592	16.577	1.832
30	450.458	0.235	7.657	1.335	14.950	0.867	1.104	2.256	15.549	0.639
45	450.617	0.096	5.706	1.554	13.713	0.721	0.000	--	13.713	0.721
60	450.528	0.278	4.646	2.188	12.673	2.855	0.000	6.830	12.673	1.281
75	450.010	0.051	3.126	2.601	9.061	2.648	0.066	7.697	9.096	1.519
90	450.804	0.047	7.311	1.526	8.784	1.207	4.532	2.580	11.457	0.543

FIGURE B.2: Fit parameters for contribution 1 of the first peak in the Ag thickness sweep, identified in figure 3.5 (B).

t (nm)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
5	450.756	1.933	8.394	3.485	0.000	97870.135	30.851	3.765	30.852	3.679
10	448.244	2.625	6.530	4.483	25.745	3.715	0.000	0.029	25.745	3.715
15	455.274	5.251	12.134	5.910	0.001	98499.661	32.816	9.745	32.816	4.878
30	462.018	1.922	13.060	5.059	0.000	165126.493	31.056	8.422	31.056	5.672
45	460.311	3.218	7.378	13.613	21.913	16.017	4.455	40.825	24.393	10.419
60	460.525	2.375	6.697	4.046	20.874	5.075	0.000	0.292	20.874	5.075
75	460.445	2.770	11.074	9.852	27.314	7.368	0.000	0.160	27.314	7.367
90	473.259	0.512	17.852	2.559	37.277	4.357	0.000	0.633	37.277	4.357

FIGURE B.3: Fit parameters for contribution 2 of the first peak in the Ag thickness sweep, identified in figure 3.6 (B).

t (nm)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
5	558.165	0.503	15.000	116.748	27.717	44.872	21.441	167.395	40.921	73.770
10	560.058	0.413	9.350	3.578	37.558	1.899	0.001	6.054	37.559	2.380
15	564.210	1.449	12.099	31.197	39.510	38.402	3.569	114.965	41.453	24.818
30	567.084	0.769	10.991	8.950	35.951	1.309	10.312	11.601	41.783	6.304
45	572.472	1.970	4.940	5.898	36.519	10.671	0.000	--	36.519	10.671
60	568.219	2.017	4.368	19.825	34.198	44.302	8.440	144.263	38.935	41.005
75	570.884	4.033	4.267	5.988	43.357	10.926	0.000	0.088	43.357	10.926
90	572.000	2.404	5.255	3.632	40.000	27.678	22.008	47.571	53.056	8.554

FIGURE B.4: Fit parameters for contribution 1 of the second peak in the Ag thickness sweep, identified in figure 3.7 (B).

t (nm)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
5	693.964	2.336	5.692	47.132	0.003	459084.690	51.620	179.922	51.620	133.537
10	686.566	0.250	1.391	3.351	19.613	15.919	10.623	46.987	25.905	15.146
15	685.908	0.254	2.499	2.030	26.138	3.955	1.885	14.979	27.161	4.626
30	685.004	0.379	7.823	4.541	26.499	1.310	7.654	7.244	30.829	3.480
45	682.643	1.422	13.881	46.366	26.686	22.420	12.973	74.610	34.296	25.853
60	682.995	1.010	13.549	3.425	27.098	3.464	11.489	4.835	33.763	0.721
75	682.646	0.901	17.177	12.918	23.090	8.367	18.632	21.669	34.625	7.380
90	682.100	0.849	17.677	37.564	19.698	14.943	23.464	41.355	35.066	18.867

FIGURE B.5: Fit parameters for contribution 4 of the third peak in the Ag thickness sweep, identified in figure 3.8 (B).

