

UNIVERSIDADE FEDERAL DE VIÇOSA

**Synthesis and Characterization of Organic Semiconductors and Topological
Crystalline Insulator Tin Telluride toward Spintronic Applications**

Robson César de Oliveira Guedes
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Thesis submitted to the Physics Graduate
Program of the Universidade Federal de
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Adviser: Joaquim B. Santos Mendes

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ABSTRACT

GUEDES, Robson César de Oliveira, D.Sc., Universidade Federal de Viçosa, March, 2026. **Synthesis and Characterization of Organic Semiconductors and Topological Crystalline Insulator Tin Telluride toward Spintronic Applications.** Adviser: Joaquim Bonfim Santos Mendes.

Spin-to-charge conversion is intensively studied in spintronics for its potential in next-generation low-power, high-performance electronics, including quantum-information architectures. Here we investigate this conversion via dynamical magnetic excitations (spin Seebeck effect and spin-pumping ferromagnetic resonance). We studied topological insulators (tin telluride) and organic semiconductors (polyaniline, PEDOT:PSS) for their respective advantages, the efficient charge–spin interconversion in the former and long-distance, chemically tunable spin transport in the latter. Structural and morphological characterizations were performed: Raman spectroscopy confirmed PEDOT:PSS film formation and UV–Vis spectroscopy yielded optical band gaps. MBE-grown tin telluride samples were characterized by Raman, high-resolution X-ray diffraction, and X-ray reflectivity. Organic-semiconductor studies used polyaniline (synthesized in-house) and PEDOT:PSS. The synthesis yielded thin films whose Raman spectra are consistent with the expected molecular structure and doping state.

The experimental study shows coherent spin transport and measurable spin–to–charge conversion across organic and topological platforms. In Pt/PANI/YIG heterostructures a 100 nm PANI film presents DC conductivities 2.5–14.3 ($\cdot\text{cm}^{-1}$) after acid doping. Ferromagnetic-resonance linewidths give $g_{\text{PANI/YIG}} = 0.56 \cdot g_{\text{Pt/YIG}}$, and spin-Seebeck analysis yields a spin hall angle of $\theta_{\text{SH}}^{\text{PANI}} = (1.70 \pm 0.06) \times 10^5$. PEDOT:PSS films (2.20 and 2.54 nm) display canonical Raman fingerprints and optical energy gaps of 2.55/2.44 eV for direct and 2.00/2.06 eV for indirect transitions, consistent with conductive polymer behavior. From the ferromagnetic resonance linewidths, we found $g_{\text{PEDOT:PSS/YIG}} = 1.46 g_{\text{Pt/YIG}}$, and spin-Seebeck analysis yields a spin Hall angle of $\theta_{\text{SH}}^{\text{PEDOT:PSS}} = (3.43 \pm 0.06) \times 10^5$. Epitaxial SnTe(111) on InP shows high crystalline quality (rocking-curve FWHM = 0.080°), out-of-plane lattice expansion ($a = 6.330 \text{ \AA}$ for 20 nm, 6.370 $\text{\AA}$ for 7 nm) and a strain-induced downshift of the A_{1g} phonon (125.3–121.6 cm^{-1}) on the Raman spectrum. Spin dynamics in Py/SnTe yield $g_{\text{eff}} = 1.4 \times 10^{18} \text{ m}^2$, $V_{\text{peak}}^{\text{SP}} = 1.71 \text{ V}$, $J_c = 3.15 \times 10^7 \text{ A/m}$, $J_s = 4.74 \times 10^4 \text{ A/m}^2$, $\lambda_{\text{IREE}} = 0.080 \text{ nm}$ and $\lambda_{\text{SH}}^{\text{sd}} = 0.24 \text{ nm}$, with $\lambda_{\text{SH}}^{\text{sd}} = 0.25 \times 10^3$, collectively supporting efficient

interfacial spin transfer and spin-to-charge conversion.

Keywords: spintronics; organic semiconductors; crystalline topological insulator; spin-to-charge conversion

RESUMO

GUEDES, Robson César de Oliveira, D.Sc., Universidade Federal de Viçosa, março de 2026. **Síntese e Caracterização de Semicondutores Orgânicos e do Isolante Cristalino Topológico Telureto de Estanho com Foco em Aplicações em Spintrônica.** Orientador: Joaquim Bonfim Santos Mendes.

A conversão de spin em carga é intensamente estudada em spintrônica por seu potencial em eletrônicos de próxima geração, de baixo consumo de energia e alto desempenho, incluindo arquiteturas de informação quântica. Aqui investigamos essa conversão por meio de excitações magnéticas dinâmicas (efeito Seebeck de spin e ressonância ferromagnética por bombeamento de spin). Estudamos isolantes topológicos (telureto de estanho) e semicondutores orgânicos (polianilina, PEDOT:PSS) por suas respectivas vantagens, a eficiente interconversão carga–spin no primeiro e o transporte de spin a longas distâncias, ajustável quimicamente, no segundo. Caracterizações estruturais e morfológicas foram realizadas: a espectroscopia Raman confirmou a formação do filme de PEDOT:PSS e a espectroscopia UV–Vis forneceu os gaps ópticos de energia. Amostras de telureto de estanho crescidas por MBE foram caracterizadas por Raman, difração de raios X de alta resolução e refletividade de raios X. Os estudos com semicondutores orgânicos utilizaram polianilina (sintetizada internamente) e PEDOT:PSS. A síntese produziu filmes finos cujos espectros Raman são consistentes com a estrutura molecular e o estado de dopagem esperados.

O estudo experimental mostra transporte coerente de spin e conversão mensurável de spin em carga em plataformas orgânicas e topológicas. Em heteroestruturas Pt/PANI/YIG, um filme de PANI de 100 nm apresenta condutividades DC de 2.5–14.3 ($\cdot\text{cm}^{-1}$) após dopagem ácida. As larguras de linha de ressonância ferromagnética fornecem $g_{\text{PANI/YIG}} = 0.56 \cdot g_{\text{Pt/YIG}}$, e a análise do efeito Seebeck de spin fornece um ângulo de Hall de spin de $\theta_{\text{SH}}^{\text{PANI}} = (1.70 \pm 0.06) \times 10^5$. Filmes de PEDOT:PSS (2.20 e 2.54 m) exibem assinaturas Raman canônicas e gaps ópticos de energia de 2.55/2.44 eV para transições diretas e 2.00/2.06 eV para transições indiretas, consistentes com o comportamento de polímeros condutores. A partir das larguras de linha da ressonância ferromagnética, encontramos $g_{\text{PEDOT:PSS/YIG}} = 1.46 g_{\text{Pt/YIG}}$, e a análise do efeito spin-Seebeck resulta em um ângulo de spin Hall de $\theta_{\text{SH}}^{\text{PEDOT:PSS}} = (3.43 \pm 0.06) \times 10^5$. SnTe(111) epitaxial sobre InP apresenta alta qualidade cristalina (FWHM da curva de rocking = 0.080°), expansão do parâmetro de rede fora do plano ($a = 6.330 \text{ \AA}$ para 20 nm, $6.370 \text{ \AA}$ para 7 nm) e um

deslocamento para menores frequências do fônon A_{1g} induzido por deformação ($125.3 - 121.6 \text{ cm}^{-1}$) no espectro Raman. A dinâmica de spin em Py/SnTe fornece $g_{\text{eff}} = 1.4 \times 10^{18} \text{ m}^2$, $V_{\text{Peak}}^{\text{SP}} = 1.71 \text{ V}$, $J_c = 3.15 \times 10^7 \text{ A/m}$, $J_s = 4.74 \times 10^4 \text{ A/m}^2$, $\lambda_{\text{IREE}} = 0.080 \text{ nm}$ e $\lambda_{\text{SH}}^{\text{sd}} = 0.24 \text{ nm}$, com 0.25×10^3 , sustentando coletivamente uma eficiente transferência interfacial de spin e conversão de spin em carga.

Palavras-chave: spintrônica; semicondutores orgânicos; isolante topológico cristalino; conversão de spin em carga

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List of Abbreviations and Acronyms

AFM	Atomic Force Microscopy
Ag	Silver
AlO _x	Aluminum oxide
AMR	Anisotropic Magnetoresistance
BEP	Beam Equivalent Pressure
BFM	Beam Flux Monitor
Bi	Bismuth
CAPES	Coordenação de Aperfeiçoamento de Pessoal de Nível Superior
CCD	Charge-Coupled Device
DFT	Density Functional Theory
e-beam	Electron-beam
FIR	Far-Infrared
FMI	Ferromagnetic (or Ferrimagnetic) Insulator
FM	Ferromagnetic
FMR	Ferromagnetic Resonance
FWHM	Full Width at Half Maximum
GGG	Gadolinium Gallium Garnet
HOMO	Highest Occupied Molecular Orbital
HRXRD	High-Resolution X-ray Diffraction
HWHM	Half-Width at Half Maximum
InP	Indium Phosphide

IREE	Inverse Rashba–Edelstein Effect
ISHE	Inverse Spin Hall Effect
LLG	Landau–Lifshitz–Gilbert equation
LUMO	Lowest Unoccupied Molecular Orbital
MBE	Molecular Beam Epitaxy
MIR	Mid-Infrared
ML	Metallic Layer
FM	ferromagnetic Material
NIR	Near-Infrared
NM	Non-Magnetic (metal)
OLED	Organic Light-Emitting Diode
PANI	Polyaniline
PEDOT	Poly(3,4-ethylenedioxythiophene)
PEDOT:PSS	Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate)
PID	Proportional–Integral–Derivative
Pt	Platinum
PVC	Polyvinyl Chloride
Py	Permalloy ($\text{Ni}_{80}\text{Fe}_{20}$)
QAHE	Quantum Anomalous Hall Effect
QCM	Quartz Crystal Microbalance
REE	Rashba–Edelstein Effect
RF	Radio Frequency
RHEED	Reflection High-Energy Electron Diffraction

RSM	Reciprocal Space Mapping
SEM	Scanning Electron Microscopy
SHE	Spin Hall Effect
SML	Spin–Momentum Locking
SnTe	Tin Telluride
SOC	Spin–Orbit Coupling
SOT	Spin-Orbit Torque
SOT-MRAM	Spin-Orbit-Torque-based Magnetic Random-Access Memory
SPE	Spin Pumping Effect
SP-FMR	Spin Pumping Ferromagnetic Resonance
SSE	Spin Seebeck Effect
STT	Spin-Transfer Torque
TCI	Topological Crystalline Insulator
THz	Terahertz
TI	Topological Insulator
TRS	Time-Reversal Symmetry
UFV	Universidade Federal de Viçosa
UHV	Ultra-High Vacuum
UV-Vis	Ultraviolet–Visible Spectroscopy
W	Tungsten
XRD	X-ray Diffraction
XRR	X-ray Reflectivity
YIG	Yttrium Iron Garnet — $Y_3Fe_5O_{12}$

rpm revolutions per minute

u.c. Unit cell

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1 INTRODUCTION

The expressive technological development driven by the Internet of Things (IoT), automation, e-commerce, and artificial intelligence is a hallmark of what some specialists are calling the Fourth Technological Revolution, or Industry 4.0. This technological advancement is shaping our workplace and how humans communicate with each other and with machines. Improvements in communication methods and multivariable data analysis have led to high volume data processing, driving the development of high-performance devices with low power consumption, low cost, and large memory storage capacity.

The size of the smallest transistor (3 nm) in a classic silicon based processor is very close to the limit of miniaturization, limiting its density and maximum clock frequency in a processor unit. The need for data processing power increases each year, in the way that the development of new parallel computing processing units, such as the Tensor Cores [1], will dictate the rate of technological evolution, mainly in the AI field. To solve this demand, industry and academia are looking for new materials or physical phenomena. Quantum computing, two-dimensional materials and quantum quasiparticles are among the most promising candidates to be used in the new revolution on data processing devices [2, 3, 4, 5].

Topological materials exhibit distinctive features that make them ideal candidates for such applications. This behavior arises primarily from dissipationless spin-polarized currents and the potential for skyrmion stabilization at topological insulator surfaces or interfaces [6]. These dissipationless channels enable electrons to propagate without energy loss. Additionally, spin momentum locking generates spontaneous spin polarization orthogonal to the electron momentum, allowing low-power electrical currents to manipulate the magnetization of an adjacent magnetic layer [7]. Moreover, the existence of skyrmions in localized magnetic nanostructures offers a promising route for binary information storage. These features make this class of materials a promising platform for quantum computing applications.

Carbon-based materials, also known as organic semiconductors, have been consolidated in the development of next-generation electronic devices due to their stable electrical performance under repeated mechanical deformations and their electrical conductivity,

which is controllable by chemical processes [8, 9]. In addition to efficient charge transport and controllable electrical resistance, certain conducting polymers exhibit high efficiency in the transport of spin currents and in spin-to-charge conversion [10, 11, 12].

In the field of electronics, conducting polymers such as Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) are widely used as hole transport and injection layers in high-performance flexible displays, such as OLEDs [13, 14, 13]. Beyond this application, organic semiconductors such as graphene, polyaniline, and PEDOT:PSS also demonstrate efficiency in the transport of spin currents generated by spin pumping ferromagnetic resonance (SP-FMR) or by the spin Seebeck effect (SSE) [15, 12, 11]. With these properties, organic semiconductors emerge as a promising platform for the study and application of flexible spintronics.

In this demanding context for flexible conducting materials and metallic semiconductors with distinctive transport properties, conducting organic polymers such as PANI and PEDOT:PSS, as well as topological insulators such as tin telluride (SnTe), emerge as promising materials for applications in high-performance and low-power information-processing devices. The dissipationless transport with spin-polarized electric currents in topological materials, as well as the scalability, mechanical flexibility, and spintronic features of organic semiconductors, make this class of materials a promising platform for the development of next-generation spintronic devices.

In this work, measurements related to spin current transport, generated by Spin Pumping Ferromagnetic Resonance (SP-FMR) and Spin Seebeck Effect (SSE) in different materials, were performed. For this, heterostructures formed by PEDOT:PSS covered with platinum on a surface of Yttrium Iron Garnet (YIG) (Pt/PEDOT:PSS/YIG) were built. The YIG film was grown on a Gadolinium Gallium Garnet substrate using the liquid-phase epitaxy method by the research group of A. Azevedo at the Universidade Federal de Pernambuco. The PEDOT:PSS was deposited by the drop-casting technique¹ and the platinum by sputtering. Heterostructures of Permalloy/SnTe grown in situ by Molecular Beam Epitaxy (MBE) on an Indium phosphide (InP) (111) substrate were also investigated using the FMR and SP-FMR techniques.

¹In Drop-casting technique a small volume of solution is directly deposited onto a substrate and allowed to dry under controlled environment conditions. The solvent evaporation leads to the formation of a solid film [16].

The investigation focused on the transport and conversion efficiency of spin current into charge currents in the Pt layer, directly in the PEDOT:PSS layer, and in the Py/SnTe/InP(111) heterostructures. For both materials, we determined the fundamental physical parameters, and the efficiency of spin current transport. This work seeks to contribute to the understanding of the main physical phenomena underlying the spin-to-charge conversion in these systems, particularly in materials with potential application in high-performance devices. We aim to contribute to innovation in high-density and low-power data processing technologies, which are key elements of the ongoing technological revolution.

This thesis is organized into three chapters. Chapter 1 presents the theoretical background related to spin-to-charge conversion phenomena, as well as the mechanisms responsible for generating spin currents. Additionally, a discussion of the materials studied in this work is provided. Chapter 2 describes the growth techniques and all the characterization methods employed for morphological, structural, and electronic analyses. Chapter 3 presents and discusses the experimental results, followed by a final discussion summarizing the main conclusions of this work.

2 THEORY AND FUNDAMENTALS

The first chapter of this work is dedicated to the presentation of the theoretical background and physical phenomena related to spin-to-charge conversion. Effects such as the spin Hall effect (SHE), Rashba–Edelstein effect (REE), and spin Seebeck effect (SSE), as well as their reciprocal effects, are presented. In addition, ferromagnetic resonance (FMR) and spin pumping ferromagnetic resonance (SP-FMR) are discussed as the mechanisms responsible for the generation of spin currents. An overview of spintronics, topological materials, and organic semiconductors is also provided. The PEDOT:PSS, PANI, and tin telluride materials are also presented in this chapter.

2.1 Spintronics

Spintronics is a branch of physics responsible for the study of spin currents and their properties. One of the main focuses is the investigation of the conversion between electric charge currents and spin currents, and vice versa. This phenomenon is called spin-to-charge conversion. The manipulation of spin currents is of great technological interest, since the manipulation of spin currents opens a new way to transport and manipulate information. One example of this is the use of spin currents to switch magnetization in thin magnetic layer through the spin-orbit torque (SOT) effect [17, 18].

In contrast to charge currents, the intrinsic energy dissipation associated with spin currents can be negligible in certain systems, such as spin-polarized currents in the topological surface states (TSS) of topological insulators, where transport occurs at surfaces or edges. Materials capable of supporting coherent spin-current transport are highly desirable for the development of new memory devices with higher storage density and lower power consumption. Spin-orbit-torque-based magnetic random-access memories (SOT-MRAM) have been demonstrated to be up to ten times more efficient than spin transfer torque (STT) devices, since no current passes through the magnetic layer, reducing energy losses due to the Joule effect [19]. Figure 1 illustrates the magnetization switching mechanisms for STT and SOT.

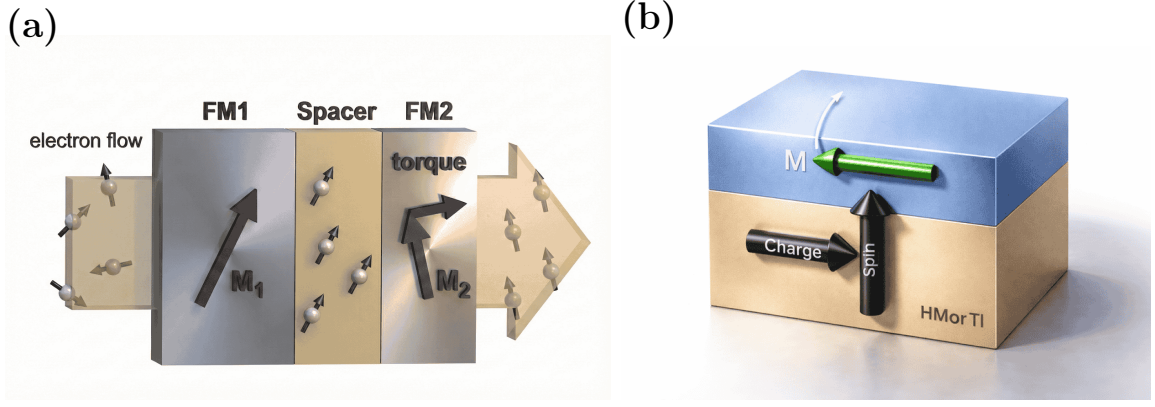


Figure 1 – Magnetization switching schemes for STT in (a) and SOT in (b). In STT, the spin-polarized electric current passes through the magnetic layer, changing its magnetization direction. In this process, the exchange interaction is dominant and is responsible for the magnetization switching. In SOT, a spin current can induce magnetization switching by means of spin-orbit coupling (SOC). The spin current can be generated by bulk (Spin Hall Effect) or surface (Rashba-Edelstein Effect) mechanisms by electric currents on TSS.

The field of study in spintronics is not limited to semiconductors or heavy metals, advanced materials such as organic semiconductors have been shown to be efficient in the transport of spin currents and their conversion into charge currents [20, 12]. This behavior can be explained by the large spin diffusion length in these materials, which allows a spin current to propagate over long distances without significant loss of its spin orientation. This feature originates from the weak spin-orbit coupling intrinsic to polymeric materials, a consequence of their low atomic number composition. This characteristic, combined with the mechanical flexibility, scalability, low cost, and electrical stability, makes them promising platforms for the development of flexible spintronic devices.

2.2 Spin currents

A spin current is the spin angular momentum flux transported by charge carriers or collective excitations called magnons. The spin angular momentum can be transported in two distinct ways, the spin-polarized current and pure spin current [21, 22]. In the case of spin-polarized current, the electric current transports a net imbalance of spins, meaning that both electric charge and angular momentum are transported. Spin-polarized currents can be generated in different ways, passing an electric current through a ferromagnetic material, by a precession in the magnetization in a resonance regime, or through the topological surface states of topological insulators. In the case of a pure spin current,

spin angular momentum is transported without an accompanying charge current. Such pure spin currents can be generated, for example, by means of SP-FMR, particularly in magnetic insulating materials such as yttrium iron garnet (YIG).

Pure spin currents can be converted into charge currents in different classes of materials, such as non-magnetic materials, magnetic conductors, semiconductors, and metals [23, 21, 22, 24, 25]. This conversion is more efficient in materials with high atomic number that leads to a strong spin-orbit coupling (SOC). Some examples are Pt, W, and Bi, as well as in topological insulators. Spin currents can also be manipulated over macroscopic length scales, for example in the magnetic TI $\text{Cr}_{0.15}(\text{Bi}_{0.1}\text{Sb}_{0.9})_{1.85}\text{Te}_3$ exhibiting the quantum anomalous Hall effect (QAHE), enabling their practical implementation in spintronic devices [18, 26, 27].

A spin current J_s can be defined mathematically in three distinct ways: through diffusive transport, quantum mechanics, or magnetization dynamics [28]. Initially, for transport in metals or semiconductors, a spin current is defined as the difference between the flux of electrons with spin-up orientation, $J_\uparrow$, and electrons with spin-down orientation, $J_\downarrow$, as

$$J_s = J_\uparrow - J_\downarrow.$$

The spin current density can be treated within a diffusive transport picture in terms of an electrochemical potential, using the concept of spin accumulation ($\mu_s = \mu_\uparrow - \mu_\downarrow$), one has

$$\vec{J}_S = \frac{\hbar \sigma_N}{2e^2} \nabla \mu_S,$$

in units of angular momentum per unit time per unit area. In this equation, σ_N is the conductivity for electrons in each spin states, e is the electron charge, and $\hbar$ is the reduced Plank's constant. Within the quantum mechanical definition, the spin current is given by the product of the spin density and the carrier velocity, both treated as operators. In this formulation, the spin current is a second-rank tensor, having both a flow direction and a spin polarization direction, and can be written as [28]

$$\mathbf{J}_s = \sum_{\mathbf{k}} s_{z\mathbf{k}} \mathbf{v}_{\mathbf{k}},$$

where $s_{z\mathbf{k}}$ is the spin density and $\mathbf{v}_{\mathbf{k}}$ is the carrier velocity.

In the context of interfaces between metals and magnetic materials, the spin current is defined in terms of the magnetization dynamics of the magnetic material, such that the spin current injected into the non-magnetic material is proportional to the vector product between the magnetization and its time derivative [28],

$$\mathbf{J}_S = \frac{\hbar g_{eff}^{\uparrow\downarrow}}{4\pi M_s^2} \left(\mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t} \right).$$

where $g_{eff}^{\uparrow\downarrow}$ is the real part of the effective spin-mixing conductance at the interface. In the present work, this latter approach is considered. Further details on spin currents will be discussed in the following sections.

Besides the extensive use of heavy metals in the spintronics field, organic semiconductors have proven to be efficient materials for the transport of spin angular momentum. In this context, conductive organic polymers have attracted interest due to their low cost and their potential application in flexible electronic devices and optoelectronics [29, 30, 31].

2.3 Inverse Spin Hall Effect

The spin Hall effect is one of the phenomena responsible for converting spin currents into charge currents in a phenomenon called spin-to-charge conversion [32]. This conversion can be driven by SP-FMR process, in which the spin current is generated by the precession of the magnetization of a ferromagnetic (FM) material, which is in turn driven by a microwave magnetic field under FMR. When the FM material is in contact with an adjacent non-magnetic metal (NM), the spin current flows across the interface and penetrates into the bulk of the NM layer, where it is partially converted into a charge current transverse to the original spin-current propagation direction. This charge current accumulates at the edges of the sample, generating a DC voltage signal that is proportional to the sample magnetization. This conversion phenomenon in three-dimensional materials is known as the Inverse Spin Hall Effect (ISHE). The first experiments of spin-to-charge conversion by means of ISHE with SP-FMR were performed with a bilayer composed by an ferromagnetic metal and a metallic layer [33, 34, 35, 36, 37]. Figure 2 illustrates the ISHE phenomenon.

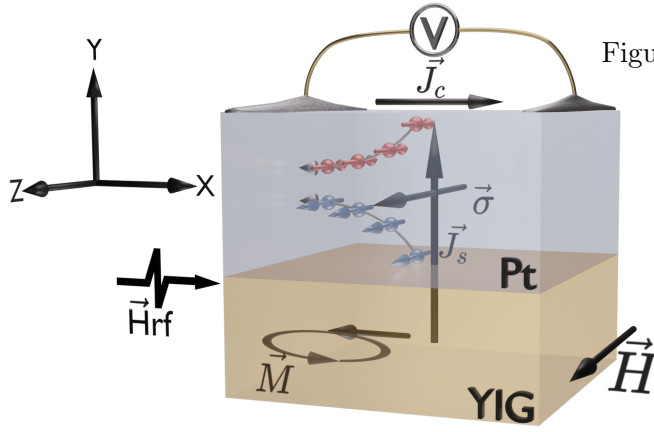


Figure 2 – Schematic representation of SP-FMR, and ISHE in spin-to-charge conversion process. The static magnetic field $\vec{H}$ induces the magnetization $\vec{M}$ along the z direction, while the microwave magnetic field $\vec{H}_{rf}$ drives the resonance of $\vec{M}$. The spin current $\vec{J}_s$ flows into the NM layer, where it is converted into a charge current $\vec{J}_c$. The spin polarization is represented by $\vec{\alpha}$.

Experiments involving spin-to-charge conversion governed by the ISHE can be performed using a metallic film as a detecting layer. This film can be a heavy metal with thickness larger than the spin diffusion length λ_N^1 , particularly when the material of interest is located between the magnetic layer and the heavy metal layer. In this regime, increasing the thickness of the heavy metal minimizes the DC signal generated by the metal itself.

The spin current density at the FM/NM interface is given by the following expression:

$$\vec{J}_S(y=0) = \frac{\hbar g_{eff}^{\uparrow\downarrow}}{4\pi M^2} \left(\vec{M} \times \frac{d\vec{M}}{dt} \right), \quad (2.1)$$

where y is the direction normal to the sample surface and also the propagation direction of $\vec{J}_S$. The solution of this expression gives the spin current density pumped out throughout the interface

$$J_S(0) = \frac{\hbar \omega g_{eff}^{\uparrow\downarrow} p}{16\pi} \left(\frac{h_{rf}}{\Delta H} \right)^2 L(H - H_r), \quad (2.2)$$

where

$$L(H - H_r) = \frac{\Delta H^2}{(H - H_r)^2 + \Delta H^2}, \quad (2.3)$$

is the Lorentzian function, and

$$p = \frac{4\omega (H_r + 4\pi M_{eff})}{\gamma (2H_r + 4\pi M_{eff})^2}. \quad (2.4)$$

¹It is the characteristic length over which a nonequilibrium spin polarization decays inside a material due to spin relaxation processes.

p describes the ellipticity factor of the precessing magnetization. Here M_{eff} is the effective magnetization inside the film, taking into account the demagnetizing field arising from the finite size of the sample. This solution can be applied to different classes of interfaces [28, 38].

In a three-dimensional system a spin current propagates into the adjacent material's bulk, this spin current undergoes attenuation due to scattering phenomena. The total equation that describes the spin current intensity as a function of its propagation distance is given by

$$J_S(y) = J_S(0) \frac{\sinh[(t_N - y)/\lambda_N]}{\sinh(t_N/\lambda_N)}, \quad (2.5)$$

where t_N represents the thickness of the ML. By means of ISHE, a spin current is partially converted in a transverse charge current by the relativistic spin-orbit coupling (SOC) which scatters electrons with opposite spins in opposite and transverse directions relative to the propagation direction of the spin current. The resulting charge current is then a consequence of the imbalance between spin-up and spin-down electrons. The density of charge current generated is given by

$$\vec{J}_C = \theta_{SH}(2e/\hbar)\vec{J}_S \times \vec{\sigma}, \quad (2.6)$$

where θ_{SH} represents the spin Hall angle and $\vec{\sigma}$ the spin polarization. It is important to note that the direction of the generated charge current is orthogonal to both the spin current and the spin polarization. Therefore, reversing the spin polarization, or the spin current direction, leads to an inversion of the measured DC signal, making this behavior a signature of the ISHE.

The efficiency of the conversion of spin current into charge current is quantified by the Spin Hall angle (θ_{SH}). In our studies, θ_{SH} is defined as the ratio between the spin current density and the charge current density, vide Equation 2.6. The Spin Hall angle is usually related to the strength of the SOC, such that high conversion values are generally observed in heavy metals due to their high atomic number. However, factors such as roughness, crystalline quality, and two-magnon scattering interfere with the magnitude of θ_{SH} [38]. As a figure of merit, topological insulators have emerged as materials with record spin-to-charge conversion rates [39].

Integrating the Equation 2.5 along the sample dimensions, we have the voltage

generated at the ends of the sample as follows [40]:

$$V_{SPE} = \frac{1}{4} R w \lambda_N e \theta_{SH} f g_{eff}^{\uparrow\downarrow} p_{xz} \tanh\left(\frac{t_N}{2\lambda_N}\right) \left(\frac{h_{rf}}{\Delta H}\right)^2 \cos \phi L(H - H_r), \quad (2.7)$$

where R is the electrical resistance of the normal metal (NM) layer, w is the width of the NM strip, λ_N is the spin diffusion length in the normal metal, e is the electron charge, θ_{SH} is the spin Hall angle of the normal metal, f is the microwave frequency, $g_{eff}^{\uparrow\downarrow}$ is the real part of the effective spin-mixing conductance at the interface, p_{xz} is the ellipticity factor of the magnetization precession, t_N is the thickness of the normal metal layer, h_{rf} is the amplitude of the radio-frequency microwave magnetic field, ΔH is the ferromagnetic resonance linewidth (half width at half maximum), ϕ is the angle between the applied static magnetic field and the direction perpendicular to the charge current (as shown in Figure 3), and $L(H - H_r)$ is the Lorentzian function describing the resonance lineshape as a function of the static magnetic field H and the resonance field H_r .

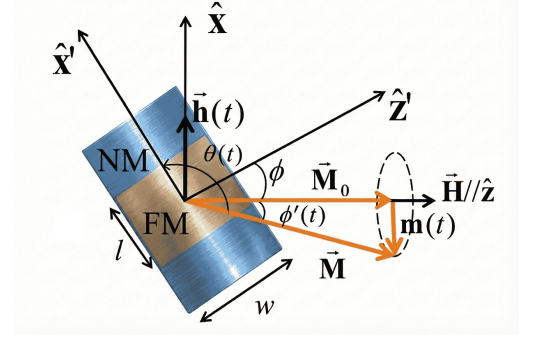


Figure 3 – Schematic of the coordinate reference system adopted in the SP-FMR measurement. Image adapted from [28].

By Equation 2.7, the inversion of the signal with the reversal of the external magnetic field is evidenced. This behavior is represented by the $\cos \theta$ dependence, which makes V_{SPE} reach its maximum and minimum at $\phi = 0^\circ$ and $\phi = 180^\circ$, respectively, while its value is zero at $\phi = 90^\circ$, assuming a material with positive θ_{SH} . At this angle, the vector product between $\vec{J}_s$ and $\vec{\sigma}$ generates a vector orthogonal to the line connecting the contacts, resulting in a zero measured voltage. This behavior is a hallmark of the ISHE.

2.4 Inverse Rashba-Edelstein Effect

Two-dimensional electron systems formed by heavy metals with high spin-orbit coupling interaction (SOC) give rise to another spin-to-charge conversion process called the Rashba-Edelstein effect (REE). This effect originates from the broken inversion symmetry at a surface or interface and from the SOC [41, 42]. An electron in a two-dimensional system, subjected to an electric field ($\vec{\epsilon}$) and a magnetic field ($\vec{H}$), has, within a relativistic treatment, a spin-dependent energy described by two terms.

$$H_Z = -\frac{e\hbar}{2mc}\vec{\sigma} \cdot \vec{H},$$

and

$$H_{SOC} = \frac{e\hbar^2}{4m^2c^2} \frac{1}{r} \frac{\partial V}{\partial r} \vec{\sigma} \cdot \vec{L},$$

where the terms are the Zeeman and the SOC energies respectively. The broken symmetry at a surface or interface of a crystal gives rise to a resulting electric field

$$\vec{\varepsilon} = -\frac{\partial V}{\partial z} \hat{z},$$

where $\hat{z}$ is the unit vector normal to the surface.

The Hamiltonian for this two-dimensional system with Rashba and kinetic term is given by

$$H = \frac{\hbar^2 k^2}{2m} + \alpha_R (\hat{z} \times \vec{k}) \cdot \vec{\sigma}, \quad (2.8)$$

where $\vec{k}$ is the wave vector for an electron in the plane, and α_R is the Rashba parameter given by

$$\alpha_R = \frac{e\hbar^2 (\partial V / \partial z)}{4m^2 c^2}.$$

From this, the energy dispersion is obtained as

$$E^{(\pm)} = \frac{\hbar^2 k^2}{2m} \pm \alpha_R k,$$

where $\pm$ signs represent the two possible orientations of spins orthogonal to the electron propagation direction. This equation shows that the energy has a parabolic dependence on the wave vector, additionally the second term makes the energy dispersion $E \pm (k_x, k_y)$ to split laterally in the $\vec{k}$ plane, as shown in Figure 4.

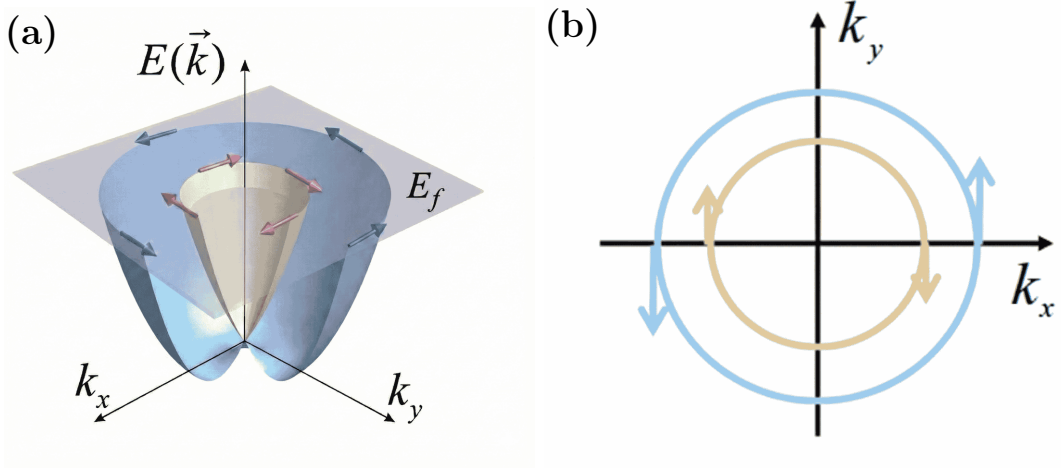


Figure 4 – Split representation of the energy dispersion for a two-dimensional electron system with the Rashba interaction (a), The red and blue arrows represent the spin orientation. The Fermi level (E_f) is presented by the constant energy defined by the plane, also the spin-momentum locking by the spin orientation tangential to the Fermi circles. Panel (b) shows a top view for a positive energy value, showing the zero net spin accumulation and electric current.

Another phenomenon that occurs as a consequence of the Rashba effect is the spin-momentum locking (SML), where the spin orientation is locked orthogonal to the momentum. The spin orientations in Figure 4 represent this effect by the circles at the Fermi level, where the spins are tangential to the circles. The SML is responsible for the spin-to-charge conversion in two-dimensional systems.

In a two-dimensional electron gas system, all the states in k -space of the spin-polarized Fermi circles are occupied, and in this sense the total momentum is zero in the equilibrium, as represented by the dashed circles in Figure 5 (a). By the application of an electric field in, for example, the $-x$ direction, a resulting force pulls the electrons in the $+x$ direction. As a result, the Fermi circles are displaced along the k_x axis. In this case, the number of occupied states with positive k_x becomes larger than those with negative k_x , so a charge current in the $-x$ direction arises with a spin polarization along the y direction.

The reciprocal mechanism of the REE is the inverse Rashba-Edelstein effect. In this effect, a pure spin current with spin polarization along the $+y$ direction, for example, when injected into the two-dimensional system, creates a net spin accumulation in the

plane. As a result, the Fermi circles are displaced in opposite directions along the k_x axis due to the non-zero resulting momentum, as shown in Figure 5 (b). With this, a charge current along the $-x$ direction is generated.

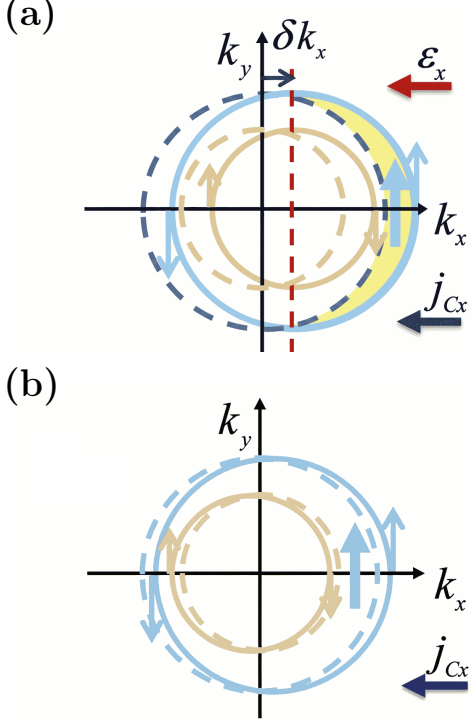


Figure 5 – Schematic representation of the direct Rashba–Edelstein effect (REE) and its reciprocal, inverse REE. The dashed lines represent the Fermi circles in the equilibrium state. Panel (a) shows the direct REE, in which a spin-polarized electric current arises when an electric field is applied. Panel (b) shows the inverse REE, in which an injected spin current produces a spin-polarized electric current.

In this sense, a two-dimensional charge current is generated from a three-dimensional spin current.

The vector relation describing the two-dimensional charge density generated is given as follows:

$$\vec{j}_C = \alpha_R (e/\hbar) (\hat{z} \times \vec{S}) \quad (2.9)$$

where α_R is the Rashba parameter, $\vec{S}$ is the nonequilibrium spin density induced by spin injection, and $\hat{z}$ is the unit vector normal to the interface related to the resulting Rashba electric field [28]. It is important to note that, in contrast to the ISHE, the direction of the generated charge current by IREE is independent of the direction of the spin current injected at the surface. It depends only on the spin polarization and on the direction of the effective Rashba electric field along $\vec{z}$. This relation can be expressed in units of (charge)/(time · area) by:

$$j_C = \left(\frac{2e}{\hbar} \right) \lambda_{\text{IREE}} J_S \quad (2.10)$$

where λ_{IREE} is called the IREE length, and it is related to the Rashba parameter as

$$\lambda_{\text{IREE}} = \frac{\alpha_R \tau_S}{\hbar}.$$

Here, τ_S is the effective relaxation time of the spin density in the two-dimensional electron gas in the presence of SML. Relating Equations 2.10 and 2.2, the charge current can be written as

$$j_C = \lambda_{\text{IREE}} \frac{e \omega g_{\text{eff}}^{\uparrow\downarrow} p}{8\pi} \left(\frac{h_{rf}}{\Delta H} \right)^2 L(H - H_r). \quad (2.11)$$

The peak voltage can be obtained from SP-FMR and SSE experiments, as described previously. Considering that $I_C = w j_C$ and that the resonance condition is $H = H_r$, one obtains

$$V_{\text{SPE}}^{\text{peak}} = \frac{1}{4} \lambda_{\text{IREE}} R w e f g_{\text{eff}}^{\uparrow\downarrow} p \left(\frac{h_{rf}}{\Delta H} \right)^2, \quad (2.12)$$

where R is the resistance of the layer and f is the microwave frequency, defined as $f = \omega/2\pi$. Thus, the IREE length can be directly determined from

$$\lambda_{\text{IREE}} = \frac{4V_{\text{SP}}^{\text{peak}}}{R w e f g_{\text{eff}}^{\uparrow\downarrow} p (h_{\text{rf}}/\Delta H)^2}. \quad (2.13)$$

By considering some examples of three-dimensional heavy metals and topological insulators, it was shown that the spin-to-charge conversion is about one order of magnitude more efficient in topological insulators, where the IREE governs the process [43]. To support this conclusion, the authors compared the λ_{IREE} obtained for the topological insulator with the product $\theta_{\text{SH}} \lambda_N$ reported for three-dimensional materials.

2.5 The longitudinal spin Seebeck effect

The spin Seebeck effect (SSE) is the phenomenon in which a spin current is generated by a thermal gradient. Its discovery in 2008 by Uchida *et al.* [44] gave rise to the field of spin caloritronics. The longitudinal SSE is usually performed using an insulating magnetic material as a source of spin currents. In this case, a ferromagnetic (or ferrimagnetic) insulator (FMI)/metallic layer (ML) bilayer is subjected to a temperature gradient ∇T applied orthogonally to the layers. This generates a magnonic spin current J_s in the FMI layer, which is injected across the FMI/ML interface into the ML layer. Upon reaching the ML, the pure spin current is converted into a charge current by the ISHE or

IREE, depending on the sample features. This current produces an electric voltage V_{SSE} at the edges of the sample, as in SP-FMR experiments.

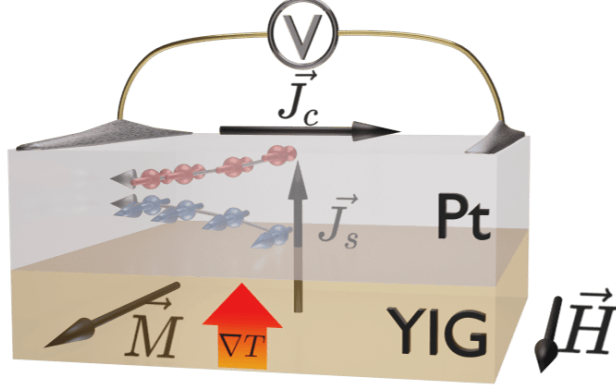


Figure 6 – Representation of an FMI (YIG)/ML (Pt) heterostructure used in a typical SSE experiment. In this scheme, the spin-to-charge conversion is governed by the ISHE.

The magnon accumulation at the FMI/ML interface produces a spin current density that flows across the interface, generating a charge current whose origin depends on the system configuration. In a three-dimensional system, a 3D spin current density generates a 3D charge current density via the inverse spin Hall effect (ISHE), given by

$$\vec{J}_C = \theta_{SH} \left(\frac{2e}{\hbar} \right) \vec{J}_S \times \vec{\sigma},$$

where θ_{SH} is the spin Hall angle and $\vec{\sigma}$ represents the spin polarization. The integration of this current density over the sample volume yields the spin Seebeck voltage V_{SSE} , taking into account the spin diffusion length λ_N , we have

$$V_{SSE} = R_{ML} w \lambda_N \frac{2e}{\hbar} \theta_{SH} \tanh \left(\frac{t_N}{2\lambda_N} \right) C_s \rho g_{eff}^{\uparrow\downarrow} \nabla T, \quad (2.14)$$

where C_s is a factor that depends on the material parameters and universal constants², and ∇T is the thermal gradient. The factor ρ depends on the effects arising from the thickness of the FMI layer, this value can be expressed by

$$\rho = \frac{\cosh(t_{FM}/l_m) - 1}{\sinh(t_{FM}/l_m)}, \quad (2.15)$$

where l_m is the magnon diffusion length. The parameter $\rho \approx 1$ for $t_{FM} \gg l_m$, and $\rho \approx 0$ for $t_{FM} \ll l_m$.

²All dependencies are discussed page 335 of Rezende, SM [28].

For the two-dimensional system case, where the IREE is the responsible for the spin-to-charge conversion, the Seebeck voltage is given by

$$V_{SSE} = R_{ML} w \frac{2e}{\hbar} \lambda_{IREE} C_{SP} g_{eff}^{\uparrow\downarrow} \nabla T. \quad (2.16)$$

Thus, by knowing the values of the electric potential and the thermal gradient, it is possible to estimate important quantities in the field of spintronics, such as λ_{IREE} .

2.6 Topological insulators

Topological insulators (TIs) are materials characterized by an insulating bulk and conducting states on their edges in two-dimensional systems or on their surfaces in three-dimensional systems. Electrons at the edges or surfaces, occupying a metallic boundary state, majority cannot be scattered by non-magnetic impurities or defects. These conducting states result from the combined effects of symmetry protection and strong SOC making them robust against perturbations that do not break the symmetry of the system [45, 46, 46, 47, 48, 49, 50]. Figure 7 shows the band dispersion in momentum space of a topological insulator. From this image, it is possible to observe the energy gap between the bulk bands, while the topological states remain gapless.

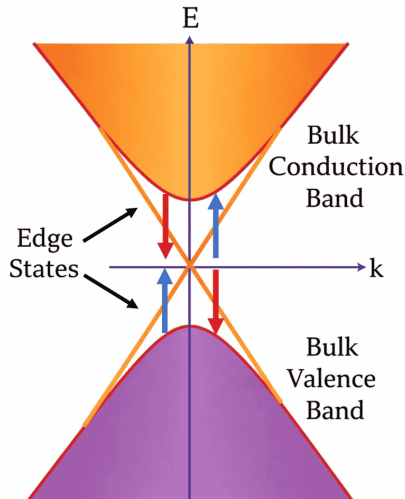


Figure 7 – Band dispersion representation constructed in the reciprocal space for a topological insulator. The TSS are the linear dispersions at the bulk band gap. Only a specific spin orientation is allowed in each edge-state band, as depicted by the blue and red arrows.

The TSS exhibit ballistic transport along the edges in 2D systems or on the

surfaces of 3D systems, meaning that charge transport occurs without energy dissipation³. Electrons propagating in the TSS experience the spin–momentum locking (SML) effect, where the electronic spin is locked orthogonally to its momentum, generating then a spontaneous spin polarization. Topological states are chiral, it means that a given electronic band transports only electrons with a specific spin orientation, while electrons with opposite spin necessarily travel with opposite momentum, as represented by the chiral edge states in the Figure 8. The SML phenomenon occurs due to the Ti band structure, combined with strong spin–orbit coupling.

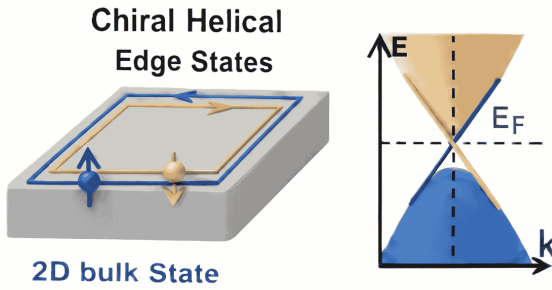


Figure 8 – Schematic representation of the quantum Hall insulator phase in a 2D material, which is analogous to that of a 2D-topological insulator. On the right-hand side, the linear band dispersion representing the edge states of this system is shown. The blue and beige colors represent the 2D bulk valence band and the 2D bulk conduction band, respectively.

The topological insulator phase arises from the nontrivial bulk-boundary correspondence of materials with strong SOC. A short explanation of this phenomenon starts with a description of an electron in a cubic, periodic, and infinite crystalline lattice where its propagation is described by the Schrödinger equation with a periodic potential [51]. As the potential is periodic, the following description is adopted

$$V(\vec{r} + \vec{R}) = V(\vec{r}), \quad (2.17)$$

where $\vec{R}$ is any vector belonging to the Bravais lattice. Due to the translational symmetry of the lattice, the Hamiltonian operator of the system is given by

$$\hat{H} = \frac{\hat{\mathbf{p}}^2}{2m} + V(\vec{r}), \quad (2.18)$$

The translation operator associated with the crystalline lattice commutes with the Hamiltonian, such that

$$[\hat{H}, \hat{T}_{\vec{R}}] = 0. \quad (2.19)$$

³At $T = 0$ K. For $T > 0$ K, inelastic backscattering processes are allowed, resulting in a finite conductivity [50].

As a consequence, it is possible to find wave functions that are simultaneous eigenfunctions of the Hamiltonian and the translation operator. From translational symmetry, one has

$$\hat{T}_{\vec{R}}\psi(\vec{r}) = \psi(\vec{r} + \vec{R}). \quad (2.20)$$

The solution of this problem is provided by Bloch's theorem, which states that the eigenfunctions of the Hamiltonian can be written as an exponential associated with the propagation of a plane wave multiplied by a function that has the periodicity of the lattice potential [51], as follows

$$\hat{T}_{\vec{R}}\psi_{\vec{k}}(\vec{r}) = e^{i\vec{k}\cdot\vec{R}}\psi_{\vec{k}}(\vec{r}). \quad (2.21)$$

A quantum number ($\vec{k}$) emerges naturally in this solution due to the periodic character of the crystalline lattice. The vector $\vec{k}$ appears in this context as the wave vector that labels the electronic states. This relation establishes that an electron propagates through the crystal as a wave modulated by the lattice periodicity, and that its energy is given as a function of the quantum number $\vec{k}$. Therefore, the different values of $\vec{k}$ enumerate all the allowed energy states of the system, constructing then its electronic band dispersion. The electronic bands are defined over the Brillouin zone, as shown in Figure 7. This definition allows their global properties (in momentum space) to be characterized by topological invariants.

A topological invariant is a property of a system that remains unchanged under continuous transformations⁴. A classical example is illustrated in Figure 9 (b), where an object with a hole can be continuously transformed from a mug into a donut without changing its number of holes, that is, the topological invariant.

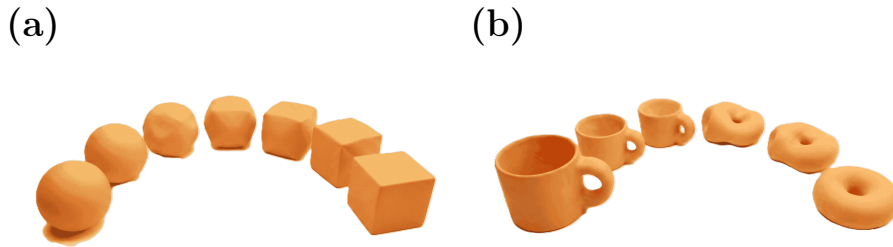


Figure 9 – Representation of real objects with topological invariant (genus) equal to 0 in (a) and 1 in (b).

⁴In the TIs case, those transformations cannot close the bulk energy gap

In this example, the genus number is the integer topological invariant associated with the topology of the surface. In the context of topological insulators with time-reversal symmetry, the topological invariant is given by the Z_2 number of the system. Accordingly, $Z_2 = 0$ represents a trivial material, whereas $Z_2 = 1$ corresponds to a topological insulator⁵. Strong SOC is central to the topological phase, as it induces band inversion, thereby characterizing the transition from the trivial state ($Z_2 = 0$) to the nontrivial state ($Z_2 = 1$). The robustness of the TSS arises from the bulk–boundary correspondence. In this context, topological insulators correspond to materials whose bulk electronic bands exhibit a nontrivial topology, which necessarily leads to the emergence of conducting states at the boundaries of the system.

The time-reversal symmetry condition ensures that the topological states of TIs are protected as long as this symmetry is preserved. This ensures that the metallic surface states are robust against nonmagnetic defects or impurities. An electron propagating in a given direction in a topological state can only be scattered by a magnetic impurity, which interacts with its spin. In contrast, backscattering by nonmagnetic impurities or lattice defects is forbidden, since the corresponding scattered energy state is not accessible without a spin flip. As conventional scattering processes do not flip the electron spin, magnetic impurities provide the only effective mechanism for backscattering. Thus, a magnetic field or a magnetic impurity acts as an agent that breaks time-reversal symmetry, lifting the topological protection of the surface states in this material.

In particular, charge currents flowing in the TSS result in a spin-polarized electric current that can generate large spin torques in an adjacent magnetic layer, thereby enabling efficient and fast manipulation of the magnetization in memory devices. This feature, together with ballistic electronic transport, makes TI materials highly promising for the development of ultra-low-power memory devices [18, 17, 26, 50, 2].

2.6.1 Topological Crystalline Insulator Tin Telluride

In contrast to conventional topological insulators protected by TRS, topological crystalline insulators (TCI) are characterized by topological phases protected by spatial

⁵These invariants are defined from the Berry connection of the Bloch wave functions. For details on the definition of the Z_2 invariant, see reference [52].

symmetries of the crystal. Mirror, rotation, glide, and screw are examples of symmetries in the lattice for a TCI [53, 54, 55]. Tin Telluride (SnTe) is a topological crystalline insulator exhibiting TSS protected by mirror symmetry. In SnTe, strong SOC leads to band inversion at multiple L points of the Brillouin zone (as shown in Figure 10). This results in multiple TSS where four Dirac cones arise on specific faces such as $\{100\}$, or $\{111\}$ as in our studies [53, 55, 54, 56].

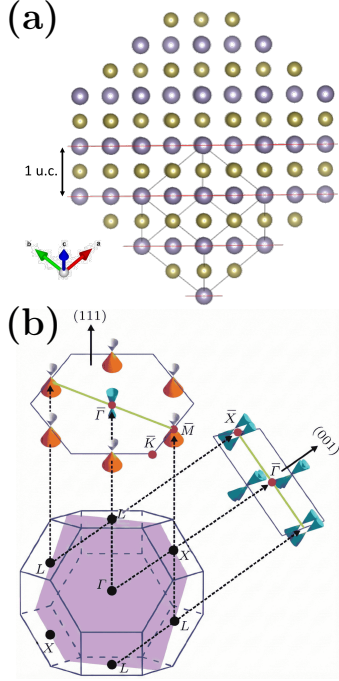


Figure 10 – SnTe (111) structure representation, the (111) direction is the up vector in the plane of the screen. Panel (b) shows the $\{100\}$ and $\{111\}$ surface projections of the first Brillouin zone for the rock-salt structure of SnTe. Four Dirac cones are present in both directions. Image (b) adapted from [57].

Due to the presence of mirror symmetry, the electronic states can be classified according to their mirror eigenvalues, giving rise to a nonzero mirror Chern number (n_M) and to the emergence of Dirac surface states. This integer topological invariant (n_M) can assume values larger than one, allowing the existence of more complex topological states. Additionally, SnTe is trivial in terms of the Z_2 invariant [53, 54].

In contrast to the TIs, the topological surface states of TCI are robust against magnetic impurities or external magnetic field, since this field doesn't create structural distortions that break mirror symmetry. In SnTe, a structural phase transition from the cubic to the rhombohedral phase can break mirror symmetry, opening a gap in the TSS. In this sense, TCIs can be more suitable for SP-FMR investigations, where magnetic fields are applied in the measurements. However, high crystalline ordering must be achieved in order to ensure mirror symmetry. These characteristics make SnTe an appealing platform for exploring symmetry-engineered topological phases and their interplay with magnetism

and spin-dependent transport phenomena.

2.7 Organic semiconducting materials

The physical properties of polymers such as flexibility, mechanical strength, low density, and high dielectric rigidity, justify their wide use as insulating materials in traditional electronics [9, 58, 59]. Although the vast majority of polymers are naturally electrical insulators, this characteristic is not always true. As early as the 1960s, organic molecules capable of transporting electrical charge were identified. However, their excessively rigid molecular chains, along with resulting handling difficulties, insolubility, and infusibility, limited their technological potential [60].

In the early 1970s, Shirakawa and Ikeda demonstrated the possibility of producing polyacetylene films through the polymerization of acetylene [61]. In 1977, MacDiarmid and collaborators [62] revolutionized this field of research by discovering that treating polyacetylene with acid (*doping*) could increase its electrical conductivity by up to 13 orders of magnitude, enabling this polymer to reach a conductivity close to that of metallic copper at room temperature. The notable conductivity achievable by these materials, also known as organic semiconductors are comparable to doped semiconductors [63]. A comparison between conjugated polymers and semiconductors are illustrated on the scale presented in Figure 11.

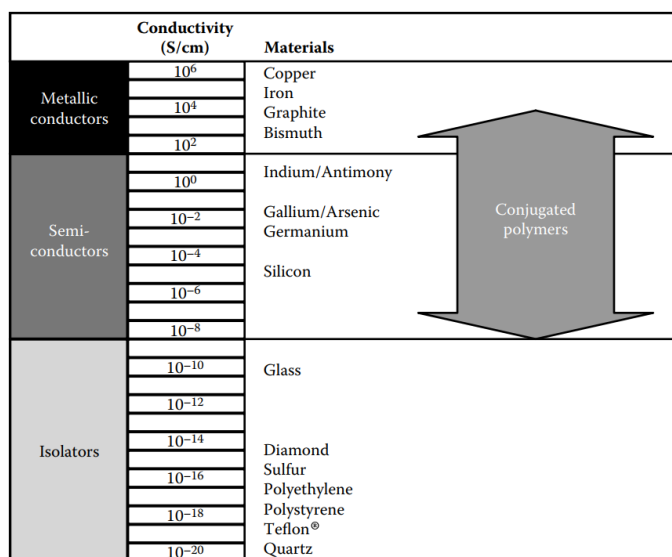


Figure 11 – Electrical conductivity of organic semiconductors compared with metals, semiconductors, and insulators. Image extracted from [64]

Electrical conduction in organic semiconductors is a complex phenomenon that differs significantly from conduction in metals or conventional inorganic semiconductors. The absence of a well structured periodic crystal lattice makes the complete description difficult of the transport phenomenon by a quantum description. One more simple description is that the electric charge conduction occurs primally in the π -conjugated bonding system along the polymer backbone (see Figure 12) and also by the formation of additional charge carriers through acid or base doping.

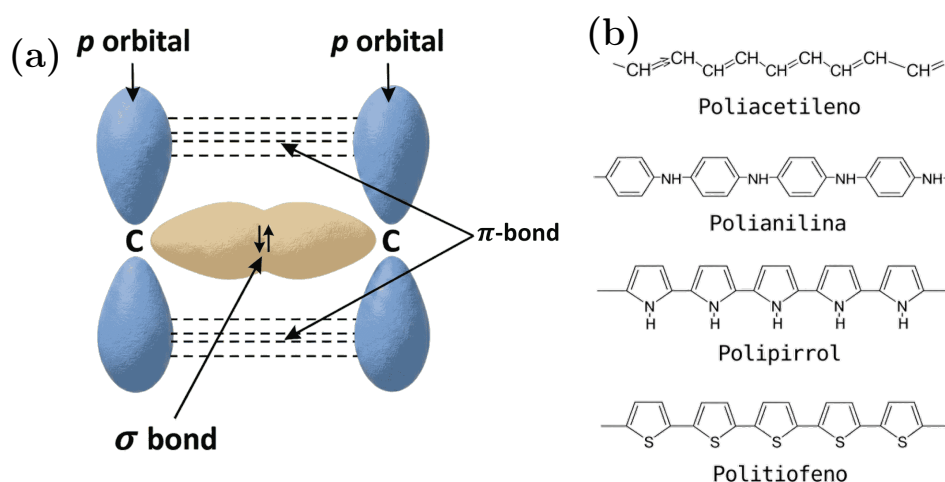


Figure 12 – Schematic representation of the formation of σ and π bonds in a $\text{C}=\text{C}$ double bond, showing the overlap of p orbitals in (a). Panel (b) shows a schematic chemical structures of conductive polymers highlighting their π -conjugated systems along the main chain. Image (a) adapted from [65] and (b) from [60].

Insulating polymers are predominantly composed of sigma (σ) bonds along their polymer backbone, resulting in strongly localized electrons. This leads to a large energy gap between the HOMO (Highest Occupied Molecular Orbital) and LUMO (Lowest Unoccupied Molecular Orbital), making them insulating⁶. In contrast, conducting polymers exhibit an alternation of single and double bonds along their polymer backbone, forming a system of π -conjugated bonds as shown in Figure 12 (b). This conjugation, also called resonance hybridization, enables the delocalization of π electrons across the atoms leading to the formation of a lower energy band gap due to the charge distribution across the polymeric structure, as depicted in Figure 13 (a) [60, 66, 67, 64]. Conjugated polymers exhibit a smaller bandgap between the LUMO and HOMO compared to insulating ones, giving

⁶In the context of organic semiconductors, HOMO and LUMO play the roles of the valence band and conduction band, respectively.

them semiconductor-like properties.

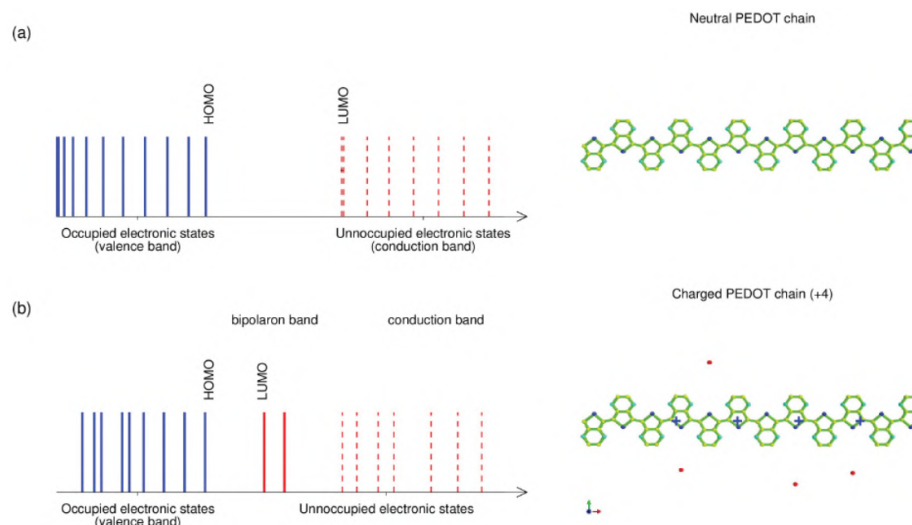


Figure 13 – Schematic energy level distribution for a neutral PEDOT chain (a) and for a positively charged PEDOT chain with charge +4 (b). Charge injection introduces new electronic states within the gap, leading to a reduction of the effective energy gap. Image taken from Ref. [63].

Most conjugated polymers, in their intrinsic state, are semiconductors with low electrical conductivity. To enhance its conductivity, a doping process can be performed. During the doping process, electric charge are added to or removed from the conjugated polymer chain through the introduction of chemical agents (dopants). The term *doping* is used by analogy with doping in inorganic semiconductors, however, the dopants (acids or bases) used in polymers are typically added in much higher quantities than those used for inorganic semiconductors. In fact, the dopant mass in polymers can reach up to 50% of the initial mass of the compound [68].

Doping in conjugated polymers can occur via oxidative (p-type) or reductive (n-type) processes. In oxidative doping, the dopant agent removes electrons from the polymer's HOMO band, creating positive charge carriers. This is the most common doping type applied to conducting polymers such as PEDOT, which becomes a cationic polymer through this process. In reductive doping, on the other hand, the dopant agent adds electrons to the polymer's LUMO band, generating negative charge carriers (electrons). n-type doping is generally less stable in air compared to p-type doping.

The field of study in organic semiconductors has gained prominence in recent

decades due to their unique combination of efficient electrical charge transport and the intrinsic mechanical flexibility of polymeric materials, enabling their application in flexible and stretchable electronic devices [69, 64, 9, 30, 31]. Due to the study of these materials, new technologies have been developed, making it possible to produce incredibly thin and light devices. The organic light-emitting diode, better known as OLED, is an example of a device manufactured with an organic semiconductor matrix. Figure 14 shows a flexible screen manufactured from OLED. Conducting polymers also have enormous potential for applications in areas such as solar cells, electromagnetic shielding films, plastic batteries [60, 61, 62, 68], and others.

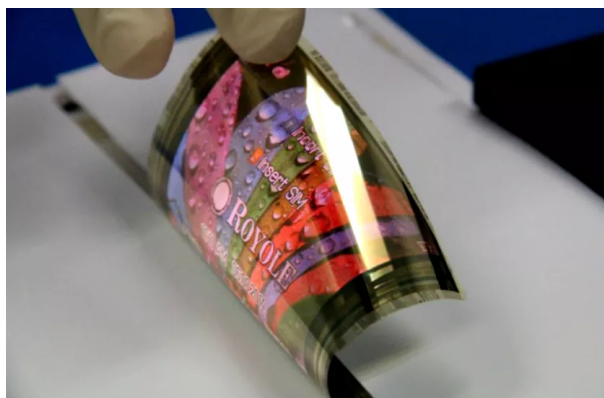


Figure 14 – Flexible OLED screen with a thickness of 0.01 mm. Image extracted from [70].

In addition to the remarkable applicability of conducting polymers in electronics and optoelectronics, as previously mentioned, organic semiconductors have attracted increasing interest in the field of spintronics. Conducting polymers such as polyaniline, PEDOT:PSS, and 2D graphene have been shown to be effective for spin-current transport and for spin-to-charge conversion [71, 20, 15]. However, the disordered manner in which the molecules of these polymers usually are arranged represents a challenge to be overcome. In order to mitigate the low degree of structural ordering, techniques such as atomic (or molecular) layer deposition can be employed to enhance control over the generation, transport, detection, and manipulation of spin currents in this class of materials [72, 73].

2.7.1 The organic semiconducting polymer PEDOT:PSS

The Poly(3,4-ethylenedioxythiophene) (PEDOT) is a carbon-based conducting polymer that exhibits a remarkable conductivity value that can be larger than high doped

semiconductors [74]. The PEDOT was developed by Bayer AG in 1988 [75] and it remains one of the most widely applied polymers in the flexible electronic industry due to its high conductivity (reaching up to 6,000 S/cm), low cost, ability to form transparent films in the visible spectrum, stability and processability, justifying its broad adoption in both industrial and academic research [76, 74, 75, 77, 78, 79, 80, 81, 82].

For optimizing its solubility, the PEDOT is commonly used in the macro-molecular salt form with the poly(estirenesulfonate) (PSS), so forming the poly(3,4-ethylenedioxythiophene):polystyrene sulfonate composite that is widely known by its acronym PEDOT:PSS. The PSS acts as a counterion stabilizing PEDOT the PEDOT in the disperse state providing the necessary structuring to the thin films development [83]. The structural part PEDOT is the oxidized component of the compound and is responsible for the conductivity of the PEDOT:PSS complex [79].

The high conductivity is due to the unbalanced distribution of charges along its polymeric chain, forming polarons and bipolarons⁷, which are responsible for the electrical conduction of the entire polymeric complex. Additionally, high carrier mobility is correlated with the degree of order and the narrow stacking between polymer chains [84]. Figure 15 represents the PEDOT:PSS structure.

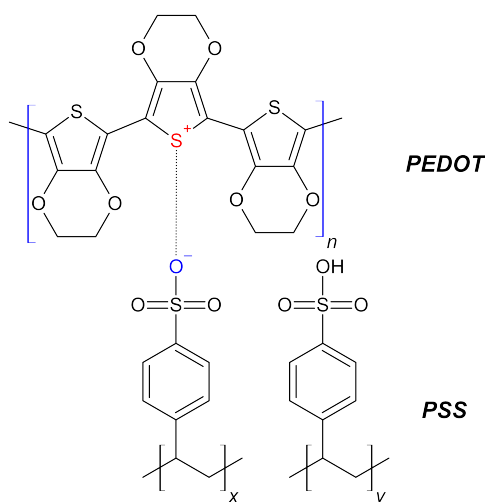


Figure 15 – Chemical representation of the PEDOT:PSS structure, where n , x , and y represent the repetition units.

⁷Polarons are quasiparticles formed by a charge carrier coupled to local lattice distortions in a material. Bipolarons correspond to bound states of two charge carriers coupled to the lattice.

PSS is the insulating component of the polymer. It contributes to the stability and processability of the material by enabling PEDOT to be dispersed in aqueous solution. PSS contains deprotonated sulfonate groups, $-\text{SO}_3^-$, meaning that they have lost a proton, H^+ , and therefore carry a negative charge. These groups act as counter-ions for the positive charges on PEDOT, stabilizing the dispersion. In an environment with excess PSS, the electrostatic interaction between positively charged PEDOT and negatively charged PSS forms a nanoscale core-shell structure, as illustrated in Figure 16.

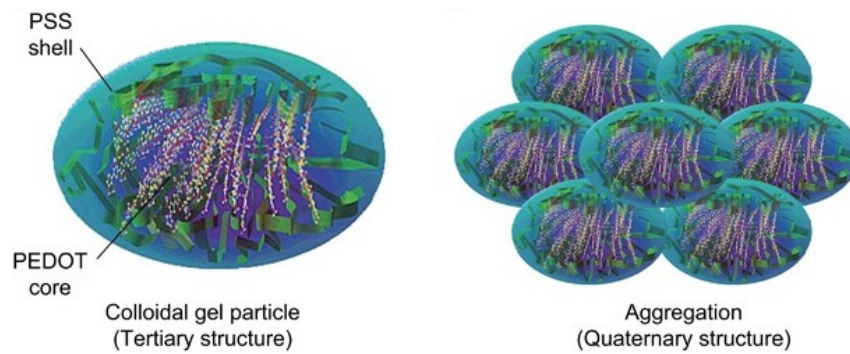


Figure 16 – PEDOT:PSS chain inside of core-shell structures and its agglomeration representation. Image extracted from [85].