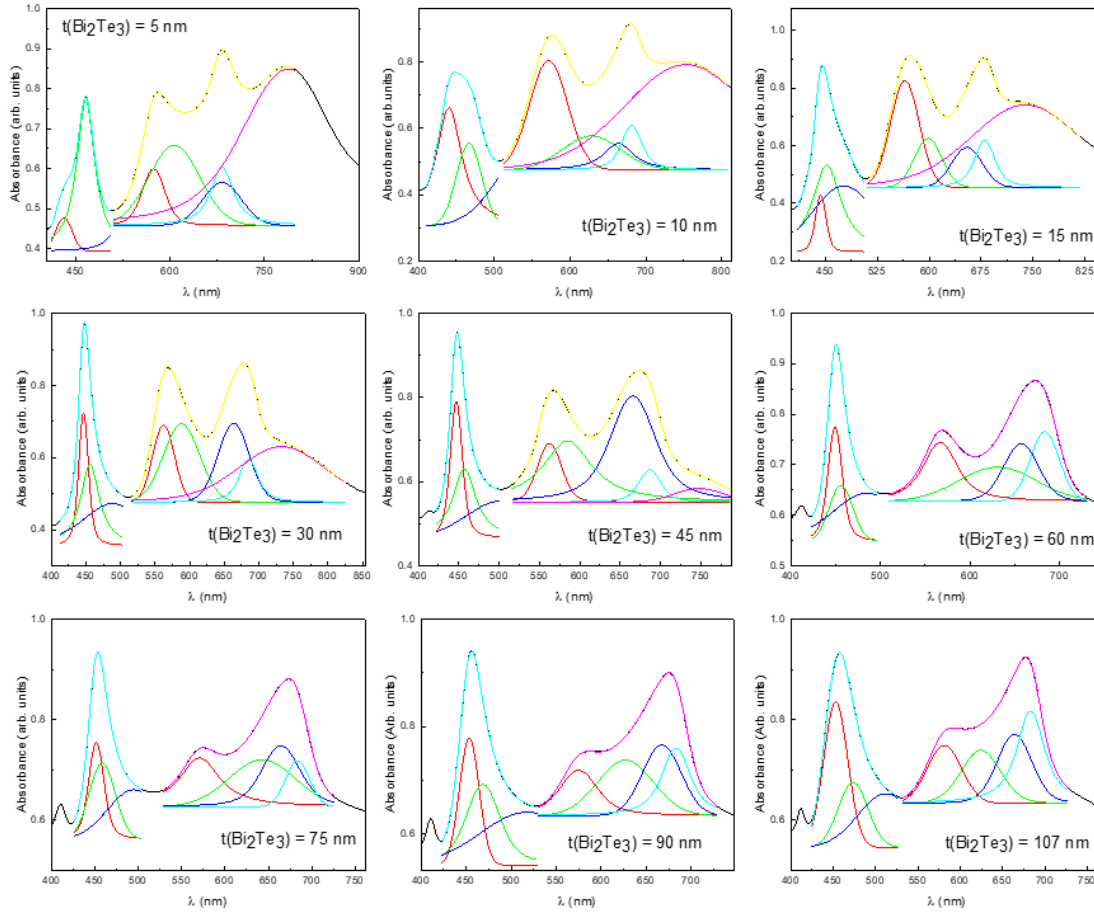


FIGURE B.6: Peak Identification for the Bi_2Te_3 thickness sweep. The Ag thickness was fixed at 15 nm.

t (nm)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
5	431.181	0.156	2.752	0.162	30.558	0.703	0.000	0.019	30.558	0.703
10	440.307	0.247	23.381	15.210	0.001	23704.138	41.702	6.653	41.703	4.782
15	443.281	0.594	4.266	9.041	15.678	7.310	4.817	26.036	18.412	8.450
30	446.522	0.292	7.840	2.121	12.497	0.477	7.203	2.034	16.790	0.972
45	446.958	0.263	7.297	1.457	13.098	0.506	7.806	1.419	17.766	0.763
60	449.367	0.062	5.408	4.787	14.205	3.381	7.935	11.187	18.919	4.024
75	451.709	0.047	5.069	2.479	15.864	2.239	8.419	7.130	20.842	2.439
90	453.643	0.337	6.486	2.381	25.708	1.329	0.000	0.288	25.708	1.329
107	453.086	3.513	10.439	3.497	33.488	11.663	0.356	27.194	33.679	3.234

FIGURE B.7: Fit parameters for contribution 1 of the first peak in the Bi_2Te_3 thickness sweep, identified in figure 3.11 (B).

t (nm)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
5	431.181	0.156	2.752	0.162	30.558	0.703	0.000	0.019	30.558	0.703
10	440.307	0.247	23.381	15.210	0.001	23704.138	41.702	6.653	41.703	4.782
15	443.281	0.594	4.266	9.041	15.678	7.310	4.817	26.036	18.412	8.450
30	446.522	0.292	7.840	2.121	12.497	0.477	7.203	2.034	16.790	0.972
45	446.958	0.263	7.297	1.457	13.098	0.506	7.806	1.419	17.766	0.763
60	449.367	0.062	5.408	4.787	14.205	3.381	7.935	11.187	18.919	4.024
75	451.709	0.047	5.069	2.479	15.864	2.239	8.419	7.130	20.842	2.439
90	453.643	0.337	6.486	2.381	25.708	1.329	0.000	0.288	25.708	1.329
107	453.086	3.513	10.439	3.497	33.488	11.663	0.356	27.194	33.679	3.234

FIGURE B.8: Fit parameters for contribution 1 of the second peak in the Bi_2Te_3 thickness sweep, identified in figure 3.12 (B).

t (nm)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
5	682.561	0.193	9.469	11.190	14.691	16.654	37.989	29.444	43.296	15.727
10	678.752	0.337	5.546	7.483	24.267	11.234	15.581	32.758	33.657	11.245
15	680.209	0.563	7.841	14.409	21.779	11.777	21.491	36.289	35.455	16.069
30	685.448	0.247	5.481	3.205	17.754	4.545	22.837	11.242	32.901	5.165
45	686.816	0.404	3.194	3.984	23.462	6.899	15.005	23.955	32.501	9.503
60	683.131	2.007	5.643	1.393	38.318	1.837	0.000	0.000	38.318	1.837
75	683.572	0.645	3.169	1.311	32.645	1.801	0.000	0.000	32.645	1.801
90	682.962	0.153	6.944	3.234	17.630	4.463	29.354	10.333	37.996	5.005
107	683.182	0.102	10.730	7.512	15.316	5.721	33.038	13.334	39.365	7.547

FIGURE B.9: Fit parameters for contribution 4 of the third peak in the Bi_2Te_3 thickness sweep, identified in figure 3.13 (B).

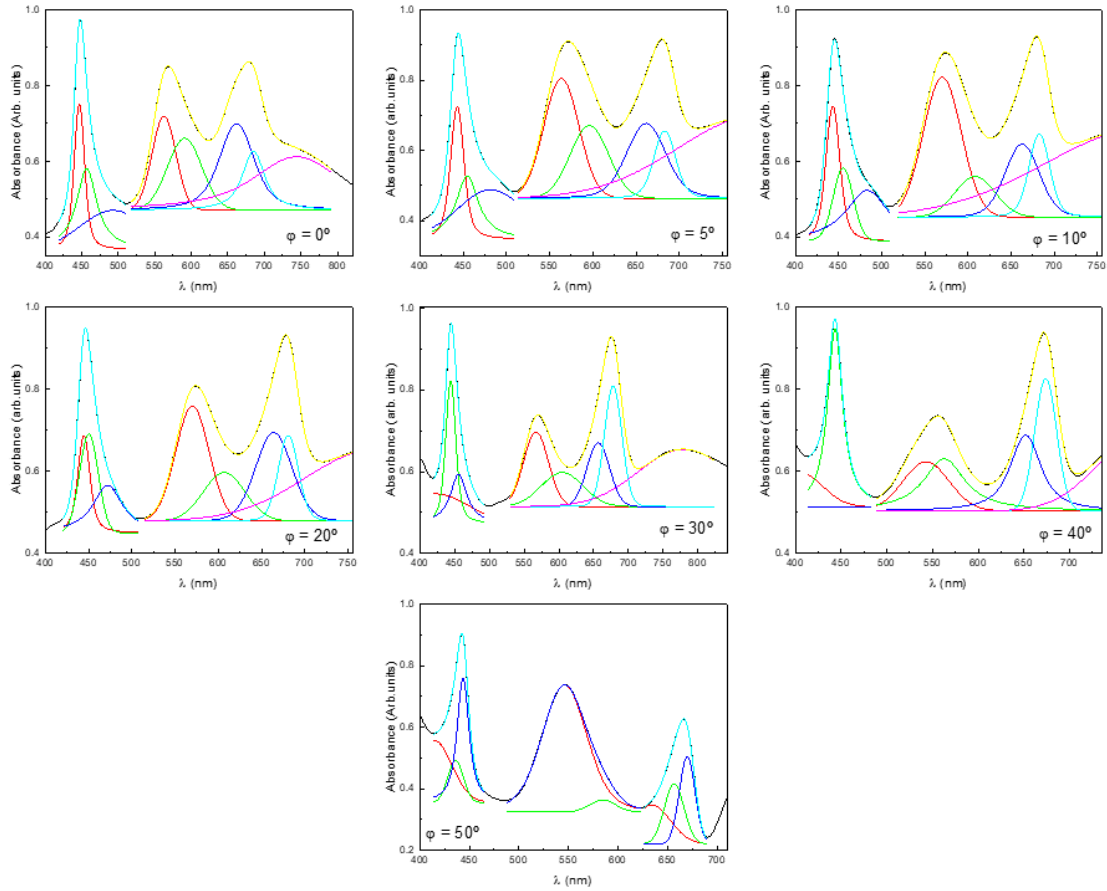


FIGURE B.10: Peak Identification for the ϕ angle sweep. The Bi_2Te_3 thickness was fixed at 30 nm and the Ag one at 15 nm.

ϕ (°)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
0	446.639	0.272	8.394	1.878	12.452	0.512	7.746	1.604	17.104	0.842
5	443.256	0.264	8.888	1.510	13.301	0.986	8.443	2.293	18.383	0.836
10	443.431	0.376	8.840	7.066	14.235	4.652	8.634	11.664	19.407	3.775
20	444.574	0.067	5.184	1.175	11.333	0.288	8.670	1.432	16.665	0.833
30	444.391	0.138	7.799	0.611	12.160	0.968	8.396	1.750	17.261	0.368
40	443.435	0.011	13.362	0.481	3.559	0.787	19.140	0.560	19.825	0.258
50	443.604	0.054	8.474	1.949	6.183	0.823	11.556	1.321	14.372	0.749

FIGURE B.11: Fit parameters for contribution 2 of the first peak in the ϕ angle sweep, identified in figure 3.15 (B).