The proportion between PEDOT and PSS compound has a direct influence on the composite conductivity. This proportion determines the charge balance in the polymeric chain. The electrical conductivity of PEDOT:PSS can be significantly increased by the addition of solvents as ethylene glycol or dimethyl sulfoxide or also by the pH tuning [86]. The high value of conductivity can be reached for pH values between 0 and 3 [87]. In an acid environment, the sulfonate groups $-\text{SO}_3^-$ can accept one proton becoming sulfonic acid $-\text{SO}_3\text{H}$ in a process called reprotonation. That change can lead to the structural reorganization or phase separation of PEDOT:PSS. This process allows the PEDOT parts approaching each other forming more efficient ways to the electrical charge transport leading to a high enhancement in the conductivity. The thiophene rings form a π -conjugated system with p -type doping and for each three thiophene rings created, one positive charge is present. This is due to the oxidative polymerization reaction triggered by radicals. In this way, PEDOT is the component responsible for the conductivity for the conductivity of the PEDOT:PSS group [64].

Due to its mechanical flexibility, transparency and high electrical conductivity,

PEDOT:PSS is widely applied in the organic optoelectronic devices field. In this case, PEDOT:PSS is commonly used to enhance hole injection in Organic Light Emitting Diodes (OLED) improving its efficiency [14, 13]. Other applications for this polymer include the fabrication of supercapacitors, organic plastic solar cells, cathode ray tubes, rechargeable batteries and other advanced electronic devices [74, 88, 78, 89, 90, 91, 92, 93, 94].

Conductive organic polymers are composed of light atoms and, consequently, exhibit weak values of SOC. Although this feature is not favorable from the perspective of spin-to-charge current conversion, it makes this material well suited for spin current transport. This is because a spin-polarized current is not strongly scattered by the SOC in this material, allowing for efficient spin propagation.

2.7.2 The organic semiconducting polymer polyaniline

Polyaniline (PANI) is a p-type organic semiconducting polymer and one of the most widely studied conjugated polymers in both academia and industry, similarly to PEDOT:PSS [95, 96, 60, 64]. PANI stands out due to its ability to be synthesized either chemically, as performed in this work, or electrochemically, which enables the fabrication of low-cost films with simple and accessible processing. From the perspective of electronic structure and charge transport, PANI exhibits metallic transport. It can be doped in acidic or basic solutions, and this doping process can be reversible [95, 96, 60, 97]. A schematic representation of the chemical structure of PANI is shown in Figure 18.

In PANI thin films, polarons act as charge carriers. The overlap of electronic wave functions along several polymer chains leads to the formation of a polaronic band, in which the Fermi level is located. This band defines the allowed energy levels for electrical charge transport [95, 98].

The manipulation of electrical conductivity is intrinsically related to the concept of secondary doping [67, 85, 8]. Phenomenologically, the treatment of an already doped polymer, in which primary doping is performed during the synthesis process, involves the use of a substance acting as a secondary dopant, either acidic or basic. This treatment can increase the electrical conductivity by several orders of magnitude. This process induces changes in the electronic spectra and in the crystallinity, which often persist even after the removal of the secondary dopant. For instance, in thin films immersed in acidic solutions,

the conductivity increases and remains stable even after complete drying [67, 85, 8].

Further details about PANI will be introduced in the section dedicated to its synthesis in **Polyaniline electrochemical synthesis** section.

2.7.3 Spintronics in Organic Semiconductors

Pure spin current presents advantages compared to conventional electronic current. It does not result in energy dissipation through the Joule effect and can propagate through a wide variety of conductors, as well as organic semiconductors. The remarkable capability for spin current injection, transport, and conversion in semiconducting materials has been demonstrated in many studies [99, 100, 101, 102, 12].

In organic semiconductors, the long spin relaxation lifetime (on the order of $10\ \mu\text{s}$) [103, 20, 104] and the spin diffusion length exceeding 500 nm (significantly larger than those observed in heavy metals) make this class of materials promising platforms for spintronic device applications [71, 97, 105, 106, 20, 99]. This behavior is mainly attributed to the weak SOC, as organic molecules are composed of light atoms such as hydrogen, carbon, and oxygen.

The spin-to-charge conversion was performed for the first time in the conductive polymer PEDOT:PSS by means of SP-FMR. A ferrimagnetic insulator substrate (YIG, $\text{Y}_3\text{Fe}_5\text{O}_{12}$) was used to produce the pure spin current that diffuses into the polymeric layer and was then converted into charge current [20]. In three-dimensional systems, the mechanism underlying spin-to-charge conversion is usually attributed to the inverse spin Hall effect. However, later studies revealed that the physical origin of the dc voltage observed in the PEDOT:PSS/YIG bilayer under FMR is mainly associated to a temperature gradient induced by the propagation of nonreciprocal magnetostatic surface spin waves [10].

Despite the significant progress and research in organic spintronics, the physical mechanisms underlying spin-to-charge conversion in organic semiconductors remain under debate. The weak SOC, which enables long spin transport distances, simultaneously limits the spin-to-charge conversion efficiency through commonly discussed mechanisms, such as the inverse spin Hall effect. Furthermore, experimental results can be influenced by interfacial effects and thermally induced voltages. Phenomena such as the spin Seebeck

effect and the Rashba–Edelstein effect can play a role in the spin-to-charge conversion process, describing the relationship between the arising of spin currents. In this sense, complementary techniques need to be employed in these materials in order to distinguish the different contributions. Finally, structural engineering of organic semiconductors is highly desirable to enhance spin-to-charge conversion and to improve the distinction between the underlying mechanisms [107, 11, 69, 8, 67].

In this context, conductive polymers emerge as promising platforms due to their chemical conductive tunability via the doping process, low cost, ease of growth, and compatibility with long spin diffusion length based approaches [66, 69, 30, 8, 97]. Among them, polyaniline and PEDOT:PSS stand out, offering controllable electrical properties and easy-to-grow processes. Their structural differences, such as molecular weight, make them suitable for distinguishing the influence of structural changes on spin-to-charge conversion and on the dominant underlying mechanism.

3 MATERIALS SYNTHESIS AND CHARACTERIZATION OF ORGANIC SEMICONDUCTORS AND TOPOLOGICAL INSULATORS

The first part of this chapter is dedicated to the presentation of the synthesis routes of the polymeric organic semiconductor polyaniline (PANI), as well as the fabrication of its thin films and of the PEDOT:PSS polymer. In addition, the method used for the growth of thin films of the crystalline topological insulator tin telluride (SnTe) is presented. The second part of this chapter is dedicated to a brief overview of the experimental techniques applied to these materials. The experimental techniques include atomic force microscopy (AFM), Reflection High-Energy Electron Diffraction (RHEED), high-resolution X-ray diffraction (HRXRD), Raman spectroscopy, spin Seebeck effect (SSE) experiments, ferromagnetic resonance (FMR), and spin pumping ferromagnetic resonance (SP-FMR). Most of these techniques were applied to all polymeric and solid-state semiconductor samples, with the exception of RHEED, which was exclusively applied to SnTe, and SSE, which was exclusively applied to PANI.

3.1 Material Synthesis and Sample Preparation

3.1.1 Polyaniline electrochemical synthesis

The synthesis of PANI is presented in this section on methodology and characterization techniques, however, this section can also be considered as a result of this work, since the synthesis successfully produced the expected polymer.

The first step in the PANI synthesis process was obtaining the purified aniline solution. This solution must mainly be composed of aniline monomers, this is achieved through the fractional distillation process [108]. During this procedure, aniline is heated in a volumetric flask at a constant temperature of 80 °C, then the vapor is condensed in a Liebig condenser and stored in an Erlenmeyer flask. All procedures were performed protected from light to avoid the aniline photo-oxidation [109, 110]. The purified aniline was stored below 10 °C and also protected from ambient light. Figure 17 (a) illustrates the aniline purification process.

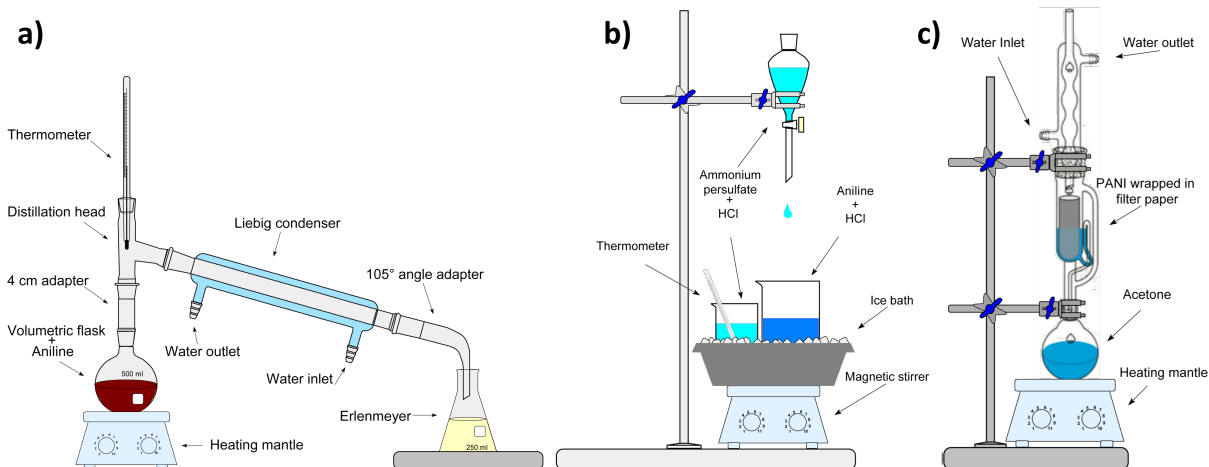


Figure 17 – Panels (a),(b), and (c) shows the experimental apparatus used for aniline distillation, PANI polymerization, and PANI filtration respectively.

During the polymerization process (see Figure 17 (b)), a solution of ammonium persulfate ($(\text{NH}_4)_2\text{S}_2\text{O}_8$) and hydrochloric acid (HCl) was slowly dripped into the solution of aniline ($\text{C}_6\text{H}_5\text{NH}_2$) and HCl [111]. The entire procedure lasted 12 hours and was carried out at a temperature below 0°C . After this procedure, the PANI solution was filtered to obtain the long chains, through the solid formed in the filter. The formed PANI solid was then washed with acetone (CH_3CH_3) using the Soxhlet extraction procedure for six hours, the apparatus is shown in Figure 17 (c) [112, 111]. This procedure was performed to remove the short-chain PANI from the obtained solid. The aniline polymerization procedure produces PANI in the emeraldine salt state (see Figure 18 (b)), which is the state with the highest electrical conductivity for this polymer [113, 114, 115, 116].

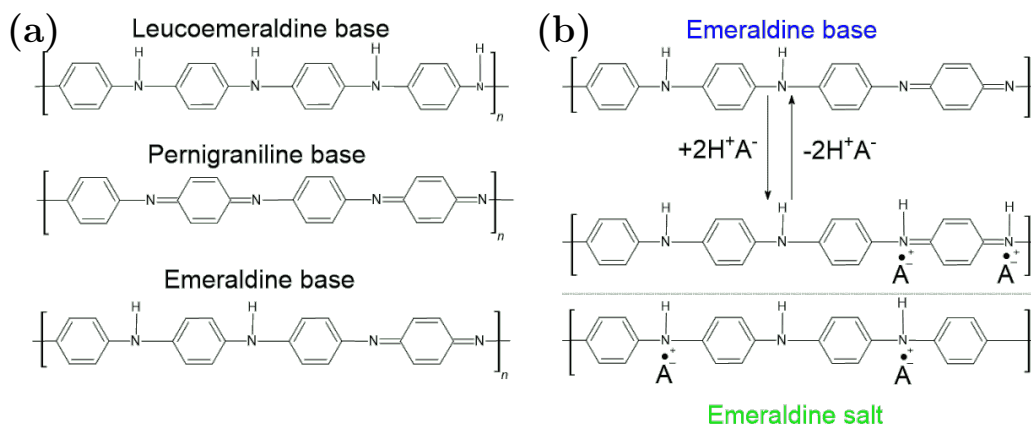


Figure 18 – Panel (a) exemplify the possible oxidation states of PANI. (b) The emeraldine base is achieved by dedoping procedure with a basic solution. The doping–dedoping process of the PANI chain. This process occurs by subjecting the polymer to acid or base treatment. Here A^- is the counter-ion of the acid molecule used to dope the polyaniline. The dot above A is a symbol indicating ionic separation. The polaron (mainly associated with amine sites¹) and bipolaron (associated with imine sites) configurations of the emeraldine salt are represented. Polyaniline films produced with emeraldine base form exhibit a bluish color, whereas those in the emeraldine salt form display a green coloration.

To ensure greater structural stability for the synthesized polymer, PANI was stored in the emeraldine base state (see Figure 18 (b)). For this purpose, the PANI solid was submitted to a **dedoping** procedure in 0.1 M ammonium hydroxide (NH_4OH) aqueous solution in order to remove the H^+ atoms bound to the polymeric chain. The solutions was filtered producing dark slabs as shown in Figure 19. For more efficient and complete drying, the PANI solid was ground using a porcelain mortar and pestle, and then stored in a desiccator containing 100 grams of silica for 72 hours. The crushed PANI (see Figure 19 (c)) was then kept in a low-humidity and light-protected environment [113, 114].

¹An amine group is characterized by a covalent bond between carbon and nitrogen atoms, while an imine group possesses a double bond between these atoms.

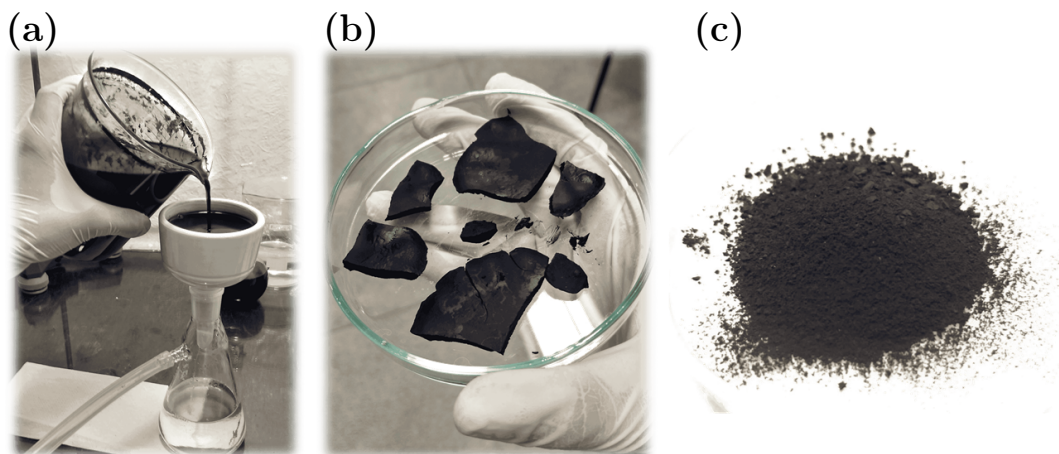


Figure 19 – Filtration process of the base emeraldine PANI. From panel (a) can be observed that the filtered solution is completely transparent. The obtained PANI solid is shown in panels (b) and (c).

The conductive PANI in the emeraldine salt state, used for the production of thin films, is obtained by mixing the previously obtained solid with formic acid CH_2O_2 85%. After mixing, the solution was passed through a porcelain filter with a porosity of 16–40 μm . The conductive PANI solution obtained by filtration was stored at low temperature and protected from light [117, 118].

At this stage, the prepared solution is ready for thin film fabrication. In this work, we employed the spin-coating technique to produce PANI thin films of varying thickness. In the spin-coating technique, a small volume of solution is deposited onto the substrate and rotated at a determined speed. The centrifugal force spreads the solution uniformly across the surface, forming a homogeneous thin layer. The final thickness depends on parameters such as rotation speed, time, and solution viscosity [119]. Further details are provided in the Result chapter.

3.1.2 PEDOT:PSS sample production

The PEDOT:PSS polymer used in this work was commercially obtained from Sigma-Aldrich [120]. The synthesis of the used PEDOT:PSS occurs through chemical oxidative polymerization in an aqueous medium, the oxidant agent usually can be ammonium persulfate $(\text{NH}_4)_2\text{S}_2\text{O}_8$ or sodium persulfate $(\text{Na}_2\text{S}_2\text{O}_8)$ [89, 64, 121]. The process is based on the dispersion of the EDOT monomer in a solution containing PSS, which acts

both as a template and as a dispersing agent. The reaction is initiated by the addition of an oxidizing agent (such as persulfate, for example), which removes electrons from EDOT and generates cationic radicals. These radicals couple to form PEDOT chains that, being insoluble and positively charged (doped state), bind electrostatically to the negatively charged PSS chains. The final result is a stable colloidal complex, in which PSS ensures water solubility, while PEDOT provides the conjugated network required for electronic conduction. A representative of PEDOT:PSS polymeric chain is shown in Figure 15.

For the purpose of studying spin-current transport, thin films of this compound were fabricated. The spin-coating technique was initially chosen for the fabrication of PEDOT:PSS films. However, the resulting films were not uniform on the YIG substrate due to the low interfacial affinity between the solution and the substrate. To overcome this limitation, the drop-casting technique was employed for film fabrication. More details about PEDOT:PSS samples are discussed in the Chapter three.

3.1.3 Molecular Beam Epitaxy

This section provides an overview of molecular beam epitaxy (MBE), a technique widely employed for growing thin films and nanostructures with high crystalline quality. The main components of the MBE system and the physical phenomena governing epitaxial growth are described in this section. The relevance of MBE for the growth of tin telluride thin films is emphasized, setting the stage for the discussion of future experimental results.

MBE is an epitaxial growth technique that operates in a non-equilibrium environment and is used for the production of epitaxial films of metals, semiconductors, and insulators. In the MBE method, which is a type of physical vapor deposition technique, growth occurs atomic layer by atomic layer in an ultra-high vacuum (UHV) environment, using atomic or molecular beams generated by heated effusion cells directed onto a heated substrate. The term *epitaxy* means the atomic (or molecular) deposition that occurs in parallel planes with the same, or nearly the same, lattice parameter as the substrate, following its crystallographic order. MBE is notable for producing films with high crystalline quality, where doping and composition can be finely tuned thanks to the precise temperature and shutter control system. High-quality effusion cells and in-situ observation techniques make MBE a distinct, reliable, and versatile growth method. Additionally,

sharp interfaces between different materials, which are crucial for the development of optoelectronic devices, can be achieved [122, 123].

High-quality films produced by MBE require ultra-high vacuum (UHV) around the effusion cells and the substrate, this condition is fundamental for ensuring the quality of the films [124]. The main growth chamber of an MBE system is maintained under UHV, with a residual pressure usually below 10^{-9} Pa (10^{-11} Torr). This condition is essential to guarantee that the mean free path of the evaporated particles is greater than the distance between the sources and the substrate, thereby minimizing collisions with residual gas molecules.

Even at 10^{-9} Pa, the number of residual N_2 and O_2 molecules inside the growth chamber can exceed approximately 10^5 cm^{-3} [124], and some of these molecules will be incorporated into the grown crystals. In modern MBE systems, UHV is achieved through careful chamber design and advanced vacuum engineering. These systems typically consist of three stainless steel chambers connected in series, with vacuum pumps attached. The inner surfaces of the chambers are specifically designed to reduce gas absorption. Additionally, some molecules are absorbed by the inner chamber surfaces, so a baking procedure is required to achieve UHV and enable the growth of high-purity materials. The baking process is performed at temperatures of approximately 200°C , and each chamber has its own powerful vacuum pump to handle the heavy load during all the process. The vacuum system usually includes a titanium sublimation pump in addition to liquid N_2 cryopanel, turbomolecular, ion, or oil diffusion pumps, as illustrated in Figure 20 (a).

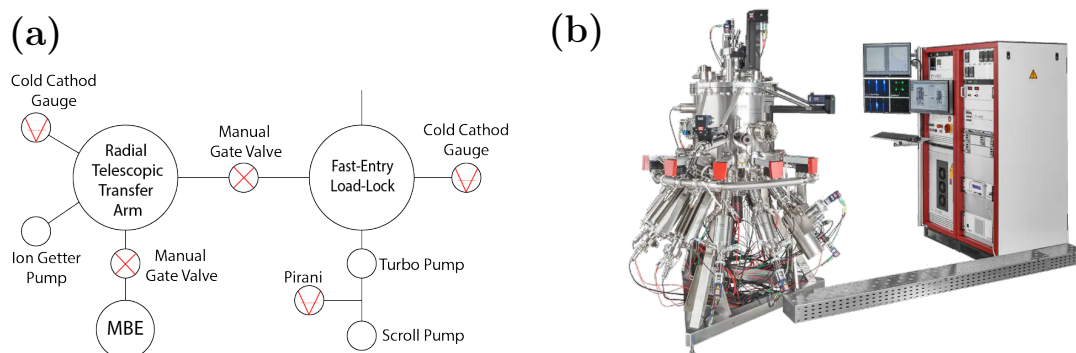


Figure 20 – Schematic diagram (a) of the MBE system, labeling the main stages. Panel (b) shows a complete MBE system. Image (b) taken from [125].

The MBE system uses Knudsen effusion cells or electron-beam evaporators (e-

beam) as material sources. Effusion cells consist of crucibles, shutters, and a resistor, they are used for materials with relatively low melting points (Ga, In, As, Sb, Se, Sn, Te for example) or high vapor pressures. The crucible is a container made of low-diffusion materials such as Ta, W, or Mo, capable of resisting high evaporation temperatures. It holds the material to be evaporated and controls the molecular beam intensity. The shutters act as gates, allowing or blocking the molecular beam flux directed toward the substrate. The resistors are responsible for heating the material to its evaporation temperature. They are made of materials with high electrical resistivity and thermal stability (Ta, W for example), even at high temperatures. Resistors are usually constructed in a spiral shape or as a folded strip around the crucible. The heat generated by the resistor is controlled by the applied current and monitored by a proportional-integral-derivative (PID) controller allowing fine tuning of the molecular flux due to the nearly constant temperature precision, with fluctuations below 1 °C.

E-beam sources are used for materials with the highest melting points and low relative vapor pressures², such as Co, Zr, Nb, Al, Mo, and Permalloy ($Ni_{80}Fe_{20}$). The e - beam is generated by thermionic emission from a tungsten filament, with a potential difference ranging from 4 kV to 10 kV. The current applied to the filament can reach 50 A, while the resulting emission current from the filament is typically between 10 mA and 100 mA. The accelerated electrons are guided to the crucible by a magnetic field and the anode-cathode electric potential difference. The source material inside a hearth can be heated to approximately 10^4 K at the point of impact of the electron beam. The current in the filament is controlled to increase or decrease the number of bombarding electrons, which in turn adjusts the amount of evaporated material. The evaporated material is directed toward the substrate, where it precipitates upon impact. The hot surface of the substrate absorbs the molecules, and through chemical reactions and kinetic diffusion, a lower-energy interaction level is established and the film is formed [122]. The growth rate in MBE is usually measured in Å/s, allowing precise control over the growth of thin films down to a few monolayers (single unit cells). Additionally, the chamber walls also absorb some molecules. For this reason, a baking process must be periodically performed to clean the chamber and avoid further contamination.

²This means that even at high temperatures, low pressure is required for the evaporated material flux to be significant.

One of the most important steps for high-quality sample growth is the proper preparation of the substrate. This procedure must be carried out carefully, avoiding as much as possible any contact with the substrate surface and preventing exposure it to the open environment. The use of a face mask, safety glasses, gloves, bouffant caps, and a cleanroom lab coat is always recommended. In addition, single-use lint-free laboratory wipes and dedicated cleaned tweezers and screwdrivers are mandatory. For some substrates, such as yttrium iron garnet (YIG), even the standard ultrasonic cleaning must be avoided, as they left organic residues and carbon, reducing wettability and leading to poor adhesion of subsequently deposited layers [126].

Purchased substrates usually require treatment to achieve atomically flat and largely defect-free surfaces, which are essential for subsequent epitaxial crystalline growth. This can be obtained through heat treatment, known as annealing. Another method used to clean the substrate surface is outgassing. In this process, the substrate is loaded into the MBE deposition chamber and heated to high temperatures ($\approx 600^\circ\text{C}$) under a UHV environment. This procedure removes water and embedded impurities from the substrate surface.

To be loaded into the MBE deposition chamber, the samples are mounted on a sample holder and then placed in the manipulator inside the growth chamber. Figures 21 (a) and 21 (b) shows the sample holder and the manipulator. In this configuration, the substrate is secured by two lateral strips of tantalum foil, tightened with four screws. Alternatively, spot welding or melted indium can also be used to attach the substrate to the holder.

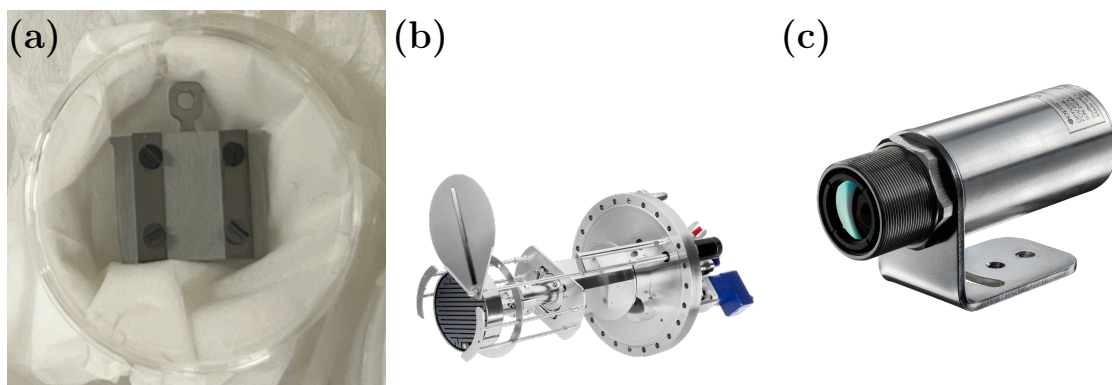


Figure 21 – (a) Standard flag-style model FP15 MBE sample holder made of tantalum. Panels (b) shows a manipulator, and (c) shows a IR optrisxi 640 thermal camera. Image (b) taken from [127], and (c) from [128].

Substrate temperature is crucial in epitaxial growth, being a critical parameter for the surface diffusion of atoms, which dictates the crystal quality of the grown material [122, 129]. The substrate temperature is controlled by the current intensity in a tungsten wire inside the manipulator platform, allowing for temperatures up to 1220 °C. The platform possesses a thermocouple³ near the substrate for temperature monitoring. However, the temperature in the substrate is not necessarily the same as in the manipulator, so an IR camera (see Figure 21 (c)) is used as a more accurate alternative for temperature measurement. This IR camera is positioned outside the deposition chamber, and measurements are taken through a glass viewport. The viewport is equipped inside the chamber with a pneumatically actuated shutter to protect the glass from material deposition. When correctly calibrated for the material's emissivity, an IR camera can achieve an accuracy of 2% for temperatures up to ≈ 2500 °C [130, 131], allowing the high precision temperature control of the substrate surface.

The effusion cell temperature regulates the flux rate of evaporated materials. Controlling this parameter is also essential for achieving high-quality MBE-grown thin films that follow the substrate orientation. The molecular flux dependence temperature follows the Arrhenius laws [132].

$$\Phi \propto \exp\left(-\frac{E_e}{k_B T}\right), \quad (3.1)$$

where E_e is an evaporation energy, k_B is Boltzmann's constant, and T the temperature. The effusion cells are also equipped with thermocouples working with PID controllers, allowing stable and finely tuned temperatures, and consequently, precise control of the molecular flux.

³A thermocouple operates based on the principle of the Seebeck effect

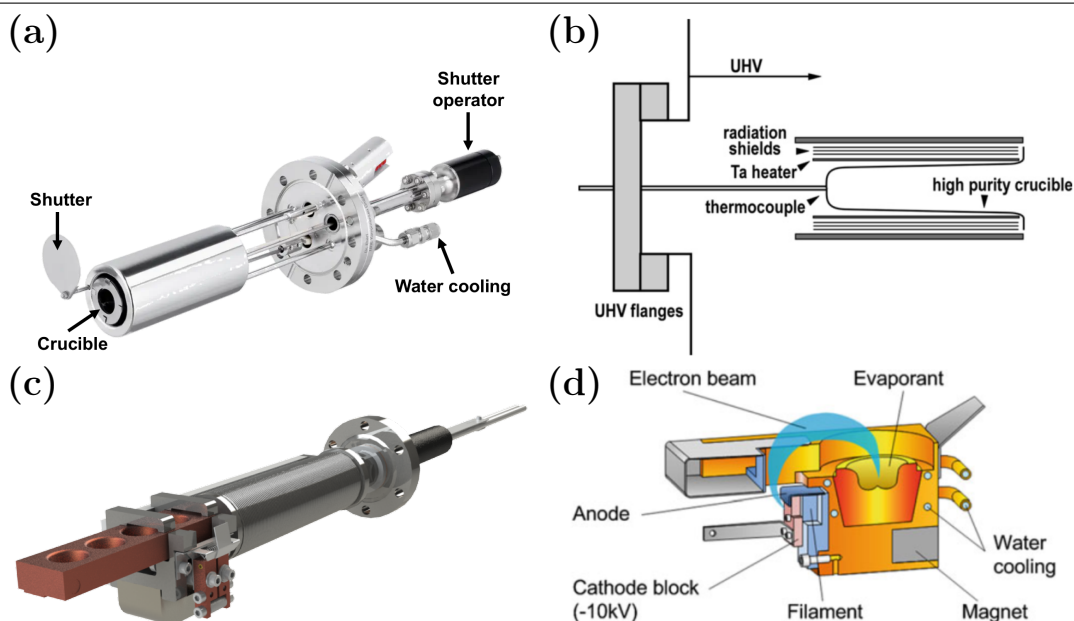


Figure 22 – Titan effusion cell and crucible used in the MBE system. (c) The model 539 is mounted on a 6-inch CF flange with 4cc pockets and no sweep coils. It is available in 3, 4, 5, and 6 pocket configurations and is ideal for research and development applications. (d) Bent beam electron gun with a water-cooled evaporant support. Image (a) adapted from [133], (b) from [129], and (d) from [134]. Image (c) taken from [135].

The molecular flux rates must be calibrated to allow control of the thickness of the grown sample through the deposition time. This calibration is usually performed using *in-situ* techniques such as a beam flux monitor (BFM) and a quartz crystal monitor (QCM), and *ex-situ* direct thickness measurements, such as step-height AFM.

The BFM is used to measure the flux of atoms or molecules that evaporate from the effusion cell before the flux reaches the substrate. The BFM is based on a Bayard-Alpert ionization measurement device, which is standard in UHV measurements [136, 137]. The BFM consists of a filament that emits electrons, a positively charged grid (anode), and a collector (cathode), as shown in Figure 23. The heated filament emits electrons that are accelerated toward the anode. Along the path, these electrons collide with the molecular beam (or atom beam) in the UHV, ionizing it and creating positive ions. These positive ions are then attracted to the collector, a wire kept at negative potential. The resulting current (I_+) is proportional to the neutral molecule density in the beam and is related to the pressure [138, 139].

The BFM can be positioned and retracted from the molecular beam path. The

measurement position is kept a few centimeters below the substrate, intercepting the molecular beams that would collide with the substrate. This allows measurement near the sample position. The measured (I_+) is then converted into a Beam Equivalent Pressure (BEP), which is relative to the molecular beam density. The BEP is useful to compare the flux rates between two different effusion cells in real time. As the BEP provides a relative measure, calibration with a direct thickness measurement is necessary for correct thickness determination.

Parasitic magnetic fields (from the e-beam or electron gun of RHEED) can interfere with the electrons inside the BFM and affect the readings. In this sense, a measurement technique that is not affected by magnetic fields, such as the QCM, can be used. The Quartz Crystal Microbalance (QCM) is an instrument that measures the real deposition rate and the total thickness of a film in real time with high precision. This equipment measures the deposited material mass at a short distance from the substrate position, acting as an extremely sensitive microbalance inside the deposition chamber. The QCM works by resonance and the piezoelectric effect, where a thin circular quartz crystal is used as a mass sensor. The quartz sensor vibrates at its natural resonance frequency when subjected to an AC signal. The crystal is cut at specific angles (AT-cut), which makes its natural resonance frequency stable, depending only on its own mechanical properties and thickness. To avoid frequency fluctuations caused by temperature changes, the crystal is also water-cooled in its holder. As material is deposited on the quartz surface by the molecular beam flux, its mass increases, causing a decrease in its natural resonance frequency.



Figure 23 – Beam Flux Monitor - Nude Bayard-Alpert Type Ionization Vacuum Pressure Sensor. Image taken from [139].

In fact, the deposited material does not have the same mechanical properties (density, viscosity, shear modulus, and Young's modulus) as the quartz crystal. This difference, called acoustic incompatibility, causes a loss of vibrational energy transferred from the vibrating quartz to the material, leading to a reading that does not perfectly match the real deposited mass. For that reason, an acoustic impedance factor correction must be applied. This correction, known as the Z-factor correction, can be determined empirically through a calibration process. The QCM registers the thickness change (d_{QCM}) based on the shift in the resonance frequency of the quartz, given by the Sauerbrey equation 3.2:

$$\Delta m = \frac{AN_q\rho_q}{\pi Z f_L} \tan^{-1} \left[Z \tan \left(\pi \frac{f_0 - f_L}{f_0} \right) \right] \quad (3.2)$$

where

$$Z = \sqrt{\left(\frac{\rho_q \mu_q}{\rho_f \mu_f} \right)}, \quad (3.3)$$

Δm is the mass change, A is the piezoelectrically active crystal area (g/cm^2), f_0 is the resonant frequency of the fundamental mode (Hz), f_L is the frequency of loaded crystal (Hz), N_q is the frequency constant for AT-cut quartz crystal ($1.668 \times 10^{13} \text{ Hz} \cdot \text{\AA}$), ρ_q is the density of quartz ($2.648 \text{ g}/\text{cm}^3$), ρ_f is the density of the film (g/cm^3), μ_q is the shear modulus of quartz ($2.947 \times 10^{11} \text{ g} \cdot \text{cm}^{-1} \text{s}^{-2}$), and μ_f is the shear modulus of the film ($\text{g} \cdot \text{cm}^{-1} \text{s}^{-2}$). This equation is used in the case $\Delta f/f > 0.05$ [140, 141, 142].