$\phi(^{\circ})$	λ_c (nm)	$u(\lambda_c)$ (nm)	A	$u(A)$	wG	$u(wG)$	wL	$u(wL)$	FWHM (nm)	$u(FWHM)$ (nm)
0	562.491	0.849	10.123	3.922	38.406	2.021	0.000	0.014	38.406	2.021
5	563.614	2.720	17.251	7.048	47.510	3.147	0.000	0.019	47.510	3.147
10	570.240	2.802	20.500	4.068	52.149	1.972	0.000	0.078	52.149	1.972
20	569.968	1.283	13.627	5.235	45.605	2.288	0.000	0.019	45.605	2.288
30	566.993	0.936	8.171	5.231	41.904	4.591	0.000	0.058	41.904	4.591
40	562.877	0.832	10.903	4.478	0.023	7048.156	54.467	9.289	54.467	5.503
50	546.008	0.134	26.058	0.680	45.770	1.322	13.574	2.290	53.461	0.272

FIGURE B.12: Fit parameters for contribution 1 of the second peak in the ϕ angle sweep, identified in figure 3.16 (B).

$\phi(^{\circ})$	λ_c (nm)	$u(\lambda_c)$ (nm)	A	$u(A)$	wG	$u(wG)$	wL	$u(wL)$	FWHM (nm)	$u(FWHM)$ (nm)
0	684.982	0.401	8.321	4.658	14.743	8.467	30.118	14.630	36.444	6.631
5	682.990	0.460	6.672	10.924	25.670	7.413	7.106	29.419	29.681	10.355
10	682.744	0.366	7.405	6.554	24.514	3.704	7.233	14.663	28.611	5.331
20	680.829	0.180	6.508	7.371	25.003	6.004	4.424	21.967	27.452	6.644
30	677.888	0.509	9.566	1.549	28.615	1.097	1.804	2.383	29.592	0.713
40	673.793	0.631	9.707	0.886	28.249	0.831	0.000	1360270000.000	28.249	0.831
50	669.874	1.558	5.645	5.458	18.744	2.695	0.000	0.010	18.744	2.695

FIGURE B.13: Fit parameters for contribution 4 of the third peak in the ϕ angle sweep, identified in figure 3.16 (B).

B.2. Chapter 4

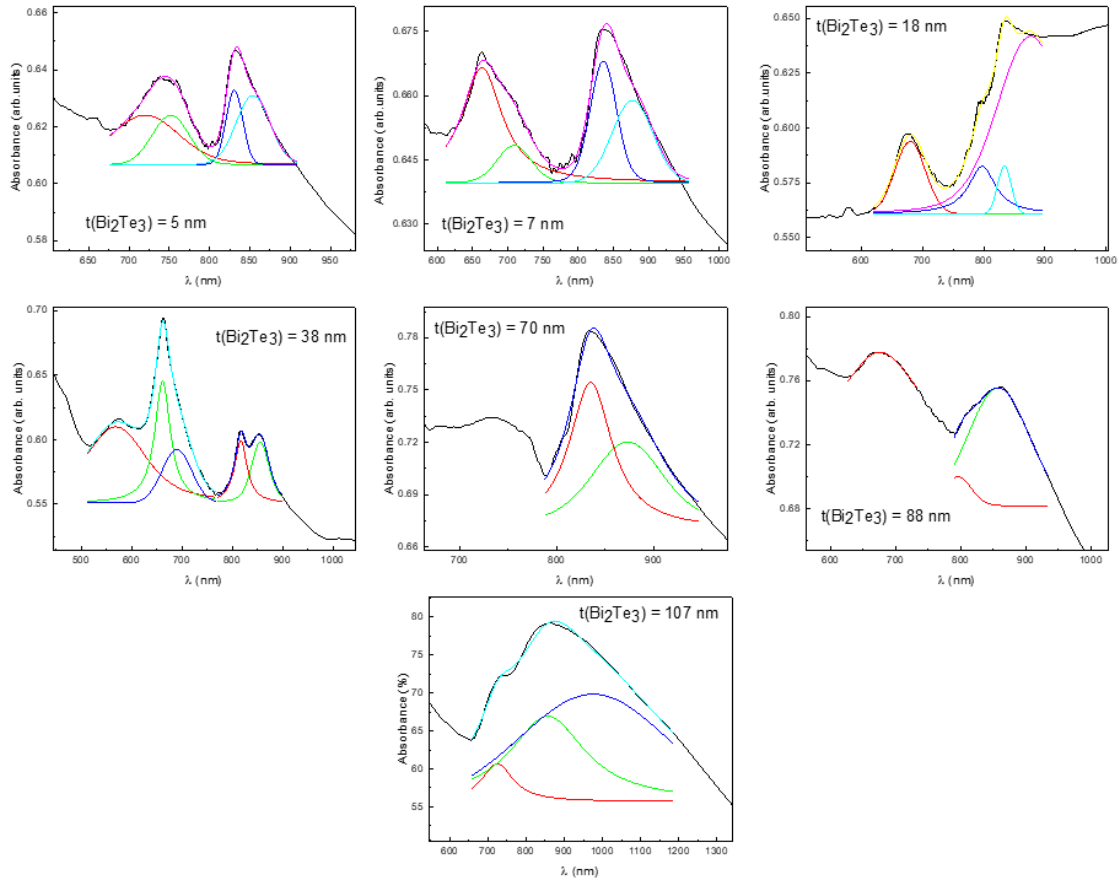


FIGURE B.14: Peak Identification for the Bi_2Te_3 thickness sweep for the UV-VIS-NIR spectra. The Ag thickness was fixed at at 15 nm.

t (nm)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
5.0	751.676	3.147	0.986	0.821	53.659	7.487	0.000	0.854	53.659	7.535
7.0	663.041	2.075	2.927	0.256	0.002	131288.205	69.340	9.138	69.340	5.358
38.3	684.803	0.170	532.038	2.906	67.677	0.419	0.000	80045000.000	67.677	0.419
88.0	676.084	0.198	4.407	0.733	105.055	7.167	0.000	0.000	105.055	7.167
107.2	724.014	1.008	100.000	9.367	40.194	2.973	0.000	114878000.000	40.194	2.973

FIGURE B.15: Fit parameters for contribution 2 of the first peak in the Bi_2Te_3 thickness sweep for the UV-VIS-NIR spectra, identified in figure 4.2 (A).

t (nm)	λ_c (nm)	$u(\lambda_c)$ (nm)	A	u(A)	wG	u(wG)	wL	u(wL)	FWHM (nm)	u(FWHM) (nm)
5.0	835.445	0.855	1.426	0.188	41.133	4.407	6.217	7.278	44.558	1.461
7.0	830.736	0.362	0.681	0.467	24.548	5.062	0.000	15.255	24.548	5.062
38.3	817.189	0.167	2.301	0.054	0.000	18716.547	30.117	0.673	30.117	0.673
70.0	834.893	0.791	7.077	1.307	0.003	31206.261	53.452	5.311	53.452	2.873
88.0	857.741	0.571	8.534	0.414	109.245	2.815	0.000	0.000	109.245	2.815
107.0	878.805	3.627	5080.887	428.865	283.709	9.610	0.000	534758000.000	283.709	9.610

FIGURE B.16: Fit parameters for contribution 3 of the second peak in the Bi_2Te_3 thickness sweep for the UV-VIS-NIR spectra, identified in figure 4.2 (A).

B.3. Chapter 5

B.4. Mott's Variable Range Hopping model fit parameters

All fit parameters used for the Mott's Variable Range Hopping model fitting of the resistivity dependence on temperatures will be displayed in this appendix section.

Structure	t(Bi ₂ Te ₃) / nm	t(Ag) / nm	ln(ρ_0)	u[ln(ρ_0)]	T ₀	u(T ₀)	γ	u(γ)	R-Square
Film	18	15	1.55E-06	2.09E-08	3.69E-04	5.88E-07	0.940	0.001	0.997
Grating	18	15	1.32E-06	2.12E-08	3.51E-04	7.04E-07	0.918	0.001	0.995
Film	18	0	1.47E-06	2.16E-08	3.61E-04	6.90E-07	0.932	0.001	0.996
Film	38	15	3.06E-06	4.54E-08	7.61E-04	1.45E-06	0.930	0.001	0.996
Grating	38	15	2.58E-06	4.59E-08	7.38E-04	1.00E-06	0.913	0.001	0.997

FIGURE B.17: Fitting parameters for the resistivity dependence on temperature data based on the Mott's Variable Range Hopping model.

B.5. HLN model fit data

All fitted data and respective fit parameters used for the HLN model fitting of our samples will be displayed in this appendix section.