After the deposition, an *ex-situ* method such as AFM or profilometry is used to obtain the real thickness (d_{real}). The Z-factor is then empirically calculated as the ratio

between the real thickness and the thickness reported by the QCM, as follows:

$$Z = \frac{d_{\text{real}}}{d_{\text{QCM}}}. \quad (3.4)$$

The correct deposition rate is then estimated by

$$R_{\text{corrected}} = R_{\text{sauerbrey}} \cdot Z. \quad (3.5)$$

For rigid materials (W, Ta, Mo, Ti) $Z \approx 1$, for soft metals with low boiling points (Pb, In, Sn) $Z > 1$, and for polymers and other organic materials $Z \gg 1$. Due to the accumulation of material on the quartz, the QCM sensor must be regularly replaced and calibrated to ensure correct deposition rate estimation.

3.2 Characterization and experimental techniques

3.2.1 X-ray Diffraction

The X-ray diffraction (XRD) technique is a useful method for analyzing the atomic ordering of materials in liquid, powder, and crystalline forms. An XRD spectrum is obtained from the coherent scattering of an incident X-ray beam by the atomic arrangement of the material, see Figure 24 (a). The interaction is described by

$$n\lambda = 2d_{hkl} \sin \theta, \quad (3.6)$$

where n is an integer, d_{hkl} is the interatomic spacing where the hkl are the Miller index, and λ is the X-ray wavelength. This relation is known as Bragg's equation and connects the interplanar spacing of a crystalline lattice with the angles at which constructive interference of scattered X-rays occurs [143, 144]. To perform this type of measurement, the electromagnetic wavelength must be comparable to the spacing between atomic planes [143]. Figure 24 (b) illustrates the diffraction geometry, where beam 2 travels an additional path length of $2d \sin \theta$ relative to beam 1. Constructive interference occurs when this path difference is equal to an integer multiple of the X-ray wavelength, $n\lambda$, determined by the radiation source [144, 143].

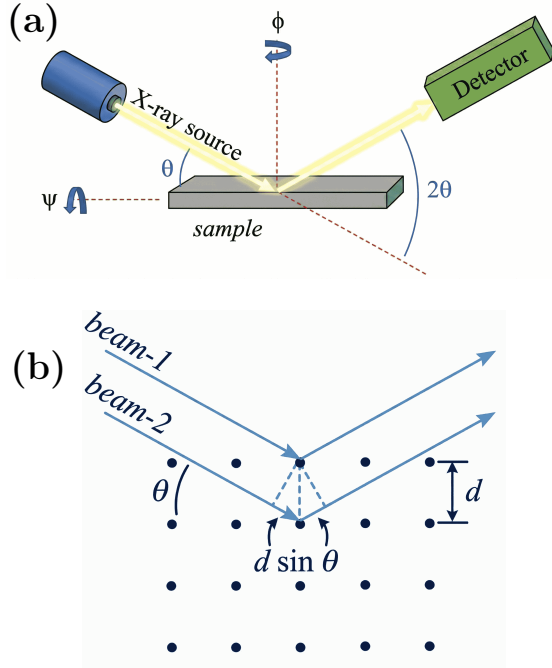


Figure 24 – Illustrations of (a) the Bragg geometry and its angular degrees of freedom and (b) Bragg reflections, with the sample represented as a set of lattice points.

With the three degrees of freedom of this configuration (Ψ , Φ , and θ), it is possible to define the diffracted wave vectors, whose components can be associated with deformation (strain) and inclination (tilt).

The high-resolution X-ray diffraction (HRXRD) is one of the most fundamental and accurate technique used in the determining the crystalline quality of MBE grown films. This experimental technique requires high precision in the determination of lattice parameters and peak profiles, which are essential for epitaxial and heterostructural analyses. To achieve such resolution (on the order of $\Delta d/d \approx 10^{-4}$ to 10^{-5}), the use of a crystal monochromator (usually Ge) and a crystal analyzer is fundamental for reducing the angular divergence and spectral dispersion of the incident beam [143, 144].

Bragg's law is a necessary condition that must be satisfied to place the detector at a position where constructive interference can occur. However, it is not sufficient to ensure the presence of a diffraction maximum, since the diffracted intensity must also be considered. The intensity of the diffracted beam depends on the atomic arrangement within the unit cell and is described by the structure factor

$$F_{hkl} = \sum_{n=1}^N f_n e^{2\pi i(hx_n + ky_n + lz_n)}, \quad (3.7)$$

where f_n is the atomic scattering factor, which represents the ability of an atom to scatter X-rays, and x_n , y_n , and z_n are the fractional coordinates of the n th atom within the

unit cell. In this sense, a diffraction peak is observed only if the structure factor at the corresponding Bragg position is nonzero [143, 144].

In HRXRD measurements, it is common to describe the diffraction phenomenon using reciprocal space. In this sense, it is convenient to express Bragg's law in vector notation. The incident and diffracted X-ray beams, together with the surface normal, define a plane. Within this framework, X-ray diffraction can be represented in terms of the incident wave vector $\mathbf{K}_0$ and the diffracted wave vector $\mathbf{K}$. The angle between the directions of the wave vectors $\mathbf{K}_0$ and $\mathbf{K}$ is equal to 2θ , as shown in Figure 25 (b). Considering a fully elastic scattering process, one has $|\mathbf{K}| = |\mathbf{K}_0| = k$. The scattering vector $\mathbf{q}$, defined as the difference between the incident and diffracted wave vectors, has a magnitude given by:

$$|\mathbf{q}| = |\mathbf{K} - \mathbf{K}_0| = 2k \sin \theta, \quad (3.8)$$

considering the wave vector magnitude $k = 1/\lambda$ and using Bragg's equation, one obtains

$$|\mathbf{q}| = \frac{1}{d_{hkl}}. \quad (3.9)$$

This corresponds to the definition of the modulus of the reciprocal space vector $|\mathbf{G}_{hkl}|$, which is associated with the (hkl) planes of a crystal. In this sense, the vector relation can be written as

$$\mathbf{K} - \mathbf{K}_0 = \mathbf{G}_{hkl}. \quad (3.10)$$

When constructive interference occurs, the scattering vector $\mathbf{G}_{hkl}$ terminates at a point of the reciprocal-space of the crystal, as shown in Figure 25 (a).

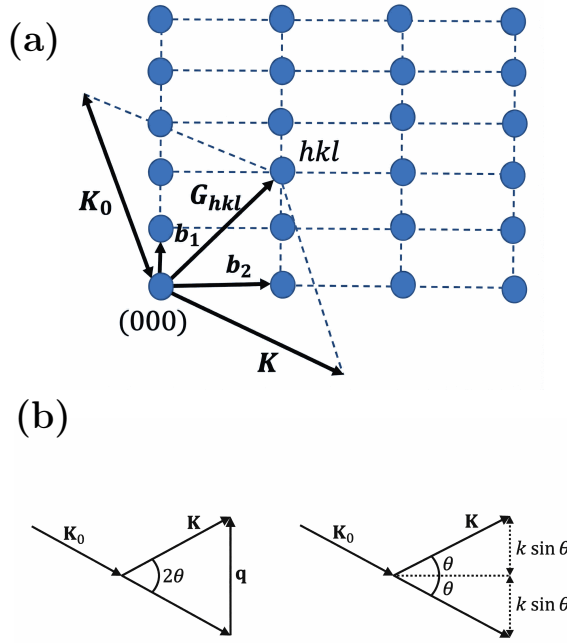


Figure 25 – Panel (a) shows a reciprocal-space diffraction condition. The scattering vector must terminate at a point of the reciprocal-space in order for Bragg's law to be satisfied and a diffraction peak to be observed. Panel (b) shows the scattering vector $\vec{q} = \vec{K} - \vec{K}_0 = \vec{G}_{hkl}$, where $\vec{K}_0$ points in the incident direction and $\vec{K}$ in the scattered direction. Also the trigonometric relation used to determine the magnitude of the scattering vector is shown.

One of the most fundamental XRD techniques is the θ - 2θ scan (or $2\theta - \omega$ in some references), in which the motion of the sample and the detector are synchronized. While the sample rotates by an angle θ with respect to the incident beam, the detector rotates by an angle 2θ . This configuration ensures that the detector always intercepts the diffracted beam, with the diffraction angle being equal to the incidence angle. Figure 24 (a) illustrates the measurement procedure.

From the reciprocal space point of view, the θ - 2θ measurement changes only the modulus of the scattering vector $\vec{G}_{hkl}$, while keeping its direction fixed, as shown by the red-dotted line in Figure 26. This measurement detects variations in the interplanar spacing (d), allowing the determination of crystallographic parameters such as the out-of-plane lattice parameters and the strain level of an epitaxial layer. A large broadening in a θ - 2θ scan indicates a larger variation in the interplanar distances, suggesting a low coherence between planes parallel to the substrate surface.

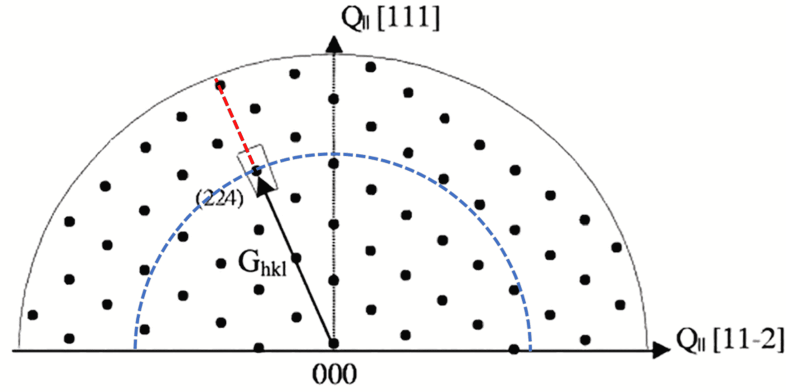


Figure 26 – Schematic representation of the reciprocal lattice points of a (111) oriented sample showing a map around the (224) reciprocal lattice point.

Another measurement method widely used for epitaxial single-crystal films is the rocking curve. Through this measurement, it is possible to evaluate the crystalline quality and the degree of perfection of the in-plane unit-cell orientation. In this measurement configuration, the detector is kept fixed at an angle that satisfies the Bragg condition and the structure factor, so that a diffraction peak is observed at this position [144, 143]. Then, the sample is rocked by small angles θ (according to the illustration in Figure 24 (a)). A broadened peak in a rocking-curve measurement indicates that the unit cells, or even crystallites, present a high degree of in-plane misalignment with respect to each other. This effect is known as mosaicity [144, 143].

In reciprocal space, the rocking-curve measurement corresponds to a variation only in the direction of the scattering vector, as indicated by the blue dotted arc in Figure 24 (b). The main quantitative metric extracted from this curve is the FWHM where the value is proportional to the defect density in the sample.

The equipment used in this work was a Bruker D8-Discover configured for high-resolution X-ray diffraction (HRXRD) measurements. The X-ray beam was generated by a copper source, and the Cu $K\alpha_1$ line with $\lambda = 1.54056 \text{ \AA}$ was used. Before reaching the sample (primary beam), a Göbel mirror was employed to focus the beam from the source. A high-perfection double-crystal germanium monochromator was used. This configuration ensures a spectral bandwidth of $\Delta\lambda/\lambda \approx 10^{-4}$ and guarantees that only the Cu $K\alpha_1$ line interacts with the sample. After interacting with the sample, the diffracted beam passes through motorized slits with an opening of 0.1 mm providing an angular resolution of

0.04°. Subsequently a nickel filter is used to block the K_{β} line. Figure 27 shows a XRD setup configured in the high resolution mode.

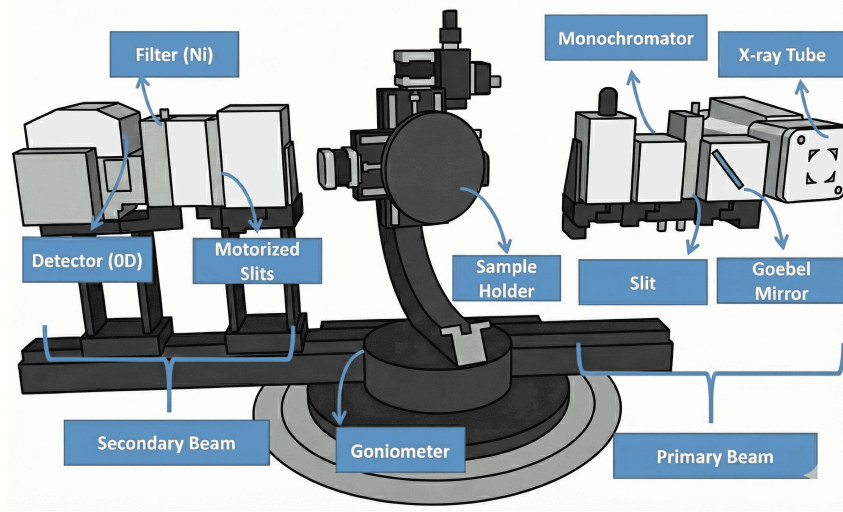


Figure 27 – High resolution XRD setup.

3.2.2 Reflection High-Energy Electron Diffraction

Reflection High-Energy Electron Diffraction (RHEED) is a technique sensitive to the surface uniformity that uses a high-energy electron beam, typically in the range from 10 to 100 keV, which interacts with the sample at a grazing angle, usually between 1 and 4 degrees [145]. Due to this geometry, the penetration depth of the electron beam is limited to only a few atomic layers, ensuring high surface sensitivity. This setup allows its simultaneous use during MBE growth, since the electron beam does not block the evaporation flux. Moreover, an ultra-high vacuum (UHV) environment is desirable to increase the electron mean free path.

Due to the de Broglie relation, an electron beam can be described as a wave, with its wavelength related to the electron kinetic energy. For non-relativistic electrons⁴, this relation can be written as

$$\lambda(\text{\AA}) \approx \sqrt{\frac{150.4}{E(\text{eV})}}, \quad (3.11)$$

where E is the electron energy. This expression shows that high-energy electrons have very short wavelengths[145, 146]. In this work, $\lambda \approx 0.10013 \text{ \AA}$ was achieved using an electron energy of 15 keV. For comparison, HRXRD uses $\lambda = 1.54056 \text{ \AA}$.

⁴Relativistic effect must be considered when the electron energy is higher than 50 keV. Usually this energy is about 20 keV.

In the kinematical theory, the Laue condition states that diffraction occurs when the difference between the scattered wave vector and the incident wave vector is equal to a reciprocal space vector (ending in a dot in the reciprocal space), as discussed previously in Equation 3.10 on **High Resolution X-ray Diffraction** section. The diffraction occurs only in the surfacial layers and, the system is therefore treated as two-dimensional. The reciprocal space is usually formed by dots when related to three-dimensional diffraction in bulk materials. In two-dimensional systems, due to the breaking of translational symmetry along the z direction (perpendicular to the sample surface), the Fourier transformation produces dots in the xy plane but extended rods along the z direction [145, 146]. Figure 28 shows blue rods, which are a representation of the reciprocal space of a two-dimensional system.

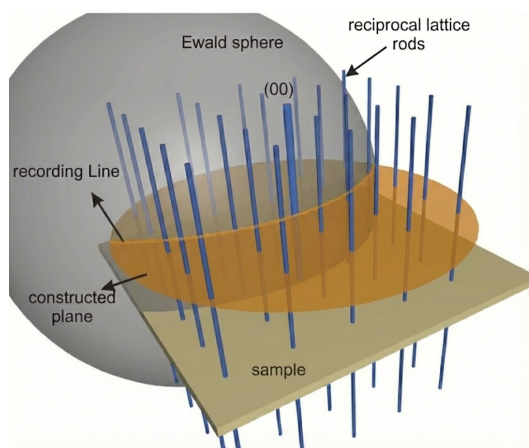


Figure 28 – Schematic representation of the two-dimensional reciprocal lattice and the Ewald sphere. The intersection between the sphere and the reciprocal lattice rods along the recording line corresponds to what is visualized in the RHEED pattern.

The streaks pattern is a diffraction pattern observed on a fluorescent screen, corresponding to the projection of the intersection between the Ewald sphere and the reciprocal lattice rods, as labeled as "recording line" in Figure 28. Since the radius of the Ewald sphere is very large and the incidence occurs at a grazing angle, the sphere intersects the rods along their length, also producing streaks rather than perfect dots. The asymmetric shape of a streak is due to irregularities in the sample surface, the finite angular dispersion inherent to the equipment, and the not perfect scattering of the electron beam[145, 146]. Those effects cause the distortions of the reciprocal rods as shown in Figure 29. Details about the equipment used in this work were given in **SnTe Sample Growth by MBE** section.

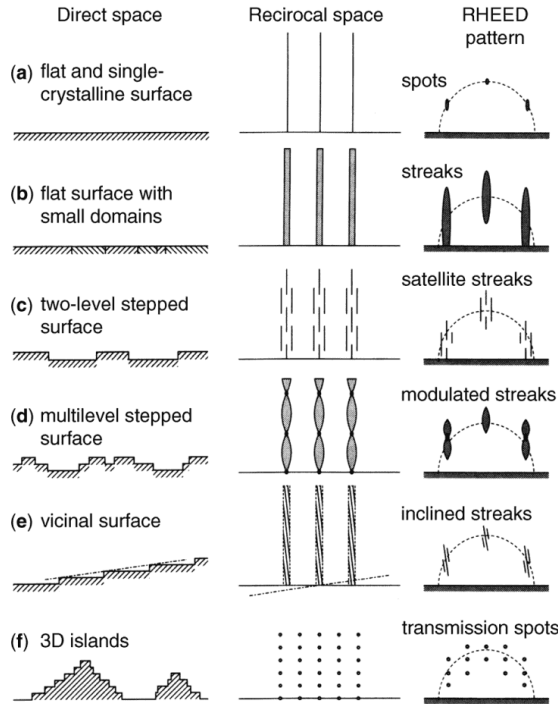


Figure 29 – Schematics of different types of realistic surfaces, showing their real space morphology, the corresponding reciprocal space representation, and the resulting RHEED patterns. Image taken from [146].

3.2.3 Atomic Force Microscope

Atomic force microscopy (AFM) is a widely employed surface characterization technique in the fields of engineering and physics. Its primary advantage lies in its ability to produce nanometric resolution topographical images of material surfaces. The high spatial resolution, operational simplicity, and minimal sample preparation requirements are key factors contributing to the broad applicability of this technique [147, 148, 149]. Based on the data acquired through AFM, parameters such as surface roughness, grain distribution, real surface area, and thin-film thickness can be quantitatively evaluated. Unlike electron microscopy techniques, AFM does not rely on lenses or electron beams and can be operated under ambient temperature and pressure conditions, making it a versatile and non-destructive method for analyzing a wide range of materials.

The operating principle of Atomic Force Microscopy (AFM) is based on scanning the surface of a sample using a highly sharp tip attached to a cantilever. The cantilever is typically made of silicon, and its tip is sharp consisting of only a few atoms, as illustrated in Figure 30 (a). The sample is placed on a piezoelectric stage that allows precise movement in the x, y, and z directions.

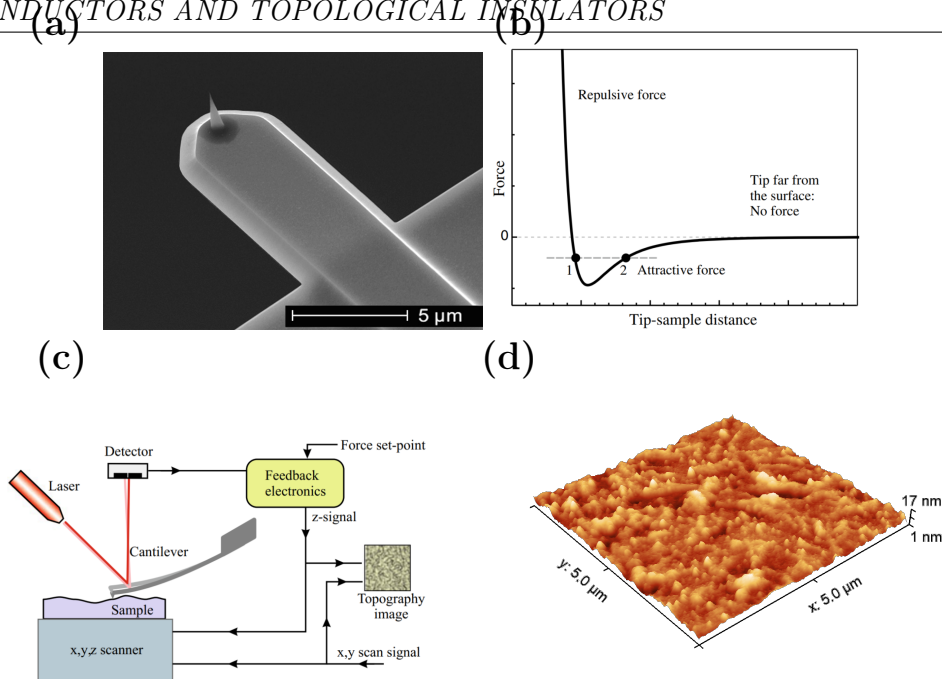


Figure 30 – Scanning electron microscopy (SEM) image (a) showing the AFM tip attached to the cantilever. (b) Qualitative representation of the force versus distance relationship between the tip and the sample. (c) Diagram illustrating the operation of AFM in contact mode. (d) Topographic AFM image of the PEDOT:PSS film surface, highlighting its surface morphology. Image (a) taken from [150], (b) and (c) from [149].

In AFM, the force between the tip and the sample is measured. The short tip-sample distance, usually 1-2 nm, enables atomic-scale interactions, allowing the detection of electrical forces between atoms of both surfaces, which leads to cantilever deflection [147, 148, 149]. Figure 30 (a) illustrates the tip-sample force as a function of distance, where repulsive, attractive and zero force are identified [149].

At very short distances, the tip-sample interaction is repulsive and at large distances the force becomes negligible. However, in the intermediate region between these two regimes, the interaction force is attractive. Point 2 in Figure 30 (b) represents the optimal operating condition, as it lies within a region where the force-distance relationship is monotonic and stable.

The tip-sample force is proportional to the cantilever deflection and is described by Hooke's law, $F_{cantilever} = -kz$ where k is the cantilever stiffness and z is the cantilever deflection relative to its equilibrium position. A laser beam reflected from the back side of the cantilever changes its direction according to the cantilever's deflection, as shown in

Figure 30 (c). This change in laser direction is detected by a four-quadrant photodiode, and the signal is processed by a microcomputer to convert it into x, y, and z positions, reconstructing a three-dimensional image of the sample surface.

The most conventional operating modes used in AFM are contact mode and tapping mode. In contact mode, the surface of the sample is mapped while the z-position is adjusted continuously to maintain a constant tip-sample force. The z-displacements applied by the piezoelectric scanner are recorded as the topographic signal. In this mode, the outermost atoms of the tip remain in continuous contact with the sample, operating in the repulsive interaction regime. Contact mode is best suited for imaging rigid, low-roughness samples, such as metals and semiconductors, and provides high-resolution images. However, the tip lifetime may be reduced due to wear caused by friction during scanning.

In tapping mode, the cantilever is driven to oscillate at its free resonance frequency. As the tip approaches the sample surface, the cantilever's resonance is altered, resulting in a change in oscillation amplitude. This amplitude variation is used both as a detection signal for z-deviation reconstruction and as a feedback signal to regulate the tip-sample distance. Tapping mode significantly reduces friction and sample damage, making it best suited for delicate samples such as polymers, biological specimens, and soft thin films. However, the calibration procedure is more sensitive and requires careful adjustment. Figure 31 presents a schematic illustration of the AFM operation mode [151].

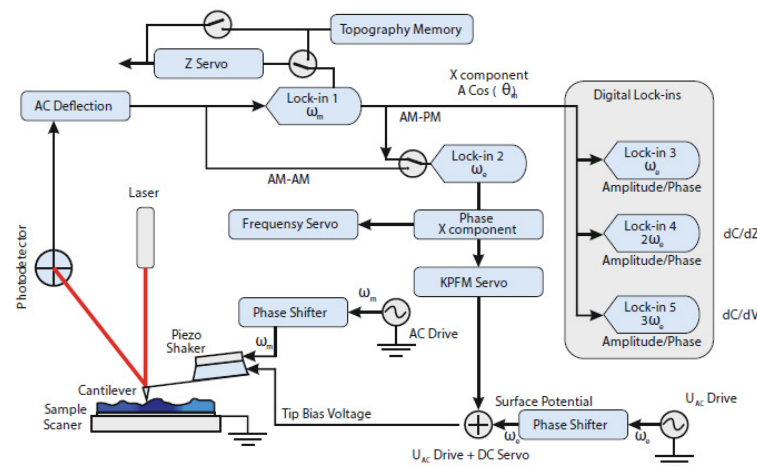


Figure 31 – Schematic diagram of the atomic force microscope (AFM) operating with lock-in detection. Image taken from [151].

3.2.4 Raman Spectroscopy

Raman spectroscopy is a non-destructive analytical technique used to obtain spectra related to the natural vibrational modes of molecules of a material. This technique is named after the Indian physicist *Chandrasekhara Venkata Raman*, who first observed it in 1928 [152]. The set of vibrational modes, which are associated with the atomic bonds within a material, forms a kind of vibrational "fingerprint." This "fingerprint" can be used both for the precise identification of a material and for the investigation of structural changes, such as those induced by the doping process.

In Raman scattering, a monochromatic, collimated, and coherent light beam generated by a laser is directed onto the sample to be analyzed. The incident light is scattered by the molecules of the material, such that the molecular electron cloud is excited by the oscillating electromagnetic field of the photon. The interaction between the photon and the molecule occurs over an extremely short time interval, which is characterized as the molecule's virtual state [153]. This state is unstable, and the photon is immediately re-emitted as scattered light. As a result of the interaction, the molecule may remain in a final energy state different from the initial one, due to the exchange of energy with the incident photon.

In most interactions between photons and molecules, their energies remain unchanged. Thus, both the photon and the molecule retain the same energy before and after the interaction. This phenomenon, classified as an elastic scattering process, is known as Rayleigh scattering and accounts for the majority of observed scattering events, see Figure 32 (a).

Raman scattering, on the other hand, is an inelastic process in which energy is exchanged between the scattered photon and the molecule. This phenomenon is much less frequent, occurring approximately once every 10^6 to 10^8 interactions with incident photons [153]. When the scattered photon loses energy to the molecule, the process is referred to as Stokes Raman scattering. In the opposite case, where the molecule transfers energy to the photon (transitioning to a lower-energy state), the process is known as Anti-Stokes Raman scattering. Figure 32 (a) and (b) illustrates these three distinct types of scattering. In principle, both Raman processes would be equally probable, however, in a collection of molecules, most are found in the ground state. Therefore, Stokes scattering is statistically

more likely.

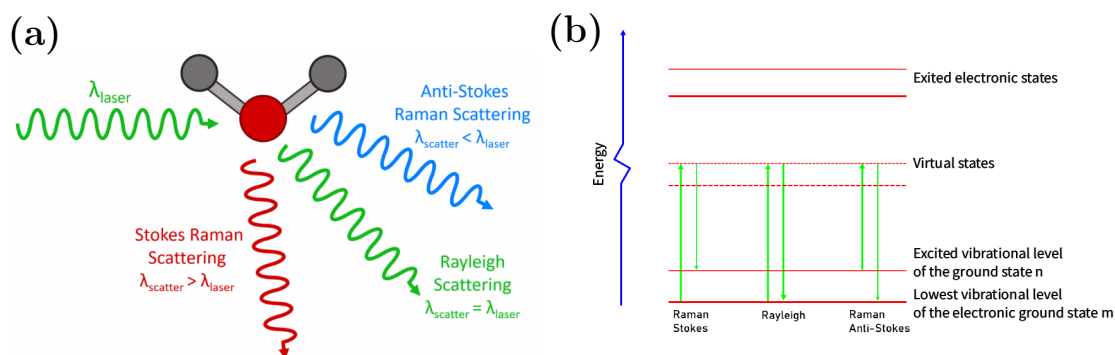


Figure 32 – Possible light scattering processes by a molecule. In Stokes Raman scattering, the scattered photon has lower energy than the incident photon. In Anti-Stokes Raman scattering, the scattered photon has higher energy. In Rayleigh scattering, the scattered photon retains the same energy as the incident photon. (b) Diagram of Raman and Rayleigh scattering processes. The lowest energy state, m , represents the fundamental vibrational level. Just above it, state n corresponds to the first excited vibrational state, differing from m by one vibrational energy unit. The upward arrows indicate the excitation energy, while the downward arrows represent the energy of the scattered photon. Image (a) adapted from [154].

The virtual energy level is associated with the vibrational modes of the molecules that make up the material and differs from the quantized electronic energy levels. In general, the virtual level has lower energy than the first electronically excited state, as illustrated in Figure 32 (b).

In the Raman spectroscopy measurement procedure, the scattered light is directed into a spectrograph, which disperses it into a spectrum through light refraction. This spectrum is then analyzed by a CCD (Charge Coupled Device) detector, enabling the construction of a graph of vibrational intensity as a function of the wavenumber (*Raman Shift*). The *Raman Shift* is the difference between the wavenumber of the incident laser and that of the scattered light. Peaks in the Raman spectrum correspond to different vibrational modes within the molecular bonds of the material. Each substance has its own vibrational "fingerprint" composed of these characteristic modes. Raman spectroscopy allows access to the normal vibrational modes and, subsequently, to the molecular structure, when analyzed through group theory [155]. Figure 50 shows an example of Raman spectroscopy of PEDOT:PSS showing its characteristic peaks.

Through Raman spectroscopy, it is possible to obtain information about the electronic state of a given material, identifying whether it is an insulator, semiconductor,

or conductor. This result can be achieved through detailed analysis of the individual peaks obtained via spectral deconvolution of the Raman spectrum. In the deconvolution process, the positions of the peaks associated with the Raman shift are analyzed separately, allowing for the identification of the material's phase. Additionally, it is possible to indirectly estimate the thickness of thin films based on the acquired spectral features.

3.2.5 Uv-Vis Spectroscopy

Spectrophotometry is a non-destructive optical characterization technique in which a monochromatic electromagnetic wave interacts with the material of interest, and the reflected or transmitted light is collected and analyzed. This technique is based on the principle that each material absorbs or transmits light differently depending on its electronic structure and composition. By measuring the intensity of the transmitted or reflected light as a function of wavelength, it is possible to determine the absorbance or transmittance spectra, which provide information about the optical properties such as optical band gap energy, refractive index, absorption coefficient, and electronic transitions of a material [156, 157, 158, 158, 159, 160].

The spectrophotometry technique can cover a wide range of the electromagnetic spectrum. However, for each spectral region, there are commercial spectrometers specifically optimized to operate within that range. The name of the spectral region usually defines the designation of both the technique and the corresponding instrument, such as UV-Vis (175-750 nm), near-infrared (NIR, 0.75-2.5 μm), mid-infrared (MIR, 2.5-50 μm), far-infrared (FIR, 50-1000 μm), and terahertz (THz, 1-10 mm) ranges⁵. Therefore, the most appropriate instrument must be selected according to the intended investigation [156, 157, 158, 158, 159, 160].

The UV-Vis absorption spectroscopy covers only a narrow region of the frequency or wavenumber spectrum. However, this range is extremely important because the corresponding energy differences correspond to the electronic transitions between quantized energy levels of atoms and molecules. UV-Vis absorption spectra describe the behavior of electronic transitions between quantized energy levels at different wavelengths, where an electron in the ground state absorbs a photon with a specific energy as illustrated in

⁵It is worth noting that the UV-Vis frequency range lies in the petahertz (PHz) region.

Figure 33 [161].

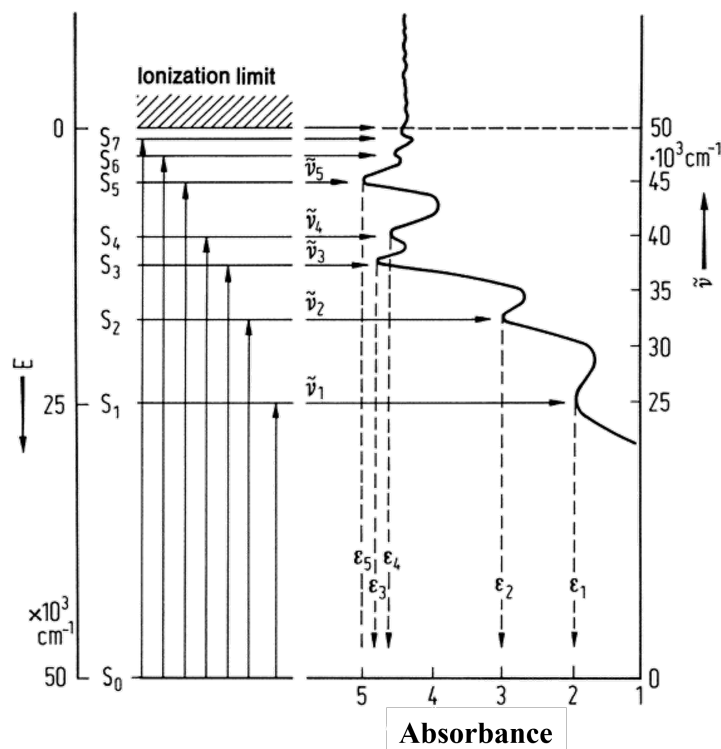


Figure 33 – Energy-level diagram illustrating singlet-singlet electronic transitions and their correspondence with absorption bands. The S_i assignments represent the quantized energy levels and $\tilde{\nu}_i$ is its corresponding wavenumber. The ϵ_i corresponds to the peak of absorbance of each transition. Image adapted from [161].

The basis of optical spectroscopy, and consequently of Uv-Vis spectra, lies on the Bohr-Einstein frequency relationship. The discrete atomic (or molecular) energy levels E_i is related with the frequency of the electromagnetic radiation ν as the following

$$\Delta E = E_2 - E_1 = h\nu,$$

where h is the Planck constant ($h = 6.626 \times 10^{-34}$ J·s) and ν the frequency of light. An electronic transition occurs when an electron goes from a discrete energy level to another one by absorbing or emitting a photon ⁶. The energy band gap (E_g) is an important parameter, as it defines the amount of energy required for electrons to jump from the HOMO to the LUMO, as depicted in Figure 34.

⁶In some cases, a phonon can participate in this process to satisfy momentum conservation.

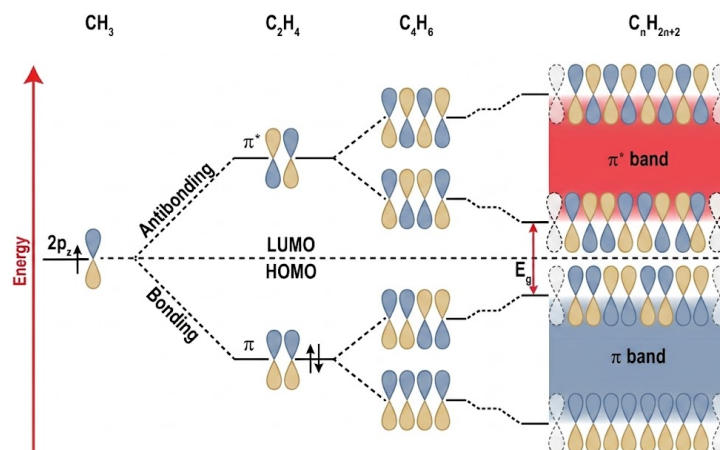


Figure 34 – Schematic representation of the formation of the π and π^* bands in conjugated polymers arising from the interaction between π orbitals of successive molecular units. A π bond is a type of covalent bond formed by the lateral overlap of two parallel atomic orbitals, usually p orbitals. Image extracted from [162].

In practical terms, the electronic transitions manifest as changes in the intensity of light transmitted through a material when it is exposed to electromagnetic radiation. The interaction between light and matter can thus be quantified by comparing the intensity of the incident radiation (I_0) with the transmitted radiation (I). This relationship provides the experimental basis for defining the absorbance (A) and transmittance (T) of a sample, parameters that describe how efficiently the material absorbs photons at each wavelength, or in other words, how transparent a material is at each wavelength.

The light absorption measurements are described by the Bouguer-Lambert-Beer law in the UV-VIS and IR-region:

$$A = -\log T = \log \frac{I_0}{I} = \varepsilon bc, \quad (3.12)$$

where T is the transmittance, I_0 is the intensity of the light arriving to the sample, I is the intensity of this light emerging from the sample, ε is the molar absorptivity, b is the path length of sample, and c is the concentration of absorber. The UV-Vis spectrophotometer scheme is shown in Figure 35. To measure the T , the incident light from the xenon lamp is passed through the sample cell. The transmitted light intensity through the sample (I) is compared with the transmitted light intensity through the reference cell (I_0). The transmittance is then calculated as $T = I/I_0$, and the transmittance percentage (T) as $T \times 100\%$. With this data, the absorbance is obtained by Equation 3.12.

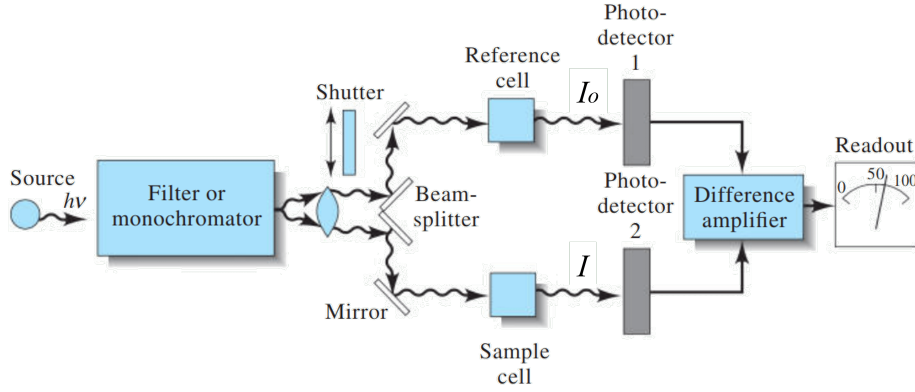


Figure 35 – A double-beam in space spectrophotometer illustration, in which the radiation emerging from the filter or monochromator is divided into two beams that simultaneously pass through the reference and sample cells before reaching two matched photodetectors. Image adapted from [163].

More details on the specific steps and configurations used to obtain the absorbance spectra of PEDOT:PSS films are provided in the **Absorbance of PEDOT:PSS films** section.

3.2.6 Ferromagnetic Resonance in Thin Films

One atomic magnetic moment has its motion described by the classical equations of mechanics and electromagnetism. Considering an infinite magnetic material without boundaries under the influence of a static magnetic induction $\vec{B}$, the interaction energy between its magnetic moment $\vec{\mu}_i$ and $\vec{B}$ is given by

$$U_i = -\vec{\mu}_i \cdot \vec{B}.$$

The torque produced by $\vec{B}$ acting on $\vec{\mu}_i$ when the magnetic moment is not parallel to the magnetic induction is given by

$$\vec{\tau} = \vec{\mu}_i \times \vec{B}.$$

The magnetic moment is related to its angular momentum by

$$\vec{\mu} = -g\mu_B\vec{J}.$$

By applying Newton's law and recalling that the atomic magnetic moment has an angular momentum $\hbar\vec{J}$, we obtain

$$\frac{d\vec{\mu}_i}{dt} = -\gamma\vec{\mu}_i \times \vec{B}, \quad (3.13)$$

where

$$\gamma = \frac{g\mu_B}{\hbar}$$

is the atomic gyromagnetic ratio, with g being the Landé g -factor, μ_B the Bohr magneton, and $\hbar$ the reduced Planck constant. Writing Equation 3.13 as a function of the magnetization vector $\vec{M}$, one obtains

$$\frac{d\vec{M}}{dt} = -\gamma\mu_0\vec{M} \times \vec{H}.$$

This is the equation of motion for the magnetization under an external magnetic field in the absence of magnetic damping. This equation, known as the Landau–Lifshitz equation, shows that a external magnetic perturbation induces a precession of the magnetization around the direction of the external magnetic field, with a precession frequency given by

$$\omega_0 = \gamma\mu_0 H.$$

When a magnetic material is subjected to an oscillating magnetic field in the radio-frequency range, its magnetic resonance frequency is given by this equation. Note that the resonance frequency depends linearly on the external magnetic field ⁷, with the gyromagnetic ratio acting as the proportionality constant.

In a real physical experiment, the magnetization precesses in spirals around the external field, with a decreasing in angular displacement until $\vec{M}$ becomes parallel to $\vec{H}$. The time required for this process is known as the relaxation time and depends on the material properties. Figure 36 illustrates the direction of the vector $\vec{T}_d$, which is responsible for the reduction of the precessional radius.

The physical origin of this relaxation is related to the structure of the magnetic material and spin interactions. However, the real magnetization dynamics can be described without a detailed microscopic treatment by introducing a phenomenological torque term that drives $\vec{M}$ toward its equilibrium direction, as follows:

$$\frac{d\vec{M}}{dt} = -\gamma\mu_0\vec{M} \times \vec{H}_{\text{eff}} + \frac{\alpha}{M}\vec{M} \times \frac{d\vec{M}}{dt}. \quad (3.14)$$

This is the Landau–Lifshitz–Gilbert (LLG) equation, where the phenomenological parameter α was introduced by Gilbert [164]. The term $\vec{H}_{\text{eff}}$ represents the effective magnetic field

⁷This equation is a simplification that does not take boundary effects into account.

acting on the magnetization. This equation is widely used to describe the magnetization dynamics in bulk metallic ferromagnets. Figure 36 shows the vector directions for a precessing magnetic moment $\mathbf{m}$.

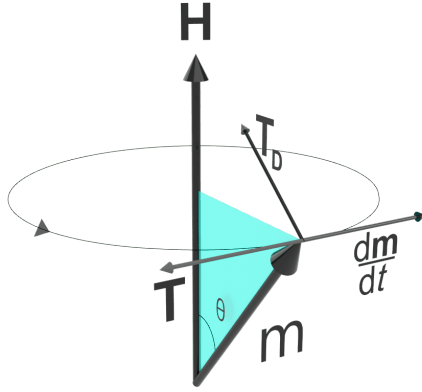


Figure 36 – Magnetic moment ($\mathbf{m}$) precession around the effective magnetic field $\mathbf{H}$. The vector $\mathbf{T}$ represents the damping torque.

When a magnetic material is subjected to a microwave driving magnetic field with frequency ω and orthogonal to $\vec{H}$, the damping parameter can be related to the linewidth of the FMR lineshape by

$$\Delta H = \frac{\alpha \omega_0}{\gamma}.$$

This relation provides a direct way to estimate the Gilbert damping parameter.

When a magnetic thin film is in the resonance regime, under the influence of demagnetizing effects generated by the sample boundaries, and considering the exchange interaction between spins, the resonance frequency ω_0 is related to the resonance magnetic field H_0 by [28]

$$\omega_0 = \gamma [H_0 (H_0 + 4\pi M)]^{1/2}.$$

This expression is known as the Kittel equation and corresponds to the uniform mode resonance frequency, where the magnetization is spatially uniform throughout the material. The specific form of the Kittel equation depends on the sample geometry and on the orientation of the external magnetic field.

3.2.6.1 Experimental Measurements

A ferromagnetic FMR experiment consists of analyzing the magnetization dynamics of a magnetic sample subjected to a radio frequency (rf) magnetic field applied

perpendicular to an external static magnetic field. Usually, FMR experiments are performed at a fixed microwave frequency while sweeping the intensity of the static magnetic field. In this configuration, the use of a lock-in amplifier provides a significantly higher signal-to-noise ratio. Alternatively, experiments can also be performed by keeping the static magnetic field intensity fixed while varying the excitation frequency.

In the FMR setup, a microwave signal with a frequency of 9.4 GHz is applied to a resonant cavity to generate the *rf* magnetic field. The microwave signal is guided from the generator to the cavity through a microwave circulator, which is used to block signal reflections and ensure unidirectional propagation from the generator to the cavity and subsequently to the detection system (Lock-in amplifier). Immediately before the lock-in amplifier, a rectifier circuit is connected in series with the coaxial transmission line, this circuit converts the high frequency microwave signal (after interaction with the sample inside the resonant cavity) from an AC signal into a DC signal, allowing the detection by the lock-in amplifier. Figure 37 shows an illustration of the SP-FMR experimental apparatus.

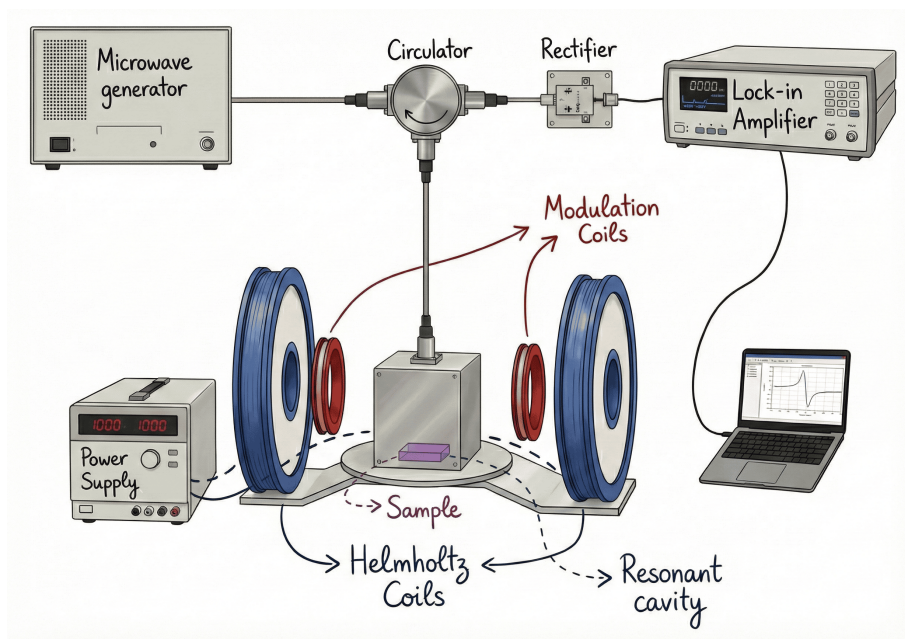


Figure 37 – Spin pumping FMR experimental setup representation. The connections between the microwave generator, circulator, and resonant cavity are made using a hollow waveguide, while other rf connections are typically performed using coaxial cables.

In this setup, the static magnetic field ($\vec{H}$) is generated by a pair of Helmholtz coils. A low-frequency modulation magnetic field, with a frequency of 1.2 kHz, is superimposed

on $\vec{H}$ using an additional modulation coil. This modulation causes a small oscillation of the static magnetic field around its nominal value. The oscillating magnetic field serves as an auxiliary technique to convert the DC absorption signal into an AC signal, which is essential for detection by the lock-in amplifier, since this instrument is sensitive only to AC signals.

The lock-in amplifier is configured to operate at the same frequency (1.2 kHz) as the modulation field, ensuring that only the signal component synchronized with the modulation is detected. This procedure significantly reduces noise contributions outside the modulation frequency.

Inside the resonant cavity, the sample is positioned at a location where the microwave magnetic field is maximized and the electric field is minimized. This positioning is crucial for FMR measurements, as it reduces spurious effects associated with electric-field-induced phenomena and ensures that the measured signal originates predominantly from magnetic interactions.

Additionally, the microwave magnetic field ($\vec{H}_{rf}$) at 9.4 GHz induces the precessional motion of the magnetic moments in the film. Since the resonance frequency depends on the value of the static magnetic field H_{dc} , the small modulation of H_{dc} also produces a slight modulation of the resonance condition, enabling the detection of the field-derivative FMR signal.

The static magnetic field ($\vec{H}$) and the microwave magnetic field ($\vec{H}_{rf}$) are applied in the plane of the film and oriented perpendicular to each other. This configuration, combined with appropriate values of the magnetic field magnitude and microwave frequency, allows the excitation of spin waves in the magnetic film, leading to the resonance condition. Under these conditions, the detected FMR signal corresponds to the derivative of the microwave power absorption with respect to the static magnetic field intensity, which is the quantity effectively measured by the lock-in amplifier. Figure 38 illustrates a typical FMR measurement of permalloy grown by molecular beam epitaxy at room temperature.

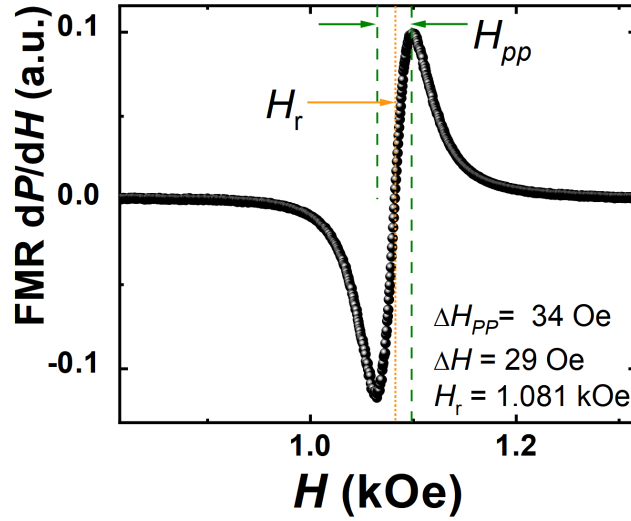


Figure 38 – Field-derivative FMR absorption spectrum of a permalloy film (20 nm) measured at a frequency of 9.4 GHz.

The linewidth (ΔH) and the peak-to-peak distance (ΔH_{pp}) were previously measured and are shown in the inset. The resonance field H_r corresponds to the field value at which the derivative of the absorption changes sign, indicating maximum absorption. From the relationship between the linewidth, the Gilbert damping parameter, and the susceptibility tensor, it is possible to obtain the following expression ⁸.

$$\Delta H = \frac{\sqrt{3}}{2} \Delta H_{pp}.$$

3.2.7 Spin Pumping Ferromagnetic Resonance

Spin pumping ferromagnetic resonance (SP-FMR) is basically characterized by the emission of a spin current driven by magnetization dynamics. In magnetic materials, FMR provides an efficient means of generating such spin currents that transports angular momentum. Spin pumping is the Onsager reciprocal effect of spin-transfer torque. In spin-transfer torque, an electrically generated spin-polarized current transfers angular momentum to the magnetization, thereby inducing magnetization dynamics through the torque exerted by the spins. In contrast, spin pumping represents the inverse process, in which the magnetization dynamics act as the source of a spin current [28].

⁸details can be found in[28]

In SP-FMR, an additional contribution to the linewidth appears due to the atomic contact between a ferromagnetic film (FM) and a nonmagnetic film (NM). The precessing magnetization generates a spin current that crosses the FM/NM interface, carrying angular momentum and consequently producing additional magnetization damping. This effect arises from the conservation of angular momentum where the spin current flowing from the FM into the NM layer removes angular momentum from the magnetization dynamics. In this sense, angular momentum is pumped out of the ferromagnetic material. Figure 39 shows the effect in a FM/NM bilayer, where the magnetization precession in the FM layer injects a pure spin current into the adjacent NM layer.

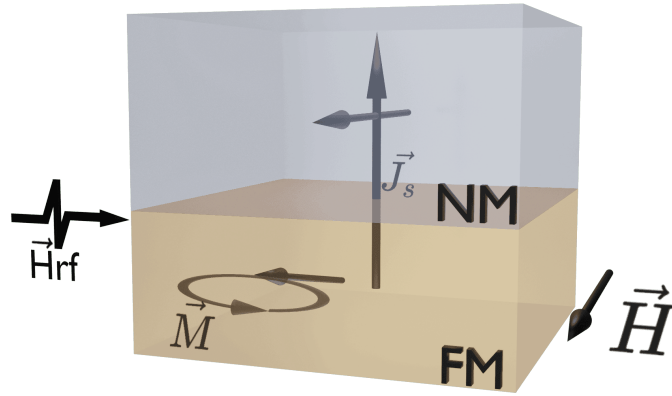


Figure 39 – Illustration of the SP-FMR mechanism. The angular momentum conservation leads to the injection of a spin current from the precessing magnetization in the FM layer into the NM layer.

The density of spin current injected at the FM/NM interface is given by the following expression [165, 166, 167].

$$\vec{J}_S^P = \frac{\hbar}{4\pi M} \left(\frac{g_r^{\uparrow\downarrow}}{M} \vec{M} \times \frac{d\vec{M}}{dt} - g_i^{\uparrow\downarrow} \frac{d\vec{M}}{dt} \right), \quad (3.15)$$

In this equation, the spin-mixing conductance is $g^{\uparrow\downarrow} = g_r^{\uparrow\downarrow} + i g_i^{\uparrow\downarrow}$ with dimension of 1/area. The terms $g_r^{\uparrow\downarrow}$ and $i g_i^{\uparrow\downarrow}$ are the real and imaginary part, respectively, defined as

$$g_r^{\uparrow\downarrow} = \frac{1}{2} \sum_{mn} [|r_{mn}^{\uparrow} - r_{mn}^{\downarrow}|^2 + |t_{mn}^{\uparrow} - t_{mn}^{\downarrow}|^2], \quad (3.16)$$

$$g_i^{\uparrow\downarrow} = \text{Im} \sum_{mn} [r_{mn}^{\uparrow} (r_{mn}^{\downarrow})^* + t_{mn}^{\uparrow} (t_{mn}^{\downarrow})^*], \quad (3.17)$$

where $r_{mn}^{\uparrow}$ ($r_{mn}^{\downarrow}$) represents the reflection coefficient (per unit area) for spin-up (spin-down) electrons at the interface, and $t_{mn}^{\uparrow}$ ($t_{mn}^{\downarrow}$) is the transmission coefficient (per unit area)

for spin-up (spin-down) electrons. The indices m and n label the electronic modes at the Fermi energy in the normal metal. Summarizing, the spin-mixing conductance is the interface transparency to the flow of spin currents. The real part represents a damping torque and is orders of magnitude greater than the imaginary part in real interfaces [165].

To quantify the additional damping produced by the contact between the NM and FM layers, the LLG equation must be supplemented with a negative additional term related to the angular momentum pumped out, as follows

$$\frac{d\vec{M}}{dt} = \gamma \vec{M} \times \vec{H}_{eff} - \frac{\alpha}{M} \vec{M} \times \frac{d\vec{M}}{dt} - \frac{\gamma \hbar}{4\pi M^2 t_{FM}} g_{eff}^{\uparrow\downarrow} \vec{M} \times \frac{d\vec{M}}{dt}. \quad (3.18)$$

Here t_{FM} is the thickness of the FM layer, and the $g_{eff}^{\uparrow\downarrow}$ is the real part of the spin-mixing conductance. With this, the total Gilbert parameter (α_T) becomes [28]

$$\alpha_T = \alpha + \frac{\gamma \hbar g_{eff}^{\uparrow\downarrow}}{4\pi M t_{FM}}. \quad (3.19)$$

The SP-FMR also produces a increase in the linewidth expression as follows

$$\Delta H_T = \Delta H_{FM} + \frac{\hbar \omega g_{eff}^{\uparrow\downarrow}}{4\pi M t_{FM}}, \quad (3.20)$$

Here, ΔH_{FM} is the linewidth of the FM layer in the absence of the NM layer, and ω is the FMR frequency. This equation reveals a signature of spin pumping induced damping, in which the linewidth exhibits a linear dependence on frequency, while $g_{eff}^{\uparrow\downarrow}$ shows an inverse dependence on the FM layer thickness[167, 168, 28]. With this, it is possible to obtain the $g_{eff}^{\uparrow\downarrow}$ by the increment in the linewidth by the expression

$$g_{eff}^{\uparrow\downarrow} = \frac{4\pi M t_{FM}}{\hbar \omega} (\Delta H_T - \Delta H_{FM}). \quad (3.21)$$

The thickness of the ferromagnetic layer, t_{FM} , should be replaced by the spin coherence length when the latter exceeds t_{FM} . A DC electric voltage in the adjacent NM layer is one of the most well-known effects associated with SP-FMR. This effect was first observed experimentally by Azevedo and coworkers in 2004, and its theoretical description was later provided by Saitoh and coworkers in 2006 [33, 34]. The SP-FMR is one of the most used ways to produce spin currents in two and three-dimensional electron systems [28]. The physical mechanism underlying the generation of a DC electric voltage conversion is the ISHE or IREE, that was already discussed in **Inverse Spin Hall effect** section. and **Inverse Rashba-Edelstein Effect** section.

3.2.8 Spin Seebeck effect experiments

The experimental setup used for the SSE measurements is based on the application of a thermal gradient ∇T perpendicular to the sample surface, allowing the investigation of spin-current generation driven by heat flow in magnetic heterostructures. The thermal gradient is generated using two Peltier modules, one placed on top of the sample and the other beneath it. This configuration allows both positive and negative thermal gradients to be applied. A static external magnetic field $\vec{H}$ is applied in the plane of the sample to define the magnetization direction of the magnetic layer. Figure 45 exemplifies the SSE configuration.

Under a thermal gradient, a magnonic spin current J_S is generated in the magnetic insulator layer (YIG). This spin current is injected across the interface into the adjacent layer, which can be either a metal or an organic semiconductor, depending on the heterostructure under investigation. The efficiency of spin transmission across the interface is determined by the interfacial properties and the spin transport characteristics of the intermediate layer. In the presence of strong SOC in the detecting layer (usually a heavy metal), the injected spin current is converted into a transverse charge current via the ISHE or IREE, depending on the material system and thicknesses. The resulting charge accumulation leads to the development of a DC voltage across the sample.

The generated SSE voltage V_{SSE} is measured using electrical contacts placed at the edges of the detecting layer and recorded with a nanovoltmeter. This setup allows the direct correlation between the applied thermal gradient, the magnetic field direction, and the measured voltage signal, providing a platform for investigating spin-to-charge conversion and pure spin-current transport in distinct material systems. More details about the SSE experimental setup and conditions are discussed in **Spin Seebeck Effect** section of the result chapter.

4 RESULTS

This chapter is dedicated to the analysis of the experimental data obtained from the characterization techniques described in the chapter **Materials Synthesis and Characterization of Organic Semiconductors and Topological Insulators**. The results are divided into three sections, the organic semiconductors PANI and PEDOT:PSS, and the topological crystalline insulator tin telluride (SnTe). Finally, a discussion is presented to connect the results and summarize the main conclusions.

4.1 PANI Results and Discussions

The creation, detection, and manipulation of spin currents are among the most extensively studied research topics in spintronics [169]. In our work entitled “*Spin transport and spin-to-charge current conversion in polyaniline by means of spin Seebeck experiments*” [71], we studied the spin transport and spin-to-charge current conversion properties in a heterostructures containing a PANI thin films. This section describes the synthesis of the organic semiconductor PANI and the fabrication of the heterostructures containing YIG and Pt. Estimation of properties such as thickness, roughness, electrical resistivity, and behavior under doping process were taken to characterize this material. In order to obtain parameters such as the spin Hall angle and the spin-mixing conductance, spin Seebeck, FMR, and SP-FMR experiments were performed, as described in the following subsections.

4.1.1 Thickness and roughness of PANI thin films

In the preparation of our samples, YIG was chosen as the substrate due to its insulating and ferrimagnetic properties and low Landau damping [28]. These features make YIG suitable for spintronic applications, since it can generate pure spin currents without charge transport. The PANI thin films were prepared by the spin-coating technique using a 6 μm -thick YIG film grown on GGG as substrate. The PANI solution (emeraldine salt state) was spin-coated onto the YIG layer under ambient conditions at a rotation speed of 3000 rpm for 60 seconds, forming a film with a thickness of approximately 100 nm. The thickness of the PANI films was estimated by AFM measurements taken orthogonally to a scratch purposely made on the film surface. Figure 40 (b) illustrates the thickness

estimation method applied.

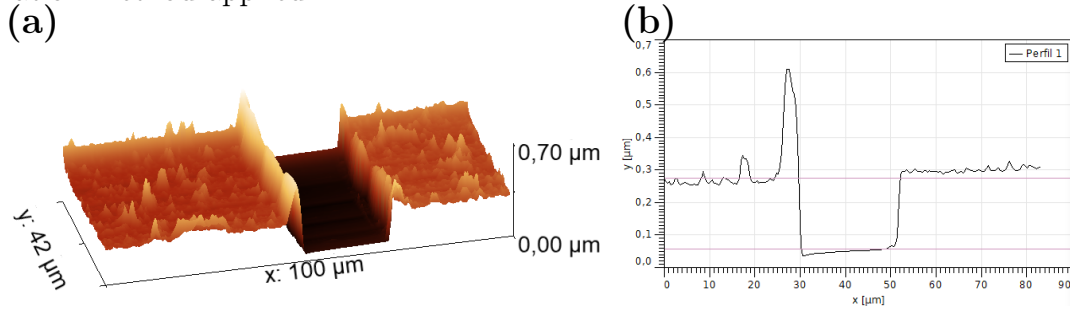


Figure 40 – Schematic representation of the thickness calibration process for PANI thin films deposited by spin-coating. Panel (a) shows a 3D AFM image of the scratch made on the film surface, and (b) the corresponding profile used to estimate the film thickness.

AFM topographic images of all surfaces of the Pt/PANI/YIG heterostructure were taken to evaluate the influence of the PANI layer on the root-mean-square roughness (R_{rms}) of the sputter-deposited Pt. The first step was to obtain the R_{rms} of the YIG surface, which was estimated to be 0.8 nm, as shown in Figure 41 (a). Subsequently, AFM images of the PANI/YIG and Pt/PANI/YIG samples with an area of $500 \text{ nm} \times 500 \text{ nm}$ were acquired. For the PANI surface, $R_{\text{rms}} = 2.652 \text{ nm}$, indicating a relatively rough surface (see Figure 41 (b)). Similarly, the Pt surface in the Pt/PANI/YIG trilayer presented an $R_{\text{rms}} = 2.758 \text{ nm}$, which is very close to that obtained for the PANI surface. Moreover, it is possible to notice a change in the granular composition of the surface. These results suggest that the PANI layer induces the formation of grains on the Pt surface, as shown in Figure 41 (c).

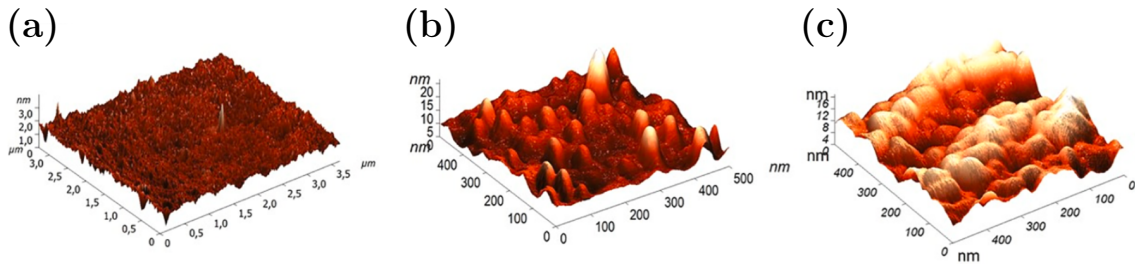


Figure 41 – AFM images of (a) the YIG film surface, (b) the PANI surface in the PANI/YIG bilayer, and (c) the Pt surface in the Pt/PANI/YIG trilayer. The r_{ms} surface roughness of the bare YIG film was approximately 0.8 nm. The PANI layer in the PANI/YIG bilayer exhibited a roughness of $R_{\text{rms}} = 2.652 \text{ nm}$. The Pt surface in the Pt/PANI/YIG trilayer showed a roughness of $R_{\text{rms}} = 2.758 \text{ nm}$, very similar to that of the PANI surface, indicating that the PANI roughness induced the formation of grains in the Pt film.

After the fabrication of the PANI/YIG structure, a secondary doping process was

performed. The sample was immersed in a sulfuric acid (H_2SO_4) solution for 10 seconds and then dried under ambient conditions. The purpose of this step is to further increase the sample conductivity. This process is known to be reversible, as the conductivity changes upon acid or base immersion, and the sample changes its color from green to blue during the transition between the emeraldine salt and emeraldine base states [96].

The step of deposition of a 10 nm thick Pt layer on top of the PANI film by the sputtering technique. Ag electrodes were also deposited by sputtering on the PANI edges, using a mask to cover the central region of the sample. The Ag was used to fabricate the electrical contacts, as shown in Figure 42 (a). The current–voltage (I–V) curve was obtained for the PANI/YIG heterostructure, and it can be verified from Figure 42 (b) that the contacts are indeed ohmic within the current range up to 1 ampere. It is also possible to observe that both PANI films exhibit relatively low resistance values 14.13 k Ω for the sample immersed for 10 seconds and 6.39 k Ω for the sample immersed for 20 seconds. These resistance values ($\sim 10^4 \Omega/\text{cm}$) are typically associated with inorganic semiconductors. This result justifies the “organic semiconductor” term commonly used for conjugated polymers [64, 123]. The decrease to almost half the resistance value for the sample doped for 20 seconds, compared with the one doped for 10 seconds, shows that this process is indeed efficient in increasing the conductivity of the samples.

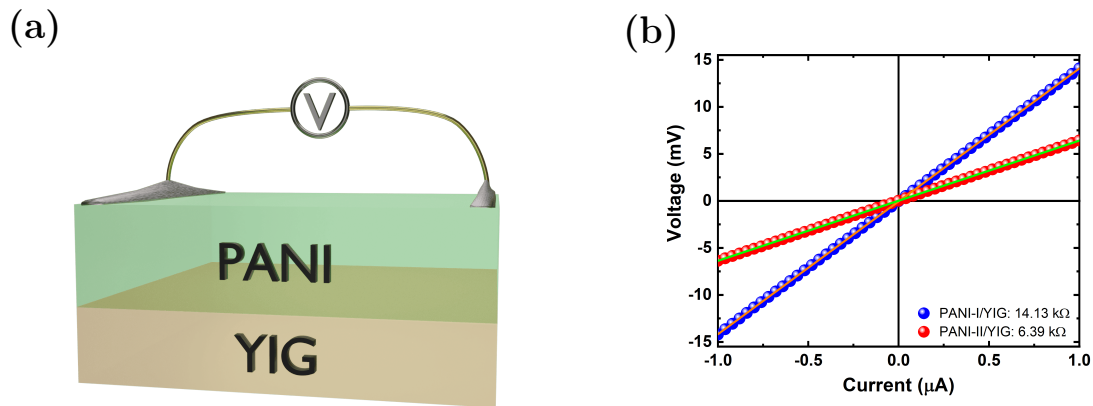


Figure 42 – Schematic representation of (a) Pt/PANI/YIG and (b) current–voltage (I–V) characteristics of the PANI/YIG structure. The linear dependence observed in the I–V curve indicates the formation of ohmic contacts between the Ag electrodes and the PANI layer. The DC conductivity of PANI ranged from 2.5 to 14.3 ($\Omega \text{ cm}$) $^{-1}$, values that are characteristic of a highly doped semiconductor.

Samples exposed to the acid solution for extended periods may exhibit surface

irregularities due to the aggressive nature of the chemical treatment. For this reason, the sample immersed for 10 seconds, which did not show significant changes in its surface morphology, was chosen for the FMR measurements, as discussed in the next section.

4.1.2 Ferromagnetic Resonance - PANI

Ferromagnetic resonance microwave excitation measurements were performed in our samples with the main objective of estimate the real part of spin-mixing conductance ($g_{\text{eff}}^{\uparrow\downarrow}$) for the different interfaces of Pt/PANI/YIG heterostructure. This parameter can be estimated by the following equation

$$g_{\text{eff}}^{\uparrow\downarrow} = \frac{4\pi M t_{\text{coh}}}{\hbar\omega} \delta H, \quad (4.1)$$

where $4\pi M = 1.76$ kG is the saturation magnetization at room temperature of Py [170], t_{coh} is the YIG film coherence length, $\hbar$ is the plank's constant, and δH is the additional linewidth that is obtained by the dP/dH versus $\vec{H}_{rf}$ spectra [170]. In this measurement, a shorted waveguide setup as used and the sample was positioned in a position of maximum rf magnetic field and zero electric field using a PVC rod. The static magnetic field ($\vec{H}$) and the microwave magnetic field ($\vec{H}_{rf}$) were applied in the plane of the film and oriented perpendicular to each other. A schematic representation of the FMR system used in our measurements is shown in Figure 43 (b). The theoretical details about the FMR is discussed in **Ferromagnetic resonance** section.

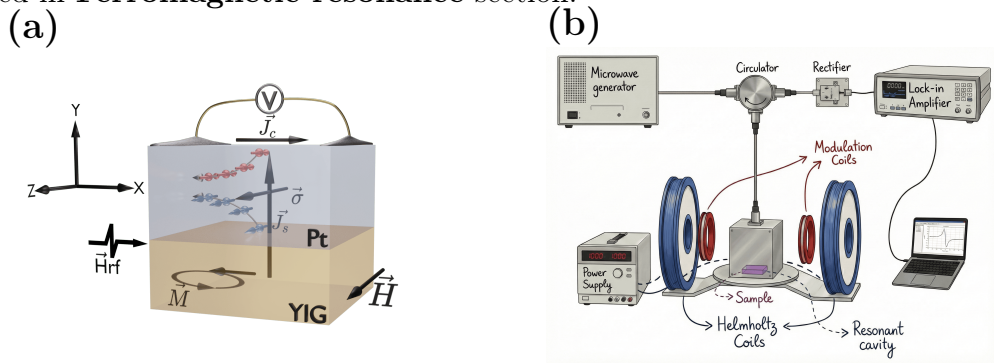


Figure 43 – The spin-to-charge schematic illustration along the heterostructure is shown in (a). Panel (b) shows the FMR setup configuration.