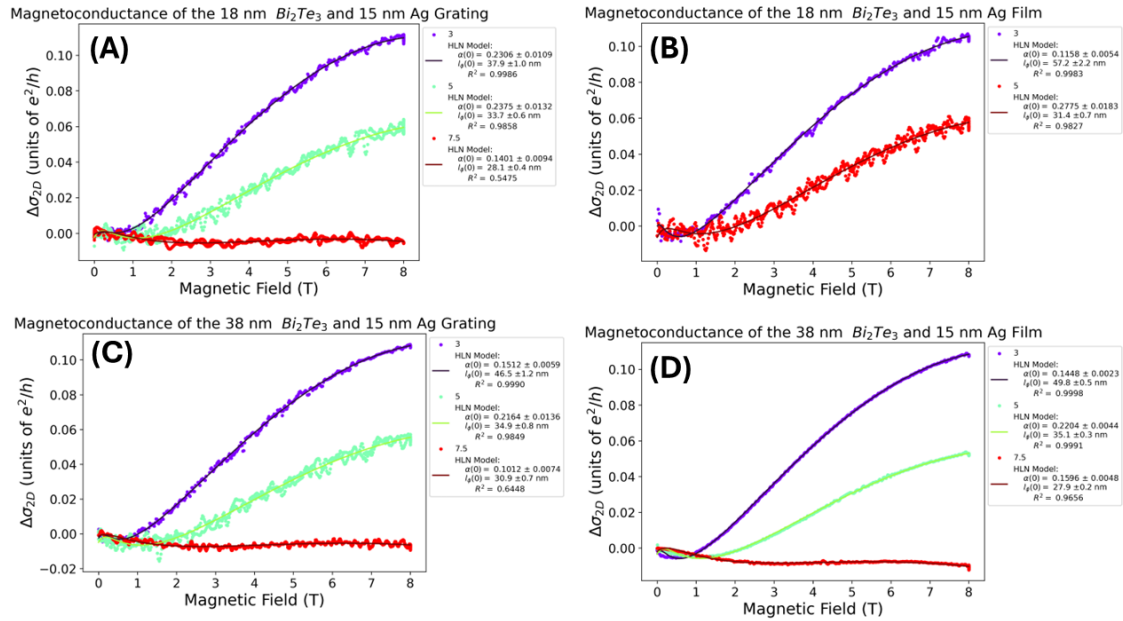


FIGURE B.18: Fitting of the magnetoconductance of samples 1 (A), 2 (B), 4 (C), and 5 (D) as a function of the in-plane longitudinal applied magnetic field at temperatures between 3 and 7.5 K, related to the WL effect dominated data.

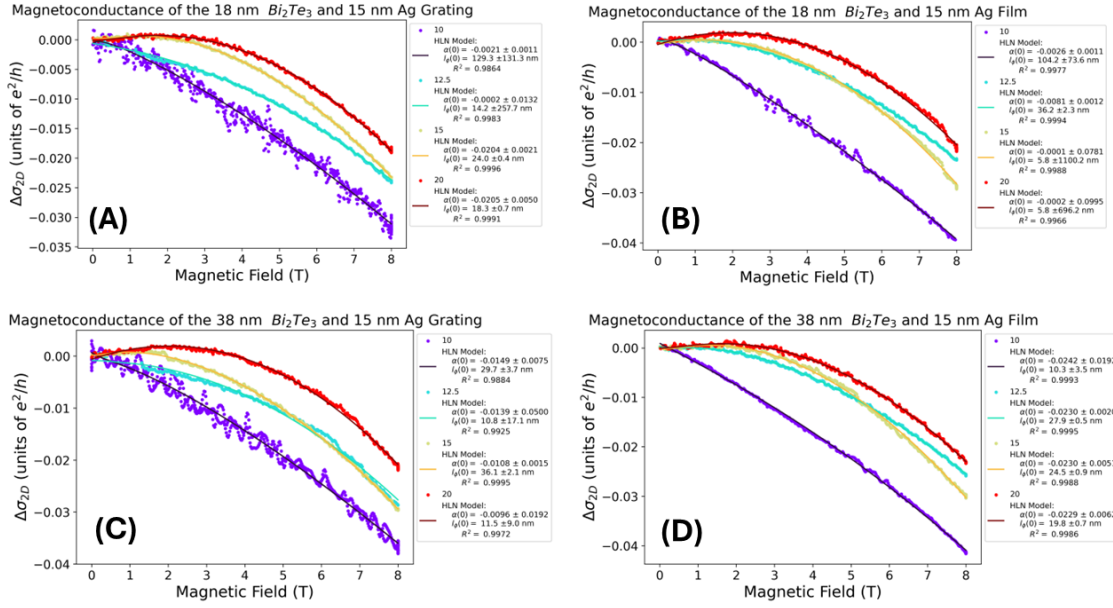


FIGURE B.19: Fitting of the magnetoconductance of samples 1 (A), 2 (B), 4 (C), and 5 (D) as a function of the in-plane applied longitudinal magnetic field, at temperatures between 10 and 20 K.