The derivative of the microwave absorption (dP/dH) as a function of the magnetic field $\mathbf{H}$ (FMR spectra) was obtained using a lock-in amplifier. A microwave field with a frequency of 1.2 kHz was applied by the coils, modulating the signal and making it

detectable by the lock-in amplifier. Figure 44 shows the FMR absorption spectra of bare YIG, Pt(10 nm)/YIG, PANI(100 nm)/YIG, and Pt(10 nm)/PANI(100 nm)/YIG respectively. The FMR spectra were recorded using a microwave frequency of 9.4 GHz and with input power of 30 mW. The static magnetic field was applied along the z -axis, as shown in Figure 43 (a).

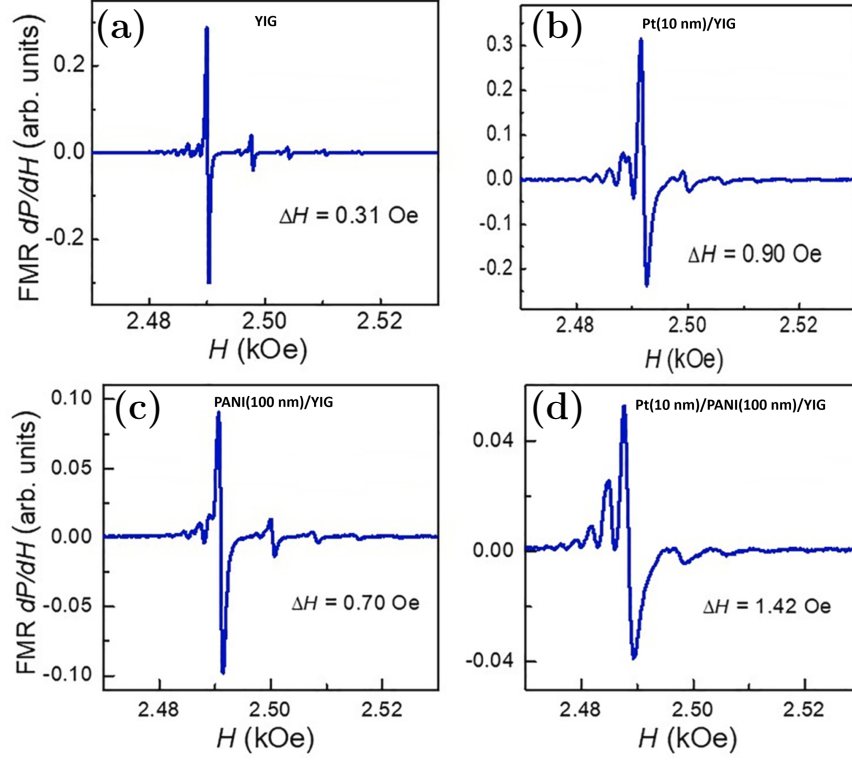


Figure 44 – Field-swept FMR derivative spectra recorded at 9.4 GHz for 6 μm -thick YIG films with different layers: (a) bare YIG, (b) Pt (10 nm)/YIG, (c) PANI (100 nm)/YIG, and (d) Pt(10 nm)/PANI (100 nm)/YIG (10 nm). The magnetic field was applied in the film plane and oriented perpendicular to the long axis. All samples have lateral dimensions of $1.5 \times 3.0 \text{ mm}^2$.

The discrete lines observed in Figure 44 (a) correspond to the standing spin-wave modes of the bare YIG. These modes have in-plane wave numbers quantized by the film dimensions, which define the boundary conditions. They are:

$$k_x = n_x \pi / L_x$$

$$k_z = n_z \pi / L_z$$

where $L_x = 3.0 \text{ mm}$ and $L_z = 1.5 \text{ mm}$ are the lengths of the sample. The most intense line corresponds to the fundamental FMR mode with $n_x = 1$ and $n_z = 1$, whose frequency

is close to that of the uniform spin-wave mode ($k = 0$), described by the equation

$$\omega_0 = \gamma(H + H_A)^{1/2}(H + H_A + 4\pi M + H_S)^{1/2}, \quad (4.2)$$

where γ is the gyromagnetic ratio ($\gamma = g\mu_B/\hbar = 2\pi \times 2.8$ GHz/kOe for YIG), $4\pi M$ is the saturation magnetization (1.76 kG at room temperature) [170], and H_A and H_S represent the in-plane and out-of-plane anisotropy fields, respectively. The resonance lines located to the left of the main FMR peak correspond to hybridized standing spin-wave surface modes, while those to the right are associated with volume modes. All modes exhibit a similar peak-to-peak linewidth of 0.36 Oe, which corresponds to a half-width-at-half-maximum (HWHM) value of $\Delta H_{\text{YIG}} = 0.31$ Oe.

From the spectra presented in Figure 44, the HWHM values obtained for the Pt/PANI/YIG, PANI/YIG, and Pt/YIG structures [Figures 2 (d), (c), and (b), respectively] are 1.42 Oe, 0.70 Oe, and 0.90 Oe, all of which are broader than the 0.31 Oe measured for the bare YIG film. The contact between the PANI layer and the YIG film leads to a broadening of the linewidth, which originates from the spin current carrying angular momentum out of the YIG layer [97].

A slight linewidth broadening is observed when the PANI layer is placed on top of the YIG, and a more pronounced broadening occurs with the addition of the Pt layer onto the PANI/YIG bilayer. From this additional broadening, the real part of the effective spin-mixing conductance $g_{\text{eff}}^{\uparrow\downarrow}$ for the PANI/YIG interface can be estimated using Equation 4.1. From the data we determined $g_{\text{PANI/YIG}}^{\uparrow\downarrow} = 0.56 g_{\text{YIG/Pt}}^{\uparrow\downarrow}$ for the PANI/YIG bilayer and $g_{\text{PANI/Pt/YIG}}^{\uparrow\downarrow} = 1.6 g_{\text{YIG/Pt}}^{\uparrow\downarrow}$ for the Pt/PANI/YIG trilayer. The relatively large $g_{\text{PANI/Pt/YIG}}^{\uparrow\downarrow}$ value indicates an efficient spin transfer across the PANI/Pt/YIG heterostructure, where the spin current pumped from YIG is transmitted through the 100 nm-thick PANI layer.

These results demonstrate that only a small portion of the spin current is absorbed by the PANI itself, while a significant fraction reaches the Pt layer, where it is efficiently absorbed due to the strong spin-orbit coupling and high spin-sink capability of Pt. This interpretation is consistent with the spin Seebeck effect (SSE) measurements discussed in the next section.

4.1.3 Spin Seebeck Effect - PANI

In order to verify the spin transport efficiency of PANI, the SSE measurements were performed. Two different heterostructures were used, as shown in Figure 45. The experiments were carried out by applying a thermal gradient ∇T perpendicular to the sample surface using two Peltier modules one on top and another under the samples. A static magnetic field $\vec{H}$ was also applied. This generates a spin current density J_S in the YIG layer, which is injected through the PANI/YIG interface and transmitted to the Pt layer. In Pt, the spin current is converted into a charge current due to its strong spin-orbit coupling and detected as an Inverse Spin Hall Effect (ISHE) voltage, V_{SSE} . The DC voltage was measured using a nanovoltmeter.

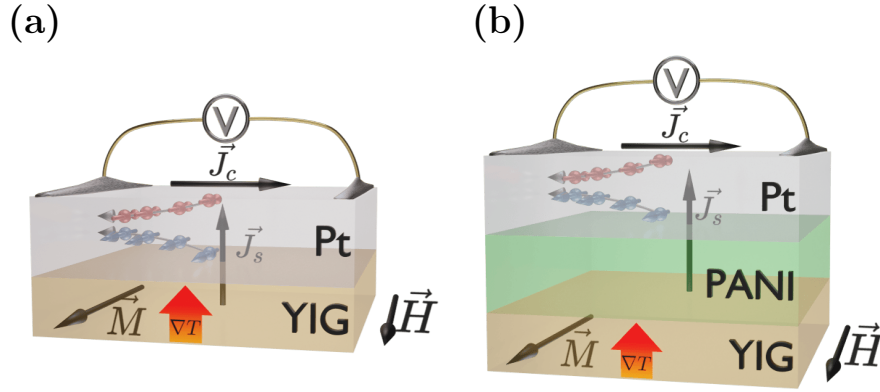


Figure 45 – Schematic illustration of (a) Pt(10 nm)/YIG and (b) Pt(10 nm)/PANI(100 nm)/YIG heterostructures used in the spin Seebeck experiments.

Figures 46 (a) and (b) present V_{SSE} as a function of the applied magnetic field $\mathbf{H}$ for the Pt(10 nm)/YIG and Pt(10 nm)/PANI(100 nm)/YIG samples respectively. Different values of ΔT were applied and maintained constant during the $\mathbf{H}$ scan. It is observed that the V_{SSE} saturation values for the Pt(10 nm)/PANI(100 nm)/YIG samples are about one order of magnitude smaller than those for the Pt(10 nm)/YIG sample, as expected due to spin current losses through the PANI layer. Additionally, in Figure 46 (b) the saturation region at higher magnetic fields shows characteristic fluctuations of the experimental setup used for SSE measurements.

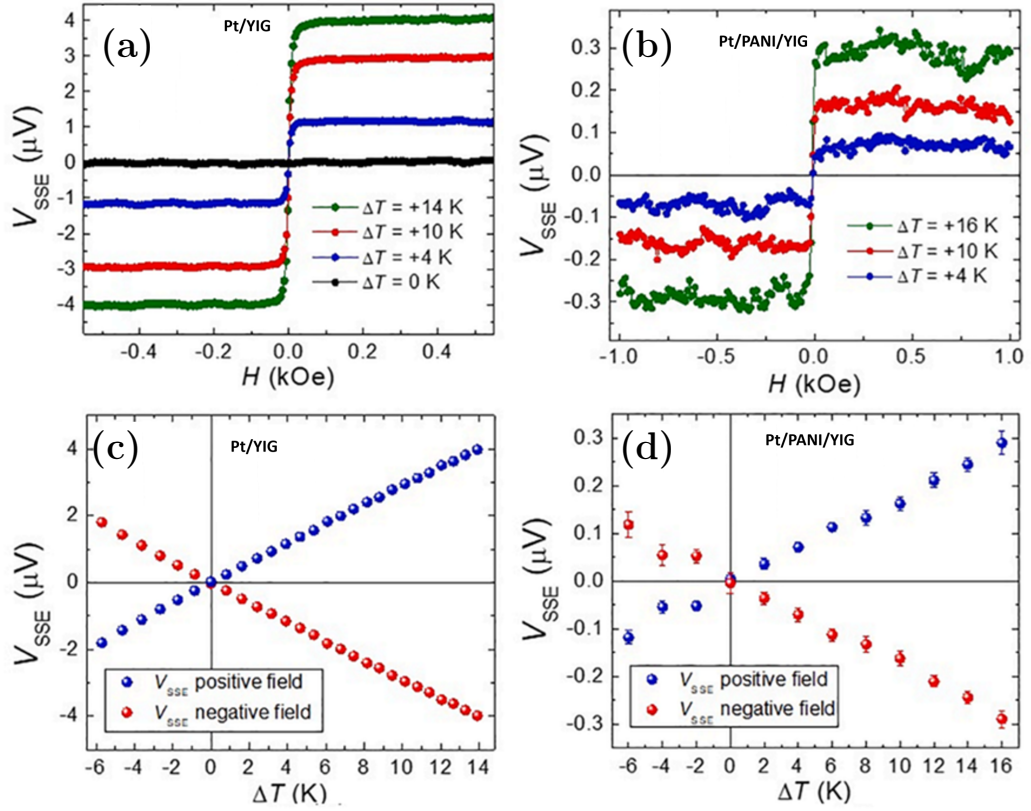


Figure 46 – Variation of the voltage difference V_{SSE} as a function of the external magnetic field at different temperatures for a Pt/YIG sample with a thickness of 10 nm. In (b), the same analysis is presented for Pt(10 nm)/PANI(100 nm)/YIG, where the PANI layer was slightly doped for 10 seconds in sulfuric acid solution. The variation of the voltage difference V_{SSE} with temperature is shown for (a) Pt(10 nm)/YIG and (b) Pt(10 nm)/PANI(100 nm)/YIG, measured under an external field of $H = 1.0$ kOe in two opposite field directions. The error bars in correspond to the V_{SSE} fluctuations in the saturation region, while for the YIG/Pt sample, the smooth saturation results in negligible error bars.

The Peltier module allows the application of both positive and negative thermal gradients across the sample. Since $V_{SSE} \propto J_S \propto \nabla T$ [171], reversing the direction of $\vec{J}_S$ also changes the sign of V_{SSE} . This behavior is illustrated in Figures 46 (c) and (d), which display V_{SSE} as a function of ΔT for the Pt/YIG and Pt/PANI/YIG samples, respectively. Each point represents the mean V_{SSE} value in the saturation region for the corresponding ΔT , considering the magnetic field direction. The voltage sign reversal with the change in field direction reflects the reversal of the spin polarization.

To better understand the spin-to-charge current conversion in PANI, we analyzed the SSE voltage in the bilayer structure of PANI(100 nm)/YIG(6 μm), as shown in Figure 45 (a). Two electrical contacts of Ag were fabricated by sputtering at the edges of the PANI

layer, as like the previous one. Figure 47 (b) presents the current-voltage (I-V) curve obtained using a pseudo-four-probe technique. The linear I-V response of the PANI/YIG bilayer indicates the formation of Ohmic contacts between the electrodes between the range analyzed. The DC conductivity of PANI ranged from 2.5 to 14.3 (Ωcm)⁻¹, values typical of a highly doped semiconductor and in accordance with literature reports [95].

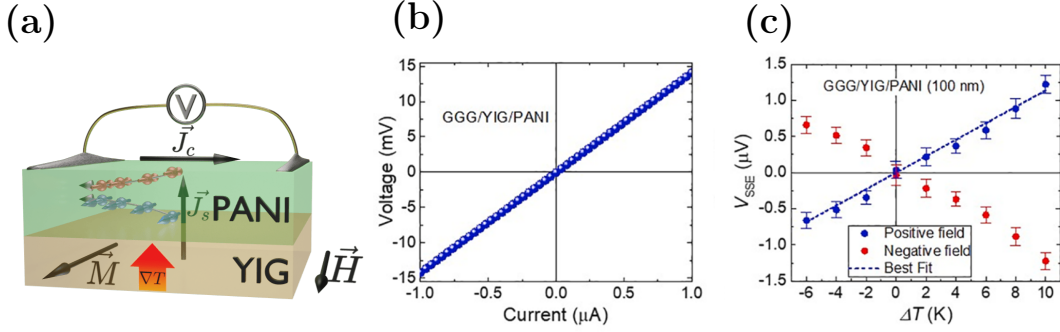


Figure 47 – Schematic illustration of the PANI(100 nm)/YIG heterostructure. The I-V curve in (b) shows the ohmic behavior of the contacts between the electrodes and the PANI. The variation of the SSE voltage with temperature difference measured in the PANI layer is shown in (c). The static magnetic field of $H = 1.0$ kOe was applied in two field directions of the PANI(100 nm)/YIG bilayer sample. The blue dashed line represents the best linear fit of V_{SSE} versus ΔT for measurements with a positive magnetic field.

By applying a thermal gradient across the sample, we observed spin-to-charge current conversion measured directly on the PANI layer. Figure 47 (c) shows V_{SSE} as a function of applied temperature differences ΔT , following the same procedure described previously. The V_{SSE} values are on the same order of magnitude as those obtained for the Pt/YIG sample. It is worth noting that the resistance of the PANI/YIG sample is $R_{\text{PANI}} \approx 20$ k Ω , much higher than that of the Pt/PANI/YIG trilayer, $R_{\text{Pt}} = 55$ Ω . Therefore, although the ISHE occurs in PANI, in the sample containing Pt the generated charge current mainly flows through the lower-resistance Pt layer.

The spin current at the PANI/YIG interface is expressed as

$$J_S = -C_S \rho g_{\text{eff}}^{\uparrow\downarrow} \nabla T, \quad (4.3)$$

where $C_S = 3.3 \times 10^{-24}$ erg $\cdot\text{cm}\cdot\text{K}^{-1}$ is a coefficient depending on the intrinsic properties of YIG such as temperature, and also the applied magnetic field, ρ accounts for the finite thickness effect of the YIG layer, and $\nabla T = \Delta T/t_{\text{sample}}$ is the thermal gradient

considering $t_{\text{sample}} \simeq 0.05$ cm as the total sample thickness, including the GGG substrate [171, 172, 173]. The resulting V_{SSE} is related to J_S by the expression [171]

$$V_{\text{SSE}} = F_P R w (2e/\hbar) \lambda \theta_{\text{SH}} \tanh\left(\frac{t_{\text{PANI}}}{2\lambda}\right) J_S \quad (4.4)$$

where R , w , λ , θ_{SH} , and t_{PANI} are, respectively, the resistance, width, spin diffusion length, spin Hall angle, and thickness of the PANI layer. The factor $F_P \simeq 2/3$ corresponds to the surface fraction covered by the Peltier heater used to generate the thermal gradient ∇T .

The spin Hall angle θ_{SH} quantifies the efficiency of the spin-to-charge current conversion. Using the SSE measurement data from the PANI/YIG bilayer, the value of θ_{SH} can be estimated by performing a linear fit of V_{SSE} as a function of ΔT . The linear relationship between V_{SSE} and ΔT is given by:

$$V_{\text{SSE}} = \theta_{\text{SH}} P \Delta T,$$

where

$$P = F_P R w \lambda \left(\frac{2e}{\hbar}\right) \left[\frac{C_S \rho g_{\text{eff}}^{\uparrow\downarrow}}{t_{\text{sample}}}\right] \tanh\left(\frac{t_{\text{PANI}}}{2\lambda}\right).$$

It represents a combination of constant parameters for this sample, obtained by substituting J_S into the expression for V_{SSE} .

The best fit is represented by the blue dashed line in Figure 47 (c), with a slope of $P\theta_{\text{SH}} = (1.14 \pm 0.04) \times 10^{-7}$ V/K. Using the parameters listed in Table 1, we obtained $P = 6.72 \times 10^{-3}$ V/K, and finally

$$\theta_{\text{SH}} = (1.70 \pm 0.06) \times 10^{-5}.$$

Table 1 – Parameters used in the calculation of spin pumping in PANI/YIG films.

Parameter	Value	Reference
Elementary charge, e	1.6×10^{-19} C	—
Resistance, R	6.0 k Ω	—
Film width, w	0.3 cm	—
PANI thickness, t_{PANI}	100 nm	—
Spin mixing conductance, $g_{\text{eff}}^{\uparrow\downarrow}$	$0.56 g_{\text{Pt/YIG}}^{\uparrow\downarrow} = 5.6 \times 10^{13}$ cm $^{-2}$	[174, 175, 176]
Pt/YIG spin mixing conductance, $g_{\text{Pt/YIG}}^{\uparrow\downarrow}$	$\sim 10^{14}$ cm $^{-2}$	[174, 175, 176]
Parameter ρ	≈ 1 for $t_{\text{YIG}} \gg \lambda$	[173]
PANI spin diffusion length, λ	$\simeq 590$ nm	[97]

This result indicates that the spin Hall angle θ_{SH} measured for PANI is consistent with values reported for similar polymeric materials [20, 107, 11]. It is important to note that the SSE technique may present uncertainties related to experimental conditions and temperature gradients within the samples [177, 178]. Therefore, these results provide an estimate of the spin Hall angle of PANI based on SSE measurements performed directly on this polymer.

4.2 PEDOT:PSS Results and Discussions

This section presents the results obtained from PEDOT:PSS thin films used to investigate spin-to-charge current conversion. Structural characterization was performed via AFM to evaluate the film thickness and roughness. Raman spectroscopy was performed providing a spectrum with the main peaks associated to this material, all they labeled according to chemical bonding. Optical absorbance measurements were also conducted to determine the optical energy band gap, using Tauc plot methodology. Finally, results from spin-to-charge conversion were obtained through the SSE and SP-FMR methods, providing a comprehensive picture of the material's performance.

4.2.1 Thickness of PEDOT:PSS films

The PEDOT:PSS films used in this analysis were produced by the drop-casting technique. The thickness value of the films is essential for the estimation of the optical energy gap by the Tauc method, for example. In our experiments, the thickness is considered the optical path length that light travels during the absorbance measurement.

Figure 48 shows the thickness values of PEDOT:PSS films, which were estimated using the AFM technique. The film thickness was determined by performing measurements at various points across a step edge. This step was created by scratching the film to expose the substrate.

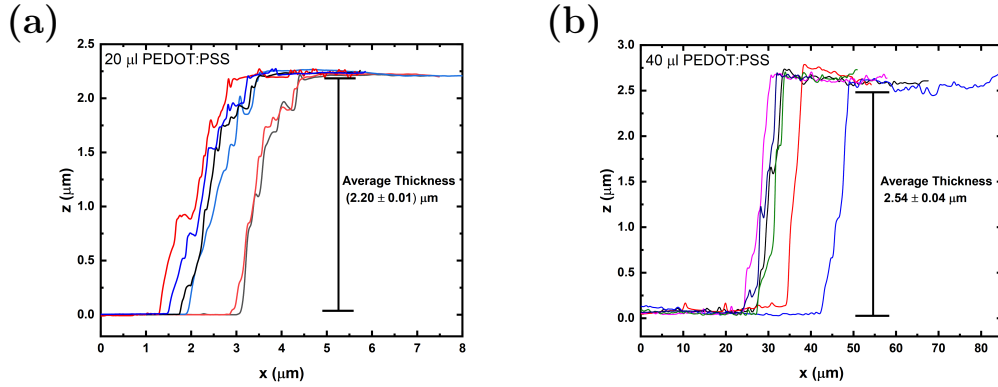


Figure 48 – Thickness of the films produced using (a) one drop (20 μL) and (b) two drops (40 μL) of PEDOT:PSS solution deposited on a quartz substrate.

From these results, we can observe that the film produced with 40 μL (two drops) is slightly thicker than the one produced with 20 μL (one drop). This result suggests that the film thickness is approaching a saturation regime as the volume of the deposited solution increases beyond 40 μL . Figure 49 shows AFM images of the PEDOT:PSS film surface. The RMS roughness value is strongly dependent on the sample preparation technique. The value of $R_{\text{rms}} = 5.11 \text{ nm}$ is relatively large, which implies that PEDOT:PSS films exhibit a higher surface area compared to PANI films produced by spin coating.

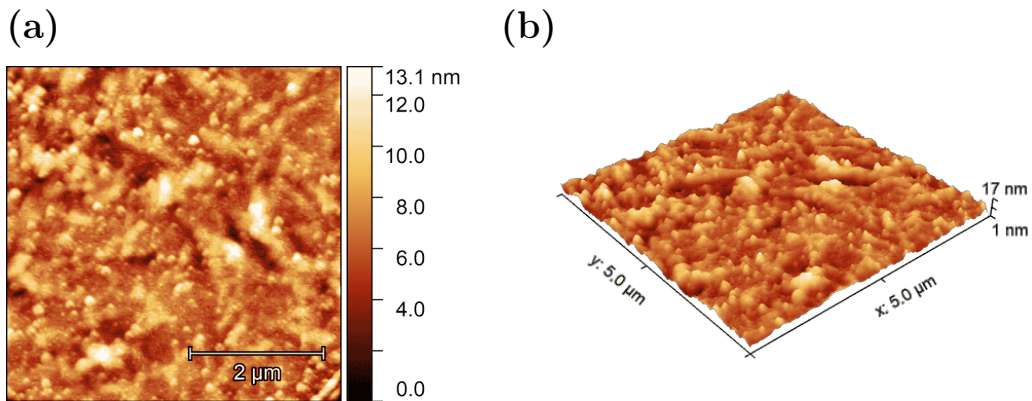


Figure 49 – AFM image of the PEDOT:PSS film surface. The RMS roughness is approximately $R_{\text{rms}} = 5.11 \text{ nm}$ over an area of $(2 \times 2) \mu\text{m}^2$. Since the PEDOT:PSS film was deposited by the drop-casting technique, its surface roughness can vary significantly near the edges of large areas. In this case, the RMS roughness can reach values of several tens of nanometers.

From Figure 49, it is possible to observe a granular morphology on the surface, which is a characteristic feature of PEDOT:PSS, since excess PSS promotes the formation of a core-shell structure, as previously discussed.

4.2.2 Raman Spectroscopy Results of PEDOT:PSS

The Raman spectrum of PEDOT:PSS deposited on a quartz substrate using the *drop-casting* technique were acquired using a Renishaw InVia micro-Raman system equipped with an argon laser operating at a wavelength of 514.5 nm. The Raman shifts and their intensities of PEDOT:PSS are correlated with the electronic structure and bonding of poly- and oligothiophenes containing π -conjugated backbones [179]. Several Raman bands associated with the PEDOT polymer backbone are clearly observed in the spectrum in the Figure 50, also its chemical structure is shown in the inset.

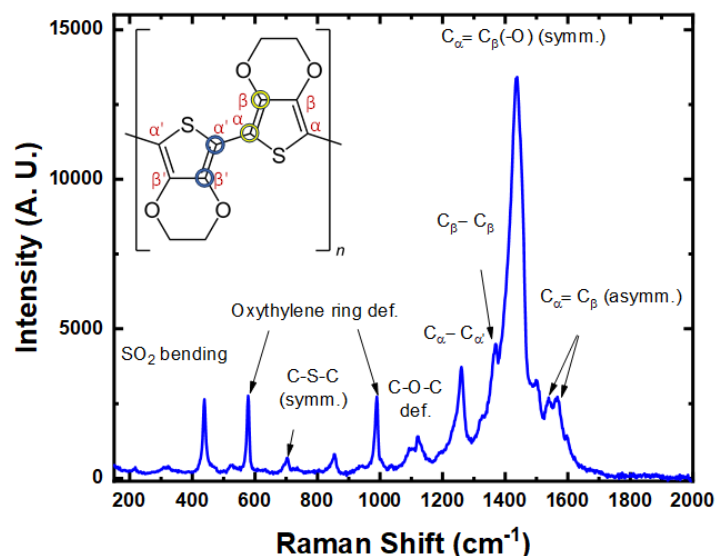


Figure 50 – Raman spectrum of PEDOT:PSS film deposited on quartz substrate. The main peak are labeled according to the corresponding molecular bonds. The inset shows the PEDOT chemical structure.

The bands at 1563 cm^{-1} and 1532 cm^{-1} are related to the asymmetric $C_{\alpha} = C_{\beta}$ stretching mode. The band at 1421 cm^{-1} corresponds to the symmetric¹ $C_{\alpha} = C_{\beta}$

¹In the symmetric stretching mode, the peripheral atoms oscillate in phase, moving away from and toward the central atom simultaneously. Conversely, in the asymmetric stretching mode, as one bond elongates, the opposite bond contracts.

stretching mode involving the $-O$ group. The peak at 1368 cm^{-1} is attributed to the $C_\beta - C_\beta$ stretching mode, while the band at 1256 cm^{-1} is associated with the inter-ring $C_\alpha - C_\alpha$ stretching. The band at 1093 cm^{-1} corresponds to the $C - O - C$ deformation mode, whereas the peaks at 989 cm^{-1} and 577 cm^{-1} are related to the oxyethylene ring deformation. The symmetric $C - S - C$ deformation mode appears at 701 cm^{-1} , and the band at 437 cm^{-1} is assigned to the SO_2 bending mode. The peak positions and their corresponding assignments are in good agreement with values reported in the literature for commercial PEDOT:PSS [179, 180, 181].

Figure 50 presents the identification of the Raman peaks associated with PEDOT:PSS. The peak positions were marked based on the reference [179]. It can be observed that most of the Raman peaks identified in this work are consistent with those reported in the literature. The main vibrational modes of PEDOT:PSS are clearly present in our results. The reference [179] discusses in detail the physical origin of the vibrational modes of PEDOT:PSS observed in Raman spectroscopy. Additionally, Raman peaks associated with the quartz substrate are expected at 540 cm^{-1} and 1092 cm^{-1} . However, their intensities are relatively weak and therefore they do not significantly appear in the PEDOT:PSS/quartz spectra.

4.2.3 Absorbance of PEDOT:PSS films

For the transmittance measurements, a manually crafted Teflon mask was used (see Figure 51) in a Fluoromax Plus R928P equipment. Its purpose was to limit the area of the film exposed to the light beam transmitted through the film by the UV-Vis equipment. The central hole in the mask has a diameter of 4 mm , which is smaller than the diameter of the PEDOT:PSS films deposited on quartz. The mask was positioned over the film so that the entire area of the central hole was covered by the film. This ensured that all the light captured by the detector had passed exclusively through the regions covered by the PEDOT:PSS film. In this way, it is possible to compare the transmittance intensity of different samples.

In order to choose the most appropriate substrate used in our UV-Vis experiments, we acquired the transmittance spectra for 1 mm glass and quartz as shown in Figures 52 (a) and (b). From Figure 52 (a) we observe that a significant amount of light in

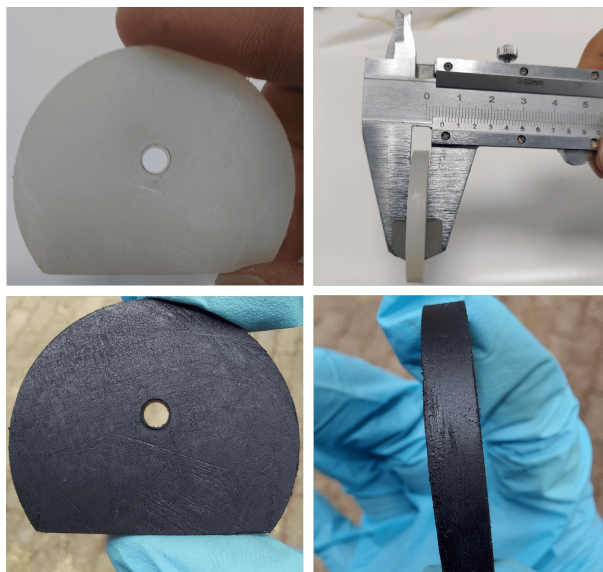


Figure 51 – A Teflon mask with a thickness of 5 mm and a central hole diameter of 4 mm coated with matte black spray paint.

the wavelength range of approximately 250 *nm* to 380 *nm* is absorbed by glass, whereas the quartz substrate absorbs considerably less in this range. From this, between the two substrates analyzed, we concluded that quartz is indeed the most suitable one for studying the interaction of light with our polymer. Due to technical limitations of the UV-Vis equipment used, light with wavelengths below 250 *nm* should not be considered for phenomenological analyses.

The influence of the Teflon mask on the light transmittance of the quartz substrate were analyzed in Figure 52 (b). The mask does not significantly affected the positions of the PEDOT:PSS transmittance peaks. We can observe that there is a noticeable change only in the peak intensity, which was expected, since the area of the substrate available to transmit light is smaller when the mask is used.

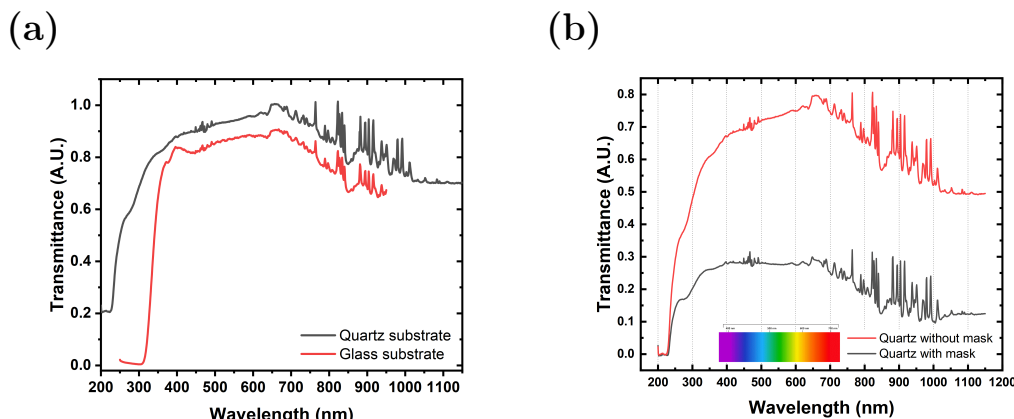


Figure 52 – The transmittance of the glass substrate is shown in (a). Panels (b) shows the transmittance of the quartz substrate (1 mm thick) with and without the Teflon mask.

The transmittance data of the PEDOT:PSS/quartz samples were obtained to estimate the optical band gap of this polymer. The curve corresponding to the quartz substrate (Figure 52 (b)), measured with the mask, was taken as the background in subsequent analyses of the PEDOT:PSS absorption spectra, as discussed below.

4.2.4 The optical band gap of PEDOT:PSS films

The optical bandgap corresponds to the photon energy that, when absorbed by an electron, leads to the formation of an electron-hole pair (exciton) within the film, bound together by the Coulomb interaction [182]. Materials with high optical band gap values (2.0–3.5 eV) exhibit maximum absorption and emission in the visible/UV range, which makes them more suitable for applications in visible LEDs, displays, and organic field-effect transistors (OFETs) [92, 183, 184]. In contrast, materials with low optical band gap values (1.0–1.8 eV) exhibit strong absorption of the solar spectrum, making them excellent candidates for use as active layers in solar cells and in near-infrared (NIR) LEDs [185, 186, 187].

For the estimation of the optical band gap, the absorbance spectra of samples were taken. From the normalized spectra in Figure 53 a "shoulder" can be observed at around 380 nm, although it is partially overlapped by the broader band on the left and the narrower peak on the right. This feature corresponds to the π - π^* transition (HOMO-LUMO) in the PEDOT backbone [188, 189], and the energy difference between these two states is referred to as the optical band gap. The absorption spectrum shown in Figure 53

also exhibits a valley going from 300 nm until 700 nm, indicating a lower absorption value in the visible spectrum. In this range, PEDOT is the dominant component of the polymer that absorbs light, while PSS does not show absorption peaks above ~ 300 nm [188]. This behavior makes the material suitable for application as a transparent conductive layer in solar panels, where the photoactive layer absorbs within this spectral range [190, 191]. Additionally, the large hump close to 800 nm is associated with intramolecular charge transfer [190] and the bipolaron subgap states [188, 190, 191], indicating the highly conductive nature of the samples. The broadening in this region is explained by additional weaker transitions arising from the lifting of degeneracies caused by polymer bending and macroionic interactions [188], and the oscillations observed at wavelengths above ~ 750 nm are attributed to substrate-related effects, as can be verified from Figure 52 (b).

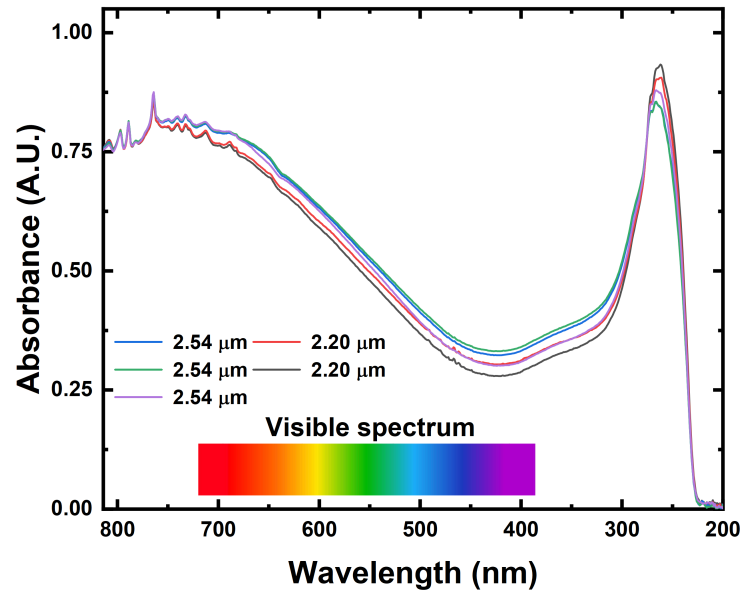


Figure 53 – Absorbance versus wavelength for PEDOT:PSS films deposited by drop-casting onto quartz substrates, with thicknesses of 2.20 μm and 2.54 μm . The sudden decrease in the curve around 220 nm is inherent to the experimental setup.

To estimate the optical energy gap of PEDOT:PSS, the Tauc method was employed, as described by Equation 4.5. The Tauc plot method has been applied to estimate the optical energy band gap in both inorganic [192, 193] and organic [191, 194, 182, 195] materials. In this model, the absorbance results (Figure 53) and the thickness of the PEDOT:PSS films were considered. The thicknesses of the films produced with volumes of

20 μL (2.20 μm) and 40 μL (2.54 μm) plus the thickness of the quartz substrate (2.4 mm) were considered as the optical path length in the Tauc equation. Internal reflections along the optical path were not taken into account. The following equation presents the Tauc model used to determine the optical energy gap

$$\alpha h\nu = \gamma (h\nu - E_g)^n, \quad (4.5)$$

where α is the absorption coefficient, ν is the frequency of the incident photon, h is Planck's constant, γ is a constant, and E_g is the bandgap energy. The absorption coefficient α accounts for both the sample thickness and its absorbance as follows:

$$\alpha = \frac{2.303}{t} A.$$

The exponent n in equation 4.5 takes the value of $n = 1/2$ for direct bandgap transitions and $n = 2$ for indirect bandgap transitions. Direct transitions are associated with the absorption of a photon by an electron, where the electron jumps from the HOMO level to the LUMO energy level. In an indirect transition, both a photon and a phonon are absorbed by the electron in order to realize the energy level transition. In this process, a phonon is absorbed to conserve the momentum. A schematic representation is shown in Figure 54.

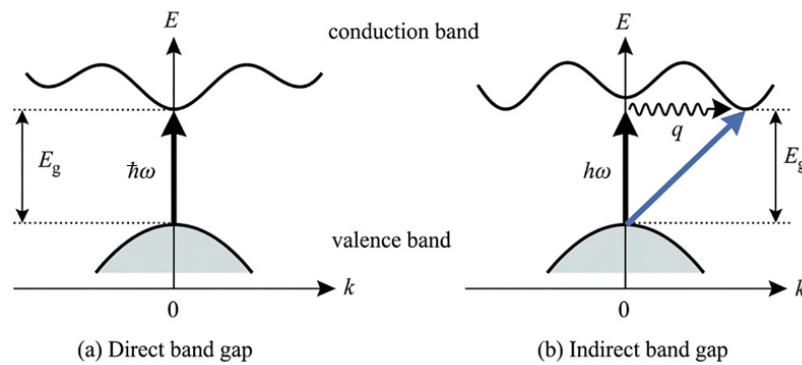


Figure 54 – Direct and indirect band gap representation. The vertical arrow represents the photon absorption process, while the wavy arrow in the right size denotes the absorption or emission of a phonon. Image adapted from [196].

Therefore, in amorphous polymers such as the organic semiconductors, the indirect bandgap value is expected to be smaller than the direct bandgap [196, 182].

The Tauc method, which consists of extrapolating the linear portion of the $(\alpha h\nu)^2$ curve to its intersection with the photon energy axis, was used to estimate the optical band gap energy. From the estimation presented in Figure 55 (a) and (b), the pristine PEDOT:PSS film showed optical band gap energies of 2.55 eV and 2.44 eV for the direct transition, and 2.00 eV and 2.06 eV for the indirect transition. The obtained values lie within the range reported for organic semiconductors, making them suitable for application in photovoltaic devices [197].

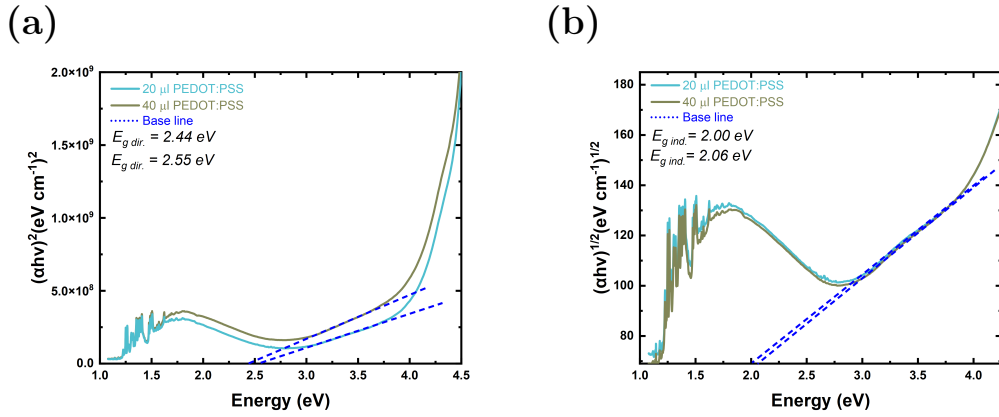


Figure 55 – Tauc plots of the pristine PEDOT:PSS film. Panel (a) represents the direct transition and (b) indirect transition, this results are used to estimate the optical band gap energies

The optical band gap value estimated for our sample lies within the expected range for organic semiconductors and shows good agreement with previous studies on PEDOT:PSS, both with experimental results obtained using the Tauc plot method and with simulation methods based on Time-Dependent Density Functional Theory calculations [188, 182, 98]. It is worth noting that several factors can influence the optical band gap energy of organic semiconductors, including doping, synthesis route, film growth technique, and solution concentration [198].

4.2.5 Ferromagnetic Resonance - PEDOT:PSS

Similar to polyaniline, PEDOT:PSS demonstrates excellent stability and versatility and has been extensively studied for numerous scientific and technological applications [66, 67, 199, 69, 8]. Using the drop-casting technique described earlier, we fabricated the PEDOT:PSS/YIG heterostructure employed in the investigation of spin-to-charge current conversion phenomena. Figure 56 (a) presents a schematic illustration of the

heterostructure used in this work. The drop-cast PEDOT:PSS layer converts the spin current into a charge current by means of the ISHE [200, 201].

As a spin current generator, we used single-crystal YIG films with a thickness of $6\ \mu\text{m}$ grown by liquid-phase epitaxy on a 0.5-mm-thick (111)-oriented gadolinium gallium garnet (GGG) substrate. The investigated samples were cut from the same GGG/YIG wafer into rectangular shapes of $2.0 \times 4.0\ \text{mm}^2$, and all measurements were carried out at room temperature.

Figure 56 (b) shows the current–voltage (I – V) characteristic curve obtained using the pseudo-four-probe technique for the structure depicted in Figure 56 (a). This technique is commonly employed for transport measurements in small samples, in which the voltage and current leads are connected separately but share the same contact pad. Because the contact resistances in our samples are negligible, they do not introduce significant errors in the resistance measurement compared with the conventional four-probe technique. The linear I – V behavior observed for the PEDOT:PSS/YIG structure in Figure 56 (b) confirms the formation of ohmic contacts between the electrodes.

For the FMR microwave excitation and spin-to-charge current conversion measurements, the sample with electrodes, as illustrated in Figure 56 (a), was mounted on the tip of a PVC rod and inserted into a small aperture in the back wall of a short-circuited waveguide, positioned at the maximum of the rf magnetic field and at a node of the electric field. No microwave resonator was used.

The static magnetic field (H) and the microwave field (h_{rf}) were applied in the plane of the film and maintained perpendicular to each other while the sample was rotated. In this configuration, the angular dependence of both the FMR spectra and the ISHE voltage can be measured. Field-swept spectra of the derivative of the microwave absorption (dP/dH) were obtained by modulating the magnetic field at 1.2 kHz and using lock-in detection.

Figure 56 (c) shows the FMR absorption spectrum of the PEDOT:PSS/YIG bilayer. The spectra were recorded with the magnetic field applied in the film plane, perpendicular to the long axis of the sample, at a microwave frequency of 9.4 GHz and an input power of 80 mW. The discrete resonance lines correspond to standing spin-wave

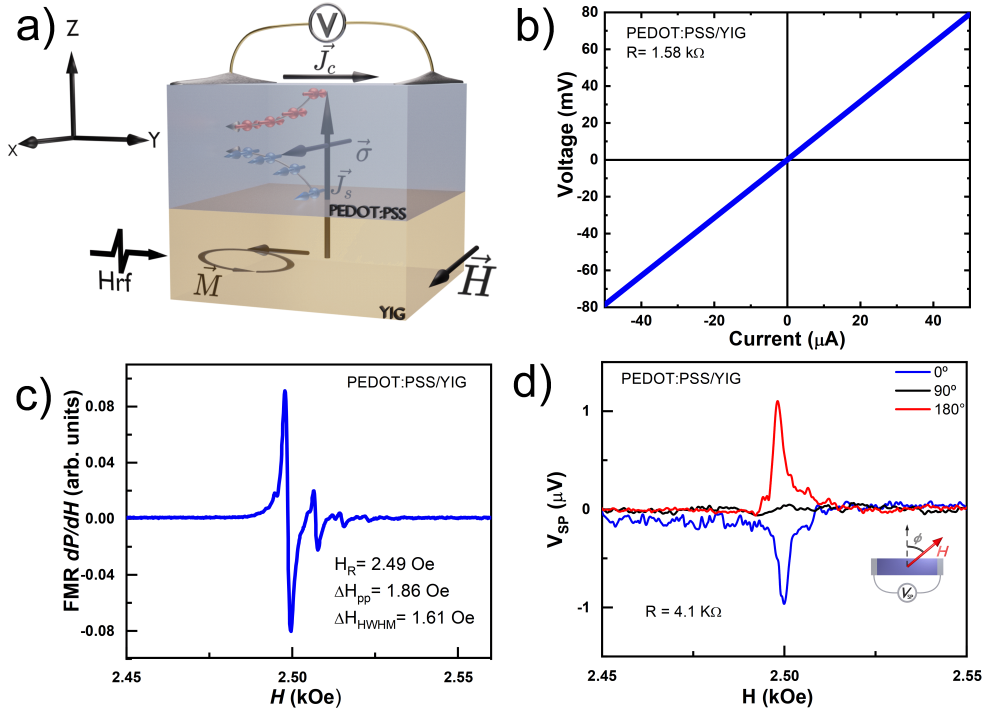


Figure 56 – Panel (a) shows a schematic illustration of PEDOT:PSS/YIG structure and the silver electrodes used to measure the dc voltage arising from spin-to-charge current conversion. (b) I – V curve demonstrating the ohmic behavior of the contacts. Panel (c) shows a typical field-swept FMR microwave absorption derivative spectrum of PEDOT:PSS/YIG sample measured at a frequency of 9.4 GHz. Panel (d) shows the field dependence of the ISHE voltage in PEDOT:PSS/YIG sample for three different in-plane angles, as indicated in the inset (0° , 90° , and 180°).

modes, with the most intense line associated with the uniform precession mode.

During the FMR process, the spin current generated by the microwave-driven FMR of YIG film is injected through the PEDOT:PSS/YIG interface. By means of the ISHE in PEDOT:PSS, the spin current is converted into a charge current and electrically detected as a dc voltage.

The contact between the polymer and the YIG layer induces linewidth broadening in the microwave absorption spectra. This effect, driven by spin pumping, results from angular momentum transfer via a pure spin current injected from the YIG into the PEDOT:PSS layer. By analyzing this broadening, we estimate the real part of the effective spin-mixing conductance, $g_{eff}^{\uparrow\downarrow}$, of the interface using the following relation:

$$g_{eff}^{\uparrow\downarrow} = (4\pi M t_{coh} / \hbar \omega) \delta H, \quad (4.6)$$

where ΔH is the additional linewidth broadening, $\omega = 2\pi f$, and λ is the coherence

length of the YIG film [170]. Using a linewidth value of $\Delta H_{\text{HWHM}}^{\text{YIG}} = 0.31$ Oe for the bare YIG [28] and measuring the additional linewidths, we found $g_{\text{PEDOT:PSS/YIG}}^{\uparrow\downarrow} = 1.46 \times 10^{18} \text{ m}^{-2}$. Comparing this value to that of Pt, we obtained $g_{\text{PEDOT:PSS/YIG}}^{\uparrow\downarrow} = 1.46 g_{\text{Pt/YIG}}^{\uparrow\downarrow}$. This result reveals a highly efficient spin transfer across the interface, as the $g_{\text{eff}}^{\uparrow\downarrow}$ of the PEDOT:PSS/YIG system is of the same order of magnitude as that of Pt/YIG, where Pt is a heavy metal and a well-known spin sink [12, 28]. This conclusion is further corroborated by the SSE electrical detection of the spin current converted within the PEDOT:PSS layer, as shown in Figure 57.

4.2.6 Spin Seebeck Effect - PEDOT:PSS

SSE measurements were performed by applying a perpendicular thermal gradient (∇T) via a Peltier module, as illustrated schematically in Figure 57 (a) for the PEDOT:PSS/YIG bilayer. The thermal gradient generates a spin current (J_s) in the YIG layer, which is injected across the interface and converted into a charge current within the polymer, being detected as an ISHE voltage (V_{SSE}). Figure 57 (b) displays V_{SSE} as a function of the applied magnetic field ($\mathbf{H}$) for several constant temperature differences (ΔT). This setup allowed the direct observation of spin-to-charge conversion within the PEDOT:PSS layer under both positive and negative gradients ($\pm \nabla T$). Since $V_{\text{SSE}} \propto J_s \propto \nabla T$ [171], reversing ∇T inverts the spin current direction and the voltage sign, resulting in the linear behavior depicted in Figure 57 (c), where each point represents the mean saturation V_{SSE} for a given ΔT . Additionally, reversing $\mathbf{H}$ inverts the voltage sign due to the reversed spin polarization. Notably, the measured V_{SSE} values are of the same order of magnitude as those of a Pt/YIG heterostructure [12]. Finally, due to the high stability of the signal at saturation, the associated error bars are smaller than the data symbols in Figure 57 (c and d).

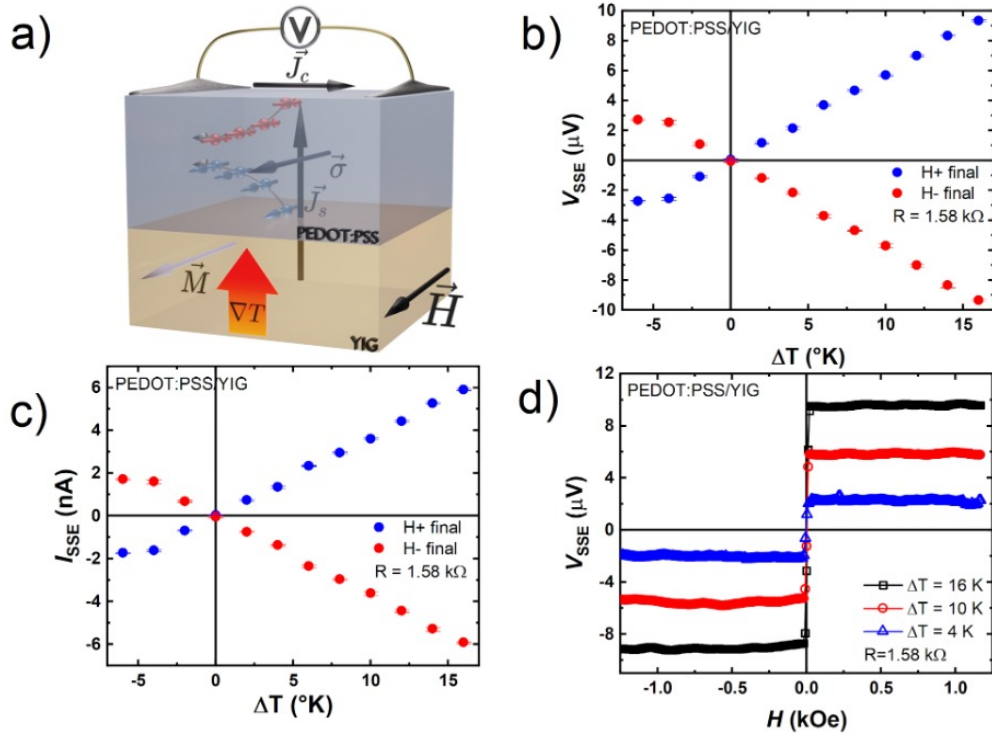


Figure 57 – Panel (a) shows a Schematic illustration of PEDOT:PSS/YIG bilayer sample. (b) Dependence of the spin Seebeck effect (SSE) voltage measured in PEDOT:PSS layer on the temperature difference (ΔT) for the PEDOT:PSS/YIG bilayer, under an applied magnetic field of $H = 1.0 \text{ kOe}$ for two opposite field directions. (c) Dependence of the SSE current on ΔT for PEDOT:PSS/YIG sample, obtained by normalizing the measured voltage by the resistance of PEDOT:PSS layer. (d) Magnetic field dependence of the dc ISHE–SSE voltage in PEDOT:PSS/YIG sample, measured for several temperature differences (ΔT).

The spin current injected across the PEDOT:PSS/YIG interface is governed by the equation:

$$J_s = -C_S \rho g_{\text{eff}}^{\uparrow\downarrow} \nabla T. \quad (4.7)$$

Here, $g_{\text{eff}}^{\uparrow\downarrow}$ is the effective spin-mixing conductance at the interface, and ρ is a parameter accounting for the finite thickness of the YIG layer. The coefficient $C_S = 3.3 \times 10^{-24} \text{ erg} \cdot \text{cm} \cdot \text{K}^{-1}$ depends on the temperature, applied magnetic field intensity, and intrinsic properties of the YIG film. The thermal gradient is defined as $\nabla T = \Delta T / t_{\text{sample}}$, with $t_{\text{sample}} \cong 0.05 \text{ cm}$ representing the total sample thickness, including the GGG substrate [171, 173].

The measured voltage (V_{SSE}) is intrinsically linked to the spin current (J_s) through

the following relation [171]:

$$V_{\text{SSE}} = F_P R w (2e/\hbar) \lambda \theta_{\text{SH}} \tanh\left(\frac{t_{\text{PEDOT:PSS}}}{2\lambda}\right) J_s, \quad (4.8)$$

where R , w , $t_{\text{PEDOT:PSS}}$, λ , and θ_{SH} correspond to the resistance, width, thickness, spin diffusion length, and spin Hall angle of the PEDOT:PSS layer, respectively. The geometric parameter $F_P \cong 2/3$ corrects for the surface fraction of the sample effectively covered by the Peltier heater used to apply ∇T . Based on the SSE measurements for the PEDOT:PSS/YIG bilayer, we can determine θ_{SH} by extracting the slope from a linear fit of V_{SSE} as a function of ΔT . This linear relationship takes the form:

$$V_{\text{SSE}} = P \theta_{\text{SH}} \Delta T, \quad (4.9)$$

where P is a proportionality coefficient comprising the system's geometric and intrinsic constants:

$$P = F_P R w (2e/\hbar) \lambda \tanh\left(\frac{t_{\text{PEDOT:PSS}}}{2\lambda}\right) \left[C_s \rho g_{\text{eff}}^{\uparrow\downarrow} / t_{\text{Sample}} \right]. \quad (4.10)$$

By substituting the expression for J_s into the V_{SSE} equation, we can derive the system parameters. From the linear fit shown in Figure 57 (b), we extract a slope of $P \theta_{\text{SH}} = (5.72 \pm 0.1) \times 10^{-7}$ V/K. To calculate the proportionality constant P , we utilized the following experimental parameters: $e = 1.6 \times 10^{-19}$ C; $R = 1.58$ k Ω ; $w = 0.3$ cm; $t_{\text{PEDOT:PSS}} = 2.20 \times 10^3$ nm; $g_{\text{PEDOT:PSS/YIG}}^{\uparrow\downarrow} = 1.46 \times 10^{18}$ m $^{-2}$; $\rho \approx 1$ (since $t_{\text{YIG}} \gg \lambda$) [173]; and a spin diffusion length for PEDOT:PSS of $\lambda \cong 180$ nm [201, 97]. This evaluation yields $P = -1.664 \times 10^{-2}$ V/K. Consequently, we obtained a spin Hall angle of $\theta_{\text{SH}} = (3.43 \pm 0.06) \times 10^{-5}$. This value agrees well with the spin Hall angles reported for similar polymeric materials [202, 12, 172]. While the SSE technique inherently carries uncertainties associated with experimental conditions and internal temperature gradients [203, 204, 178], our direct SSE measurements provide a reliable estimate of the spin Hall angle in the PEDOT:PSS layer, demonstrating that it is on the same order of magnitude as that of Pt.

In summary, we have investigated the spin current transport in the YIG/PEDOT:PSS heterostructure by probing three interconnected physical phenomena: (i) the thermal generation of a pure spin current via the spin Seebeck effect, (ii) the spin injection across the YIG/PEDOT:PSS interface, and (iii) the spin-to-charge current conversion governed by the inverse spin Hall effect directly within the PEDOT:PSS layer. These results clearly establish the viability of this polymer as an active spintronic material. Considering its

accessible synthesis routes, straightforward doping processes, high flexibility, and stable optical properties, PEDOT:PSS emerges as a highly promising candidate for spintronic applications, opening new avenues in the field of organic spintronics.

4.3 Tin Telluride Results and Discussions

In this section, the MBE sample growth procedure is presented and described. In addition, a set of characterization techniques was employed to investigate the structural, morphological, vibrational, and magnetic properties of the samples, as well as their spin-to-charge conversion behavior. The structural analysis included in situ RHEED during growth, AFM imaging, and Raman spectroscopy. High-resolution XRD with 2θ scans, X-ray reflectivity (XRR), rocking curve, and reciprocal-space map reconstruction were carried out to assess the crystalline quality and strain state of the epitaxial SnTe layers. Finally, FMR and SP-FMR measurements were performed to evaluate the spin-to-charge conversion efficiency.

4.3.1 SnTe Sample Growth by MBE

This section is dedicated to the molecular beam epitaxy (MBE) growth details of samples containing the topological crystalline insulator SnTe. All samples discussed in this section were grown during my period as a visiting scholar under the Brazilian sandwich doctorate program sponsored by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES). The growth was carried out in the research group of Professor Cui-Zu Chang at the Department of Physics of Penn State University in Pennsylvania, USA.

The samples involve the epitaxial growth of the crystalline topological insulator tin telluride (SnTe) on insulating indium phosphide (InP) substrates, whose surface normal is oriented along the (111) direction. The growth was carried out using an EVO-50 III-N MBE system (Instrument DOI: 10.60551/jb0f-5784), and a RHEED system from STAIB Instruments equipped with a kSA 400 RHEED analysis software package was used to monitoring the real-time epitaxial growth. Since SP-FMR experiments require a ferromagnetic material (FM) in contact with SnTe, a layer of Permalloy was grown on top of the SnTe layer. In order to construct a pristine topological insulator/ferromagnetic

material (TI/FM) interface, the Py growth was performed *in situ* over the SnTe layer. In this way, all layers in our samples were grown *in situ* by MBE.

The InP(111) substrate was mounted on the sample holder, as shown in Figure 21. The InP(111) substrate was then outgassed inside the MBE chamber in order to remove volatile contaminants such as oxides and water. The RHEED pattern of the as loaded InP(111) substrate is initially poor, since the InP surface absorbs contaminants when exposed to the ambient environment. For this reason, a well-controlled outgassing procedure is required before initiating the SnTe growth. The substrate temperature is slowly increased from 250°C to 400°C in small steps while the RHEED pattern is continuously monitored. During the outgassing stage, the chamber pressure is around 2×10^{-9} Torr, with fluctuations occurring due to gas molecules being released from the substrate as the temperature increases. A rapid temperature increase must be avoided because indium decomposition may occur at the surface, leading to the formation of 3D islands [205, 206]. This effect is clearly reflected in the RHEED pattern as stripes with different widths and the appearance of dots. The outgassing must be stopped when the RHEED pattern becomes uniformly composed by sharp stripes before a complete degradation of the pattern. This procedure was conducted with the MBE chamber pressure below 10^{-10} Torr, avoiding an excess of particles ejected from the substrate contaminating the chamber.

At the end of the outgassing procedure, the Te source is opened to create a Te-rich environment. During outgassing, some indium atoms decompose at the surface and form islands, which is undesirable for the subsequent epitaxial growth. To minimize this effect, the Te flux reacts with the indium, forming InTe. Since the substrate temperature is 400°C (well above the Te evaporation temperature) only the Te that reacts with indium is incorporated to the substrate surface, preventing the formation of a Te layer. This surface stabilization mediated by Te lasts for approximately 10 minutes. After this treatment, the substrate temperature is reduced to approximately 280°C, which is the SnTe growth temperature. Before starting the SnTe deposition, the Te treatment is repeated for an additional 10 minutes at this temperature.

The growth was carried out using a Knudsen thermal effusion cell as the source for tin (Sn) and tellurium (Te). The effusion cell temperatures for Sn and Te were 1050°C and 250°C, respectively, while the substrate temperature was set to 280°C. These temperatures

resulted in a deposition ratio of $0.0417 \text{ \AA/s}$. Based on the deposition flux and AFM thickness measurements, the thickness of the SnTe layers was estimated in SnTe unit cells (u.c.), where $1 \text{ u.c.} \approx 0.68 \text{ nm}$ [207]. Samples with SnTe thicknesses were grown with of 5, 8, 10, 15, 20, 30, and 38 u.c, where the SnTe deposition ratio is around 0.5 u.c/min . Figure 58 illustrates the different heterostructures produced.

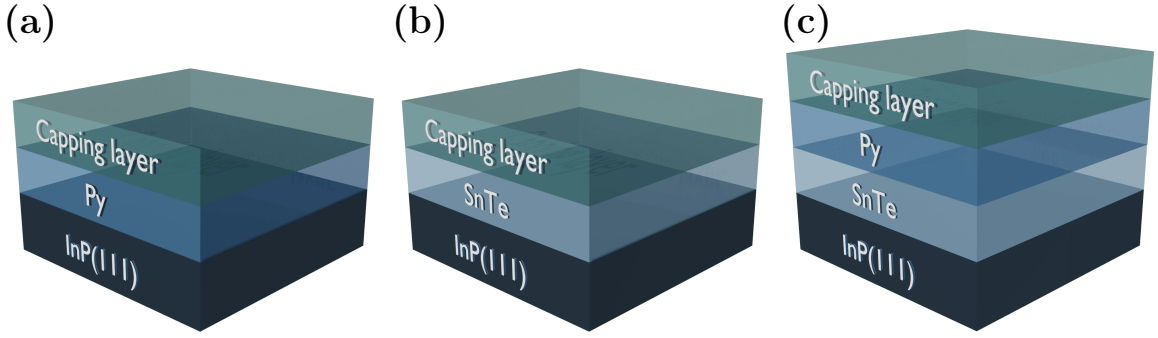


Figure 58 – Different heterostructures produced by MBE and investigated in this work.

At the beginning of the SnTe growth, the RHEED pattern is initially poor, indicating a 3D growth mode. After about 10 minutes, the RHEED pattern becomes sharp and well defined, signaling the transition to a 2D epitaxial growth regime [208]. The growth pressure during the SnTe deposition is approximately $8.78 \times 10^{-10} \text{ Torr}$.

To perform the SP-FMR experiments, a magnetic layer is required. In this sense, a 20 nm Permalloy (Py) ($\text{Ni}_{80}\text{Fe}_{20}$) layer was grown on top of SnTe in all samples. It is important to mention that this magnetic layer was grown *in-situ* over the SnTe layer at room temperature. The Py deposition ratio was performed with an electron-beam voltage of 7 kV and a current of 36 mA and sample was rotated at 4 rpm during the process. The MBE chamber pressure during the deposition was $9 \times 10^{-11} \text{ Torr}$. In this way, we have a pristine, contamination-free, and well-defined interface between the topological insulator and the magnetic layer. All samples were covered with a nominal thickness of 10 nm Te (or Al, when specified) capping layer to avoid oxidation of the magnetic layer. The capping layer was also grown at room temperature. The heterostructure in Figure 58 (a) was used to determine the crystalline phase, while Figure 58 (c) shows the complete sample used in the spintronic measurements.

An electron beam energy of 15 keV and 1.55 A was employed to the RHEED to monitor the InP substrate, the SnTe, and the Py surfaces. From Figure 59, it is possible

to see well-defined streaks for the SnTe surface after the growth, which is characteristic of a smooth and 2D surface with a low degree of imperfections or island formation. These lines are a typical indication of the epitaxial growth of a textured SnTe layer, in which the SnTe tends to follow the crystallographic ordering of the substrate.

In Figure 59 (b), the diffuse background together with the presence of rings indicates that the Py layer is usually related to a polycrystalline phase. This is expected, as the Py layer is grown at room temperature. This result will be rediscussed in the HRXRD section.

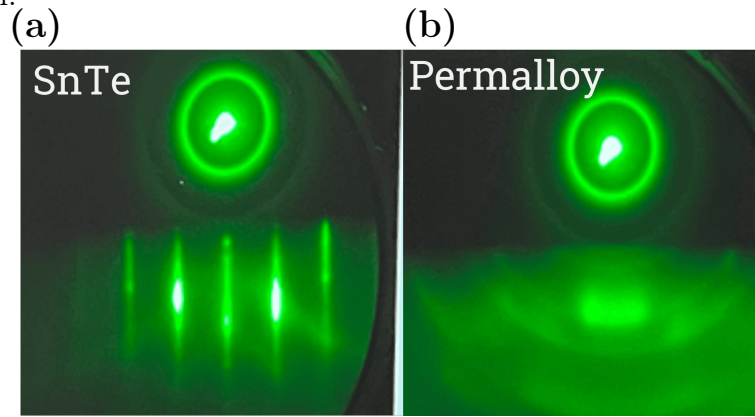


Figure 59 – Diffraction RHEED patterns captured after the growth of the SnTe and Py layers. The streaks lines observed on the SnTe surfaces (a) are a characteristic of epitaxial growth textured, in contrast, the blurred rings (b) suggest the polycrystalline phase of the Permalloy layer.

4.3.2 Surface Roughness Analysis by AFM

The surface morphology of all samples were examined using an atomic force microscope model NT-MDT PX ULTRA operating in tapping mode under a low-humidity environment ($< 15\%$) at room temperature. The measurements were performed using a BudgetSensors Tap300Al-G probe. The cantilever has a typical force constant of 40 N/m and a resonance frequency of ≈ 300 kHz, with a length of 125 μm , width of 30 μm , and thickness of 4 μm . An aluminum reflective coating $\approx 30\text{nm}$ on the detector side enhances laser signal detection [209]. The topographic images and RMS surface roughness was taken with dimension of $2 \times 2 \mu\text{m}^2$ area.

Figure 60 (a) displays the surface of the SnTe layer without any capping layer. In the top-center region of the image, a bright triangular feature with a height of 3.496 nm (≈ 9.5 u.c.) can be observed. This triangular morphology is characteristic of SnTe grown

along the (111) direction, where the cubic rock-salt structure exhibits a trigonal symmetry in its surface projection [208, 57]. Some holes are also visible along the surface. These holes are characteristic of the natural desorption of Te, which inevitably occurs to some level during the growth [210, 211]. Figure 60 (c) shows the uniform and granular Te capping layer, covering the entire SnTe surface. The measured RMS roughness values of all surfaces are displayed in the Table 2.

Analysis of Figures 60 (b) and (d) reveals a clear change in the AlO_x surface morphology when the SnTe layer is located beneath the Py layer. This modification results in a threefold increase in the AlO_x surface roughness from 0.33 nm to 1.49 nm. The sample with SnTe present the AlO_x surface to evolve from small, uniform grains to larger agglomerates, with the lateral grain average size increasing from approximately 0.05 μm to 0.20 μm .

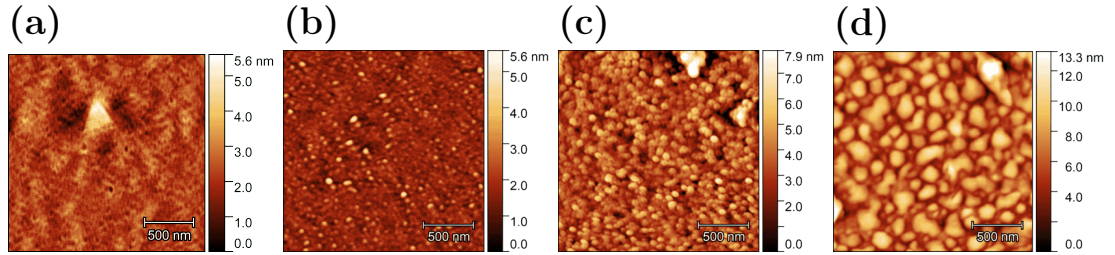


Figure 60 – Surface images taken by AFM of (a) SnTe/InP (81 u.c. $\approx$ 30 nm), (b) $\text{AlO}_x/\text{Py}/\text{InP}$, (c) Te/SnTe(54 u.c. $\approx$ 20 nm)/InP, and (d) samples $\text{AlO}_x/\text{Py}/\text{SnTe}$ (20 u.c. $\approx$ 7.36 nm)/InP.