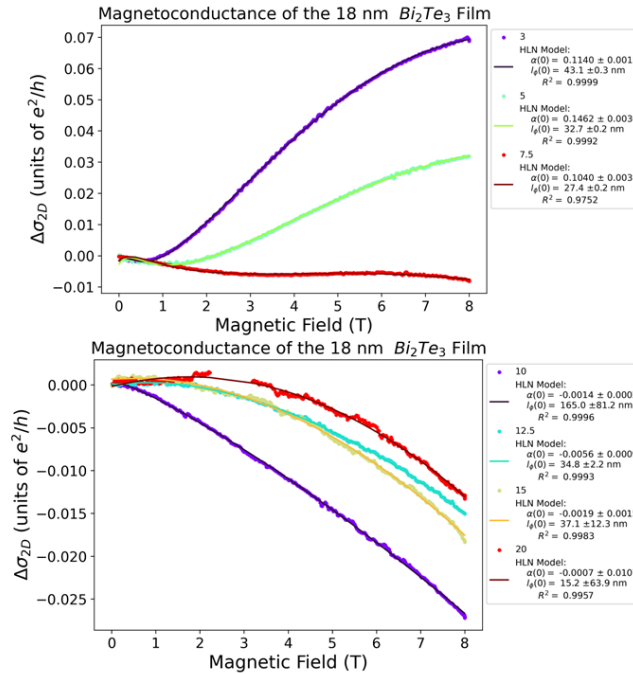


FIGURE B.20: Fitting of the magnetoconductance of sample 3 as a function of the in-plane longitudinal applied magnetic field at temperatures between 2 and 7.5 K (top) and 10-20 K (bottom), related with the WL and WAL fitting model, respectively.

	$\alpha(0)$	$l_p(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 3 K	0.1158 ± 0.0054	57.25 ± 2.21	-0.89 ± 0.02	17.81 ± 0.33	-0.08 ± 0.32	0.9983
Temperature: 5 K	0.2775 ± 0.0183	31.43 ± 0.65	-0.76 ± 0.04	17.77 ± 0.81	-2.43 ± 0.32	0.9827

FIGURE B.21: Fitting parameters for the 18 nm Bi_2Te_3 and 15 nm film for temperatures in the range of 3 K to 5 K.

	$\alpha(0)$	$l_p(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 10 K	-0.0026 ± 0.0011	104.22 ± 73.59	-0.04 ± 0.01	-1.75 ± 0.09	0.13 ± 0.21	0.9977
Temperature: 12 K	-0.0081 ± 0.0012	36.21 ± 2.25	-0.13 ± 0.00	-0.39 ± 0.06	0.08 ± 0.03	0.9994
Temperature: 15 K	-0.0001 ± 0.0781	5.82 ± 1100.16	-0.24 ± 0.01	0.55 ± 0.21	-0.15 ± 0.06	0.9988
Temperature: 20 K	-0.0002 ± 0.0995	5.84 ± 696.25	-0.24 ± 0.02	0.93 ± 0.27	-0.19 ± 0.07	0.9966

FIGURE B.22: Fitting parameters for the 18 nm Bi_2Te_3 and 15 nm film for temperatures in the range of 10 K to 20 K.

	$\alpha(0)$	$l_p(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 3 K	0.2331 ± 0.0110	37.77 ± 0.94	-1.18 ± 0.03	23.43 ± 0.51	-1.80 ± 0.37	0.9986
Temperature: 5 K	0.2375 ± 0.0132	33.71 ± 0.60	-0.72 ± 0.03	16.56 ± 0.62	-0.90 ± 0.21	0.9858
Temperature: 7 K	0.1401 ± 0.0094	28.12 ± 0.40	-0.30 ± 0.02	6.13 ± 0.39	-0.27 ± 0.11	0.5475

FIGURE B.23: Fitting parameters for the 18 nm Bi_2Te_3 and 15 nm grating sample for temperatures in the range of 3 K to 7.5 K.

	$\alpha(0)$	$l_\phi(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 10 K	-0.0021 ± 0.0011	129.34 ± 131.25	-0.05 ± 0.01	-1.23 ± 0.10	-0.22 ± 0.26	0.9864
Temperature: 12 K	-0.0002 ± 0.0132	14.15 ± 257.73	-0.10 ± 0.01	-0.32 ± 0.31	-0.25 ± 0.05	0.9983
Temperature: 15 K	-0.0204 ± 0.0021	24.01 ± 0.35	-0.15 ± 0.00	-0.39 ± 0.08	0.12 ± 0.02	0.9996
Temperature: 20 K	-0.0205 ± 0.0050	18.28 ± 0.69	-0.17 ± 0.01	0.13 ± 0.15	-0.08 ± 0.03	0.9991

FIGURE B.24: Fitting parameters for the 18 nm Bi_2Te_3 and 15 nm grating sample for temperatures in the range of 10 K to 20 K.

	$\alpha(0)$	$l_\phi(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 3 K	0.1448 ± 0.0023	49.80 ± 0.49	-0.98 ± 0.01	19.64 ± 0.14	-1.21 ± 0.08	0.9998
Temperature: 5 K	0.2204 ± 0.0044	35.14 ± 0.27	-0.70 ± 0.01	15.88 ± 0.21	-1.31 ± 0.09	0.9991
Temperature: 7 K	0.1596 ± 0.0048	27.95 ± 0.20	-0.34 ± 0.01	6.76 ± 0.19	-0.71 ± 0.07	0.9656

FIGURE B.25: Fitting parameters for the 38 nm Bi_2Te_3 and 15 nm film for temperatures in the range of 3 K to 7.5 K.

	$\alpha(0)$	$l_\phi(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 10 K	-0.0242 ± 0.0192	10.30 ± 3.50	-0.08 ± 0.01	-1.56 ± 0.30	0.37 ± 0.06	0.9993
Temperature: 12 K	-0.0230 ± 0.0020	27.94 ± 0.54	-0.10 ± 0.00	-1.07 ± 0.08	0.13 ± 0.03	0.9995
Temperature: 15 K	-0.0230 ± 0.0051	24.52 ± 0.87	-0.21 ± 0.01	-0.33 ± 0.19	-0.04 ± 0.05	0.9988
Temperature: 20 K	-0.0229 ± 0.0062	19.77 ± 0.68	-0.20 ± 0.01	0.03 ± 0.19	-0.02 ± 0.04	0.9986

FIGURE B.26: Fitting parameters for the 38 nm Bi_2Te_3 and 15 nm film for temperatures in the range of 10 K to 20 K.

	$\alpha(0)$	$l_p(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 3 K	0.1512 ± 0.0059	46.46 ± 1.15	-0.97 ± 0.02	19.49 ± 0.33	-1.03 ± 0.24	0.9990
Temperature: 5 K	0.2164 ± 0.0136	34.90 ± 0.83	-0.67 ± 0.03	15.61 ± 0.64	-1.90 ± 0.28	0.9849
Temperature: 7 K	0.1012 ± 0.0074	30.86 ± 0.67	-0.24 ± 0.02	4.74 ± 0.32	-0.54 ± 0.12	0.6448

FIGURE B.27: Fitting parameters for the 38 nm Bi_2Te_3 and 15 nm grating sample for temperatures in the range of 3 K to 7.5 K.

	$\alpha(0)$	$l_p(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 10 K	-0.0149 ± 0.0075	29.71 ± 3.65	-0.04 ± 0.02	-1.90 ± 0.33	0.26 ± 0.10	0.9884
Temperature: 12 K	-0.0139 ± 0.0500	10.84 ± 17.14	-0.17 ± 0.03	-0.05 ± 0.88	-0.33 ± 0.14	0.9925
Temperature: 15 K	-0.0108 ± 0.0015	36.09 ± 2.07	-0.17 ± 0.00	-0.45 ± 0.07	0.09 ± 0.03	0.9995
Temperature: 20 K	-0.0096 ± 0.0192	11.47 ± 9.00	-0.25 ± 0.01	0.94 ± 0.35	-0.23 ± 0.07	0.9972

FIGURE B.28: Fitting parameters for the 38 nm Bi_2Te_3 and 15 nm grating sample for temperatures in the range of 10 K to 20 K.

	$\alpha(0)$	$l_p(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 3 K	0.1140 ± 0.0015	43.10 ± 0.33	-0.68 ± 0.00	13.45 ± 0.08	-1.02 ± 0.05	0.9999
Temperature: 5 K	0.1462 ± 0.0030	32.73 ± 0.23	-0.41 ± 0.01	9.68 ± 0.14	-1.10 ± 0.06	0.9992
Temperature: 7 K	0.1040 ± 0.0031	27.44 ± 0.18	-0.22 ± 0.01	4.27 ± 0.12	-0.60 ± 0.04	0.9752

FIGURE B.29: Fitting parameters for the 18 nm Bi_2Te_3 film for temperatures in the range of 3 K to 7.5 K.

	$\alpha(0)$	$l_\phi(0)$ (nm)	β ($\cdot 10^{-7}$)	a ($\cdot 10^{-7}$)	c ($\cdot 10^{-7}$)	R -score
Temperature: 10 K	-0.0014 ± 0.0002	165.05 ± 81.21	-0.04 ± 0.00	-1.10 ± 0.02	-0.01 ± 0.10	0.9996
Temperature: 12 K	-0.0056 ± 0.0009	34.77 ± 2.22	-0.08 ± 0.00	-0.27 ± 0.05	0.05 ± 0.02	0.9993
Temperature: 15 K	-0.0019 ± 0.0015	37.08 ± 12.33	-0.11 ± 0.00	-0.05 ± 0.08	0.26 ± 0.04	0.9983
Temperature: 20 K	-0.0007 ± 0.0107	15.21 ± 63.94	-0.14 ± 0.01	0.48 ± 0.27	-0.07 ± 0.05	0.9957

FIGURE B.30: Fitting parameters for the 18 nm Bi_2Te_3 film for temperatures in the range of 10 K to 20 K.