Table 2 – Summary of AFM measurements for the samples shown in Figure 3.

Sample	Layer structure	SnTe thickness	RMS roughness (nm)
(a)	SnTe/InP	81 u.c. ($\approx$ 30 nm)	0.55
(b)	$\text{AlO}_x/\text{Py}/\text{InP}$	–	0.33
(c)	Te/SnTe/InP	54 u.c. ($\approx$ 20 nm)	0.58
(d)	$\text{AlO}_x/\text{Py}/\text{SnTe}/\text{InP}$	20 u.c. ($\approx$ 7.36 nm)	1.49

4.3.3 Raman Spectroscopy of SnTe

Raman measurements were conducted using a Renishaw InVia micro-Raman system equipped with a 514.5 nm argon laser. A 100 $\times$ objective lens (N.A. 0.75) was used to focus the laser producing a spot size of approximately 1 μm^2 on the sample surface. The spectra were acquired using 10 accumulations with an integration time of 10 seconds per

accumulation, while the laser power was maintained below 10 mW to avoid local heating and damaging effects.

Figure 61 (a) shows the Raman spectrum of the Te/SnTe(20 nm)/InP sample. The longitudinal optical (LO) phonon mode associated with the SnTe (222) orientation and A_{1g} symmetry is observed at 125.33 cm^{-1} . In addition, the transverse optical (TO) phonon mode with E_{TO} symmetry is detected at 142.04 cm^{-1} . These peaks correspond to the most intense phonon modes of SnTe. Their positions are in excellent agreement with those previously reported in the literature [212, 213, 214].

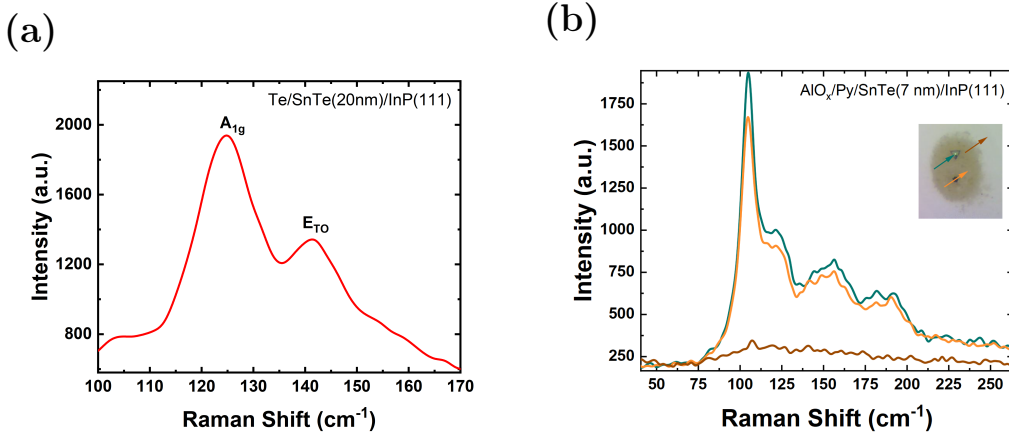


Figure 61 – Panel (a) shows the Raman spectra highlighting the most intense peaks associated with the SnTe(20 nm) layer. Panel (b) shows the Raman spectra of the $\text{AlO}_x/\text{Py}/\text{SnTe}(7 \text{ nm})/\text{InP}$ heterostructure near a surface defect. The inset in (b) shows a defect on the surface where three Raman spectra were collected. The colors of the circles correspond to the colors of the respective Raman curves.

The Raman spectra of the $\text{AlO}_x/\text{Py}/\text{SnTe}(7 \text{ nm})/\text{InP}$ sample acquired at different laser focus positions is shown in Figure 61 (b). The green curve corresponds to a measurement performed on a triangular crystallite (SnTe feature, as discussed in AFM section) located inside a defect in the Py and AlO_x layers. From Figure 61 (b), it is possible to observe that the peak intensities decrease when the measurement is taken outside the crystallite. This behavior can be explained by the presence of additional layers on top of the SnTe, which partially mask the Raman signal of the SnTe layer. Another noticeable feature is the pronounced increase of the peak located near 104 cm^{-1} of $\text{AlO}_x/\text{Py}/\text{SnTe}(7 \text{ nm})/\text{InP}(111)$, when compared with the Te/SnTe(20 nm)/InP sample. This peak is associated with an optical phonon mode from the Brillouin-zone boundary at the W point. Its appearance is attributed to first-order Raman scattering that becomes

allowed due to disorder in the SnTe crystal structure [212].

A comparison between the Raman spectra of SnTe films with thicknesses of 20 nm and 7 nm is shown in Figure 62 (b). The peaks labeled M2 and M3 are associated with phonons at the W and L points of the Brillouin zone, while the M4 peak originates from a second-order two-phonon scattering process. By analyzing the M3 peak, which corresponds to the longitudinal optical A_{1g} mode, it is observed that the thinner film exhibits a Raman shift toward lower frequencies, from 125.3 cm^{-1} to 121.6 cm^{-1} . This anomalous behavior is well known and has been previously reported for SnTe under hydrostatic pressure conditions [212]. Specifically, for hydrostatic pressures ranging from 0 up to approximately 1.5 GPa, the phonon frequency shifts toward lower values. Above this critical pressure, the phonon frequency shifts toward higher values, following the conventional hardening behavior.

This phenomenon, reflects its anomalous response to compressive strain such that an increase in strain induces a reduction in the phonon frequency. In the present case, the compressive strain induced by the InP substrate during epitaxial growth leads to a downshift of the M3 mode for thinner SnTe films. In thin-film systems, where strain can be induced both by lattice mismatch with the substrate, this behavior becomes particularly relevant. Thinner films, which typically experience higher internal strain or enhanced confinement effects, tend to exhibit the M3 phonon mode at slightly lower frequencies when compared to thicker films or bulk SnTe.

The origin of this anomalous behavior is associated with a phonon instability at the Γ point of the first Brillouin zone and with the band inversion [212]. This phonon peak behavior is predicted to persist up to approximately 5.8 GPa, as estimated by density functional theory (DFT) calculations. Above this pressure, the system becomes stabilized, indicating the occurrence of a structural phase transition. This transition is accompanied by a change in the electronic phase, from a crystalline topological insulator to a semimetallic state [212].

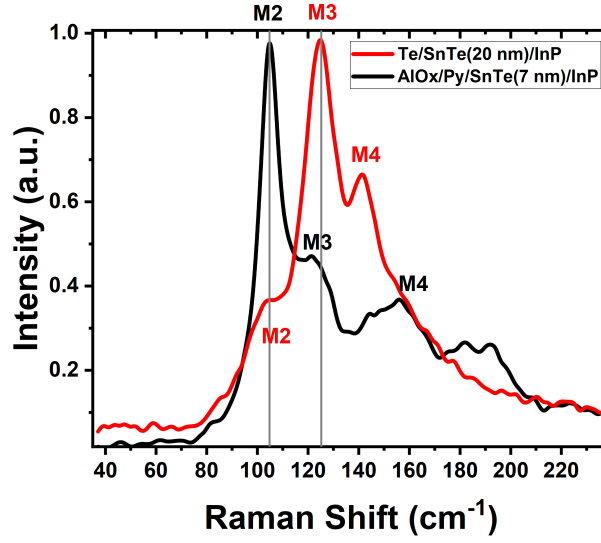


Figure 62 – Raman spectra of Te/SnTe(20 nm)/InP and AlO_x/Py/SnTe(7 nm)/InP are shown. The peaks labeled M2, M3, and M4 correspond to the most intense phonon modes. In particular, the M3 peak is the longitudinal optical (LO) mode with A_{1g} symmetry. Its shift toward lower frequencies as the film thickness decreases is an anomalous effect associated with the strain state of the layer and with the topological nature of this material.

In addition, an inversion of intensity between the M2 and M3 peaks is observed, along with a decrease in the M4 intensity as the thickness decreases from 20 nm to 7 nm. These results are in good agreement with previous experimental observations and DFT calculations reported in the literature [212]. From these results, we can infer that the SnTe(7 nm) layer is more compressed than the SnTe(20 nm) layer, following a behavior analogous to that observed for SnTe subjected to external hydrostatic pressure. Further discussion of the strain level will be presented in the following section.

4.3.4 High-Resolution X-Ray Diffraction

The structural characterization of the MBE grown Py/SnTe/InP(111) samples and the InP(111) substrate, were performed in a Bruker D8-discover diffractometer. To obtaining information about the crystalline structure and crystal orientation of the SnTe layer, High Resolution XRD measurements and $2\theta - \omega$ scans were performed. Reciprocal space mapping (RSM) was carried out on the SnTe layer and the InP(111) substrate using the symmetric (222) and asymmetric (224) orientations, respectively. In the symmetric measurements, the crystalline planes parallel to the substrate surface aligned along the

[111] direction of InP were analyzed. Also, the mosaic spread of the crystallites in the SnTe layer was investigated in the asymmetric measurements along the [224] direction. In addition, X-ray reflectivity (XRR) measurements were conducted to estimate the thickness, roughness and densities of the Tellurium, Aluminum oxide, Permalloy and SnTe layers.

High-resolution X-ray diffraction (HRXRD) measurements of the $\text{AlO}_x/\text{Py}/\text{InP}$ sample were performed to analyze the substrate quality and the phase state of the Py layer. From Figure 63 (a), it is possible to confirm that only peaks related to the (111) family of InP planes are present. Although the Py layer was grown with the substrate at room temperature, a weak peak at $2\theta \approx 77^\circ$ is observed. This weak peak is typically associated with the NiFe (220) orientation [215, 216], indicating a low degree of plane ordering of this phase. The presence of this peak may be attributed to the deposition of a secondary NiFe phase caused by the different evaporation temperatures of Ni (1453°C at a vapor pressure of 10^{-3} mBar [217]) and Fe (1180°C at a vapor pressure of 10^{-3} mBar [218]) in the e-beam system. This difference in evaporation temperatures for each material can alter the effusion flux of each element, in a small level. As the effusion cell is unique, being made of permalloy ($\text{Ni}_{80}\text{Fe}_{20}$), the e-beam spot heats up. The permalloy is evaporated from this unique effusion cell, and the temperature at the e-beam spot can cause the separation of nickel and iron atoms, creating a secondary phase that is deposited. In this case, the lower evaporation temperature of Fe can lead to a higher Fe flux, increasing its relative content from 20% to nearly 50% during deposition, while the Ni flux remains comparatively lower. This phenomenon changes the stoichiometry of part of the deposited film from $\text{Ni}_{80}\text{Fe}_{20}$ to a more Fe-rich NiFe composition. Nevertheless, no peaks related to the $\text{Ni}_{80}\text{Fe}_{20}$ are detected, suggesting that the Py layer is in an amorphous state.

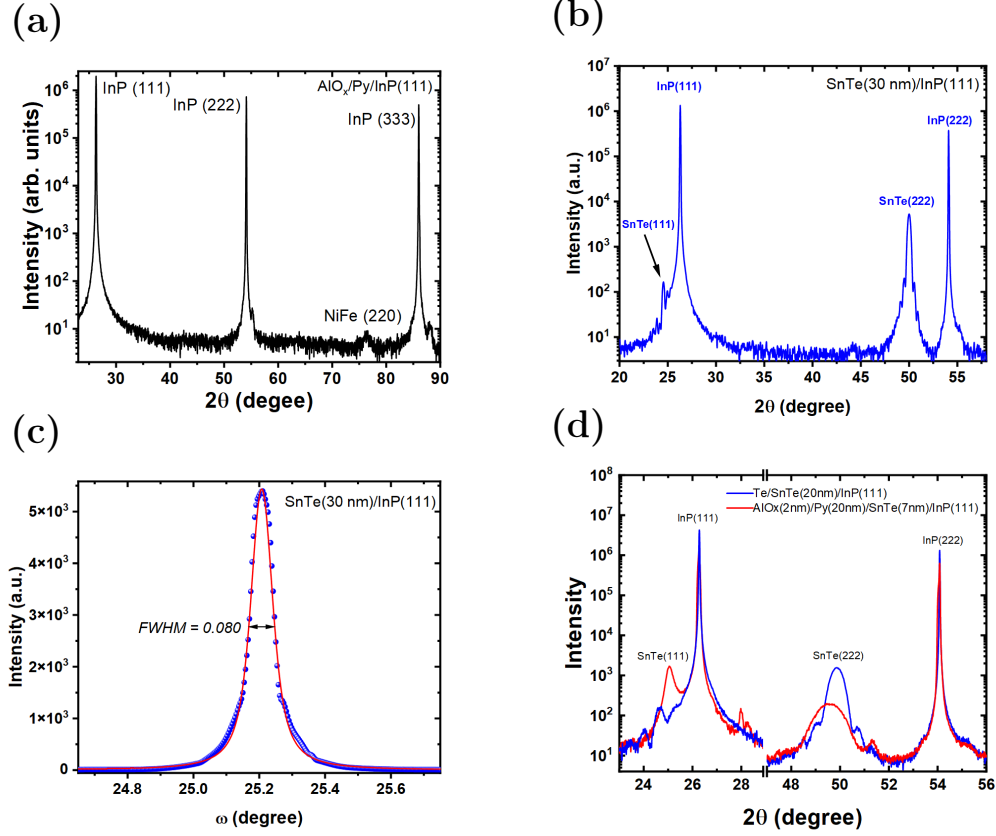


Figure 63 – The panel (a) exhibits a High-resolution X-ray diffraction (HRXRD) spectrum of the $\text{AlO}_x/\text{Py}/\text{InP}(111)$ sample, just peaks related to the (111) family of InP planes are present. Also, one peak related to NiFe (220) orientation is observed at $2\theta \approx 77^\circ$. The panel (b) shows $2\theta-\omega$ XRD scans for the $\text{SnTe}(30\text{ nm})/\text{InP}$ sample, exhibiting single-phase SnTe (111) layer with diffraction peaks corresponding to SnTe (111) near 25° and SnTe (222) around 50° . Laue fringes are observed for the SnTe (111) peak, indicating a high degree of coherence between the SnTe film planes and sharp interfaces. Panel (c) shows the rocking curve of the (222) reflection from the 30 nm SnTe layer, which exhibits a FWHM of 0.080° . In panel (d) the curves for one sample with and without the Py layer are plotted. The broadening of the SnTe (222) peak in the $\text{AlO}_x/\text{Py}/\text{SnTe}/\text{InP}$ sample is attributed to its reduced thickness.

The $2\theta-\omega$ XRD spectrum of the $\text{SnTe}(30\text{ nm})/\text{InP}$ sample shown in Figure 63 (b) exhibits a single phase for the SnTe layer, with peaks corresponding to the (111) family of planes, consistent with the substrate orientation. The SnTe (111) peak near 25° and the SnTe (222) peak around 50° are well defined and display Laue fringes. This is an indication of the a high degree of coherence between the SnTe film planes and the well-defined interface between the substrate and the SnTe layer. The right-side fringes of the SnTe (111) peak are not visible due to the overlap by the intense InP (111) peak.

Figure 63 (c) shows the rocking curve of the (222) reflection from the 30 nm SnTe layer. The curve exhibits a sharp single peak, which was fitted using a Voigt-like

profile. Rocking curves are commonly used to assess the mosaicity of epitaxial layers by estimating the full width at half maximum (FWHM), a parameter that is proportional to the defect density of the film, since narrower peaks indicate a lower density of crystalline defects. For 30 nm SnTe epitaxial layer, we obtained a FWHM of 0.080° , confirming the high crystalline quality of the film. For comparison, previously reported SnTe(111) films grown by MBE show FWHM values of 0.11° (≈ 25 nm) [219], 0.06° (≈ 15 nm) [208], and 0.09° (≈ 100 nm) [208]. Our result is in consonance with these reports, demonstrating that high-quality, large-area SnTe(111) epitaxial layers were successfully obtained.

The out-of-plane lattice constant $a_\perp$ for cubic SnTe was determined by applying Bragg's Law to the (222) peak of SnTe in the spectrum of Figure 63 (d) as follows

$$2d_{hkl} \sin \theta = n\lambda.$$

In this case, $n = 1$, corresponding to first-order diffraction. The wavelength (λ) of the XRD Cu source is $1.5406 \text{ \AA}$. For a cubic lattice, the interplanar spacing is related to the out-of-plane lattice parameter by the following expression:

$$d_{hkl} = \frac{a_\perp}{\sqrt{h^2 + k^2 + l^2}}.$$

The SnTe diffraction peak analyzed has Miller indices $(hkl) = (222)$. The measured angle of the (222) diffraction peak is $\theta = 24.935^\circ$. With these values, the out-of-plane lattice parameter of the SnTe layers is estimated as:

$$a_\perp^{\text{SnTe}(20 \text{ nm})} = 6.330 \text{ \AA}$$

$$a_\perp^{\text{SnTe}(7 \text{ nm})} = 6.370 \text{ \AA}.$$

The reported lattice parameter of bulk SnTe is $6.318(3) \text{ \AA}$ [207]. Accordingly, for both samples, the out-of-plane lattice parameter is larger than the bulk value. This result corroborates the estimated values of the out-of-plane lattice parameter estimated here, where $a_\perp^{\text{SnTe}(7 \text{ nm})} > a_\perp^{\text{SnTe}(20 \text{ nm})}$, consistent with cubic elastic theory, in which in-plane strain induces an out-of-plane elastic response known as the Poisson effect [220]. The crystalline structure of the InP substrate (with lattice parameter $a = 5.86870 \text{ \AA}$)² induces an in-plane compressive strain in the SnTe lattice parameter $a_\parallel$ during growth. In response,

²The lattice mismatch between SnTe and InP is 7.5 %.

the out-of-plane lattice parameter $a_{\perp}$ increases. Figure 64 illustrates the compression constraint induced by the InP substrate on the SnTe layer. To verify these statements, we reconstructed the asymmetric reciprocal space map for these samples, obtaining an estimate of the in-plane lattice constant and, subsequently, the strained state of the SnTe layer. The results will be discussed in the **Asymmetric Reciprocal Space Mapping** section.

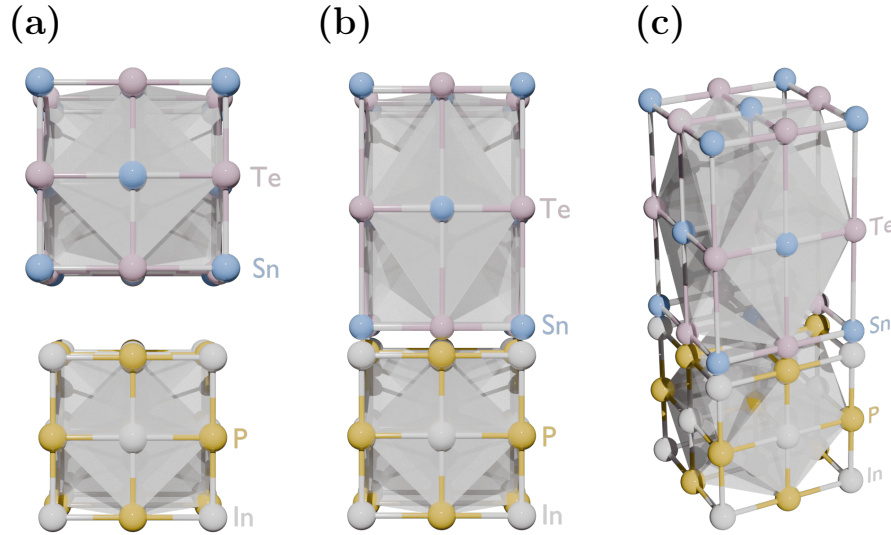


Figure 64 – Panel (a) shows the FCC unit cells of the InP substrate and SnTe. Panels (b) and (c) illustrate the in-plane compression of the SnTe lattice parameter, while the out-of-plane lattice parameter is expanded. This deformation of the SnTe unit cell increases the interplanar spacing, as confirmed by the peak positions in the XRD spectrum.

The spectra shown in Figure 65 shows a comparative between the SnTe(7 nm) and SnTe(20 nm) samples. As the capping layers are amorphous, they do not appear in the HRXRD spectrum. The broadening of the (222) diffraction peak in the SnTe(7 nm)/sample, shown in Figure 63 (c), is attributed to its smaller thickness when compared with the 20 nm SnTe layer. Additionally, the SnTe (222) peak position have a large angle for the SnTe(7 nm) than SnTe(20 nm). This result suggests that the thinner layer has a smaller in-plane lattice parameter, and consequently more strained than the SnTe (20 nm) layer and tends to approach the in-plane lattice parameter of InP substrate.

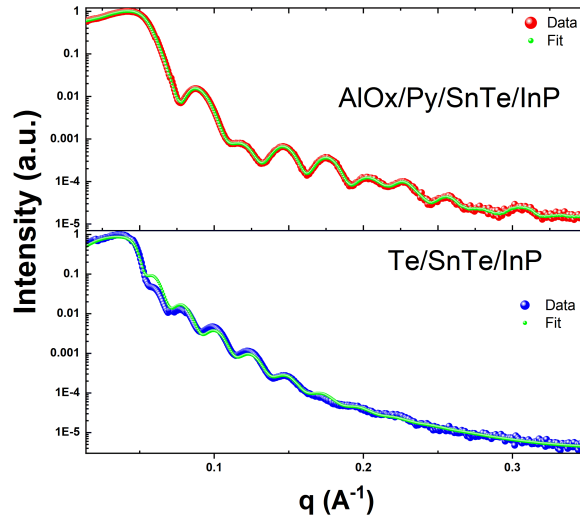


Figure 65 – X-ray reflectivity (XRR) measurement and fitting adjusts for SnTe(7 nm) and SnTe(20 nm) samples, the estimated parameters are shown in the Tables 3 and 4.

Figure 63 (d) shows the XRR spectra and the corresponding fitting results for the $\text{AlO}_x/\text{Py}/\text{SnTe}/\text{InP}$ and $\text{Te}/\text{SnTe}/\text{InP}$ samples. The density, thickness, and roughness of each layer in both samples are listed in Tables 3 and 4.

Layer	Density (g/cm^3)	Thickness (nm)	Roughness (nm)
Al	2.7 ± 0.2	8.3 ± 0.1	0.9 ± 0.2
Py	8.3 ± 0.1	12.0 ± 0.3	0.4 ± 0.1
SnTe	6.5 ± 0.2	8.9 ± 0.2	1.2 ± 0.3
InP	4.8 ± 0.1	–	0.5 ± 0.1

Table 3 – Structural parameters for $\text{AlO}_x/\text{Py}/\text{SnTe}/\text{InP}$ structure obtained by the curve adjust of the XRR data.

Layer	Density (g/cm^3)	Thickness (nm)	Roughness (nm)
Te	5.41843	2.04255	0.64704
SnTe	6.02317	22.2028	0.345
InP	4.81352	–	0.97127

Table 4 – Structural parameters for $\text{Te}/\text{SnTe}/\text{InP}$ structure obtained by the curve adjust of the XRR data.

4.3.4.1 Symmetric Reciprocal Space Mapping

To obtain information about the strain state of the SnTe films and their in-plane crystalline ordering, reciprocal space mapping (RSM) measurements were performed. The map shown in Figure 66 (a) illustrates that a symmetric configuration provides

information about lattice planes parallel to the substrate surface. In Figures 66 (b) and (c), the RSM spectra around the symmetric (222) reflection for the SnTe(20 nm)/InP and Py/SnTe(20 nm)/InP samples are presented. In both maps, two distinct diffraction peaks are observed, one associated with the InP substrate and the other with the SnTe film.

The well-defined centers of the SnTe (222) and InP (222) spots, highlighted in red, indicate a high degree of crystalline alignment and structural coherence in the SnTe layers. In Figure 66 (b), the SnTe (222) and InP (222) diffraction peaks lie on the $q_{\parallel} = 0$ axis, demonstrating that the SnTe (111) planes are perfectly aligned with those of the InP substrate, with no detectable tilt or miscut, thereby confirming the epitaxial relationship. Despite the evident epitaxy, the peak broadening observed in the 2θ - ω scans for Figures 66 (b) and (c) indicates the presence of in-plane crystallite misorientation, characteristic of mosaicity.

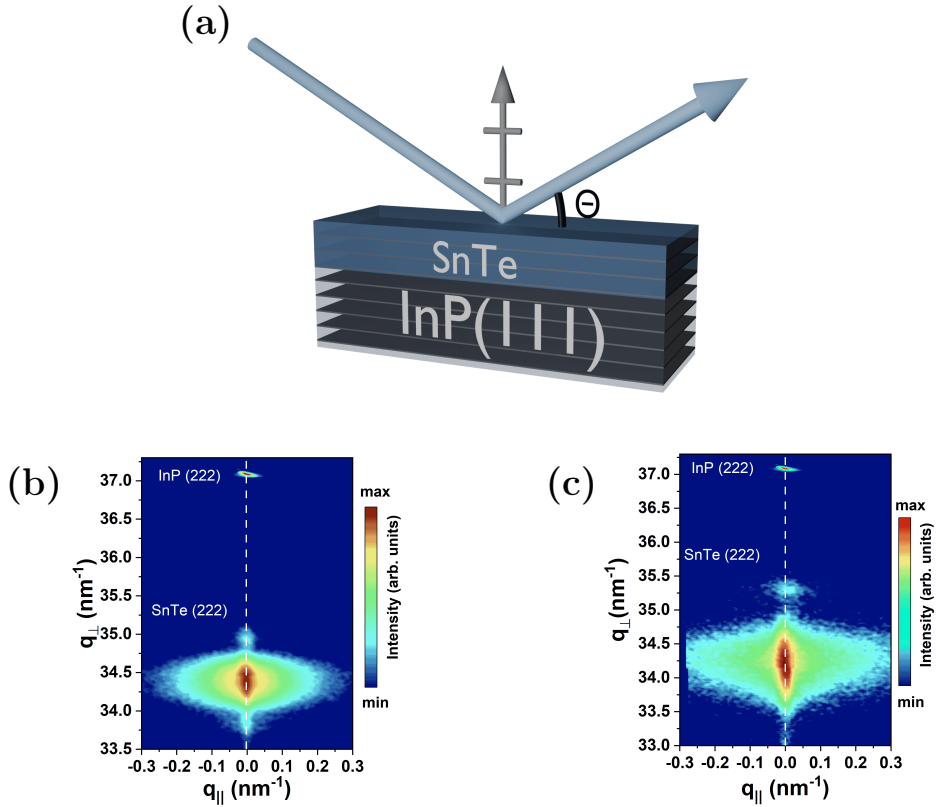


Figure 66 – (a) Schematic illustration of the planes examined in a symmetric RSM measurement. Panels (b) and (c) show the symmetric RSM reconstruction of the (222) planes for the SnTe(20 nm)/InP and Py/SnTe(7 nm)/InP samples, respectively. In both images, it is possible to observe slight oscillations along the $q_{\parallel} = 0$ line, which are characteristic signatures of Laue fringes.

4.3.4.2 Asymmetric Reciprocal Space Mapping

Figure 67 (a) exemplifies an asymmetric measurement providing information about a specific family of inclined planes relative to the substrate surface. From the RSM shown in Figure 67 (b), the relaxation state of the epitaxial SnTe layer relative to the InP substrate was evaluated by examining the relative position of the asymmetric (224) reflection. The in-plane lattice parameter of the 20 nm SnTe layer was estimated to be 5.951 Å, which is very close to the InP lattice parameter of 5.90 Å. The corresponding relaxation factor is 0.214, indicating that the 20 nm-thick SnTe layer remains largely strained and tends to follow the substrate lattice parameter. This result is consistent with the out-of-plane lattice parameter obtained using Bragg's law, which is larger than the bulk value of SnTe.

Figure 67 (c) shows peak broadening in both the 2θ - ω and ω directions in the RSM. This indicates an increased in-plane misorientation among crystallites (mosaicity) and a reduced coherence of the atomic plane spacing when compared to the Te/SnTe(20 nm)/InP sample. The broader ω scan for the Py/SnTe(7 nm)/InP sample evidences an increase in mosaicity. However, caution must be exercised when directly comparing these samples, as differences in layer stack and thickness inherently affect the mosaic spread and peak broadening in HRXRD, particularly for ultrathin films.

The asymmetric map in Figure 67 (c) exhibits significant ω broadening, which prevents a reliable determination of the in-plane lattice parameter. However, it is possible to notice that the region of highest intensity is closer to the dashed line connecting the InP (224) peak and the $q_{\parallel} = 0$ point when compared to Figure 67 (b). This indicates that the SnTe(7 nm) layer is more strained than the SnTe(20 nm) layer.

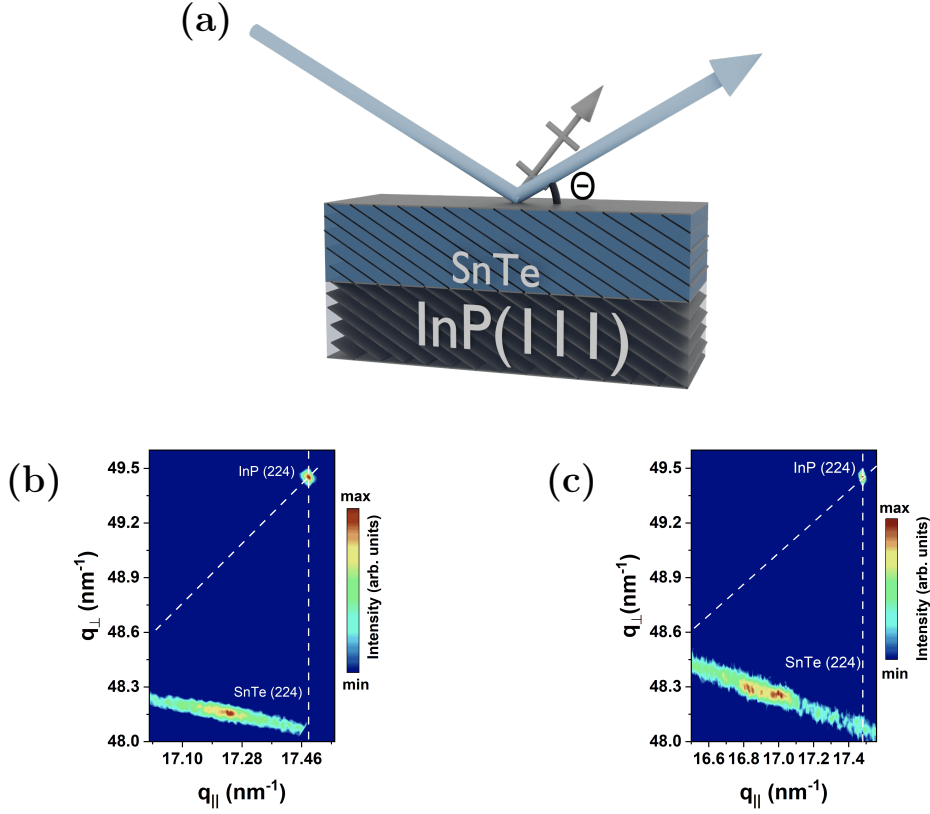


Figure 67 – (a) Schematic illustration of the planes examined in a asymmetric RSM measurement. Panels (b) and (c) show the asymmetric RSM reconstruction of the (224) planes for the SnTe(20 nm)/InP and Py/SnTe(7 nm)/InP samples, respectively. The inclined dashed line connects the InP(224) position to the origin of the reciprocal space.

4.3.5 Ferromagnetic Resonance

Using the FMR technique is possible to estimate the linewidth (ΔH), that is a fundamental parameter for investigating magnetic relaxation processes. In ferromagnet/normal-metal bilayers, variations in the linewidth enable the quantification of the spin pumping effect through the spin-mixing conductance ($g_{eff}^{\uparrow\downarrow}$). The precession of the magnetization injects a pure spin current into the adjacent non-magnetic layer, leading to a loss of angular momentum in the ferromagnet and, consequently, to an enhancement of the FMR linewidth.

For the FMR and SP-FMR experiments, a rectangular microwave cavity was used. The cavity was configured to operate in the transverse electric TE $_{102}$ mode, with a quality factor $Q \approx 2000$, at a frequency of 9.4 GHz. The static magnetic field H , generated by an Helmholtz coil setup, was kept in the film plane and orthogonal with the microwave field h_{rf} . The dP/dH versus magnetic field spectra, shown in Figure 68, were obtained using

lock-in detection using magnetic field modulation at 1.2 kHz. Both FMR and SP-FMR measurements were performed at room temperature. For this configuration, the sample was mounted at the tip of a PVC rod and positioned at a location inside the cavity where the rf electric field is minimized and the rf magnetic field is maximized. Under these conditions, galvanic effects driven by electric fields are avoided.

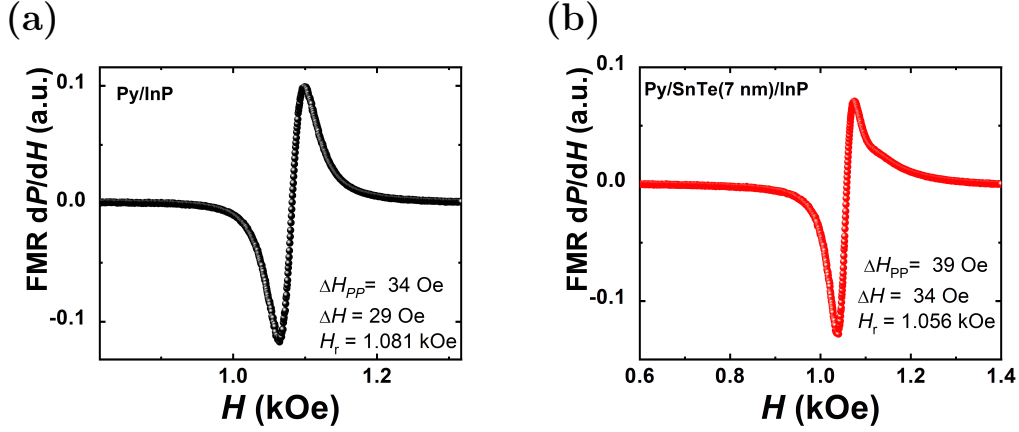


Figure 68 – Field-swept FMR microwave absorption derivative spectra for (a) Py/InP and (b) Py/SnTe(7 nm)/InP samples measured at an excitation frequency of 9.4 GHz, with an applied microwave power of 13.54 mW in a resonant cavity. Using the increase in linewidth, it is possible to estimate the spin mixing conductance. The resonance magnetic field is equal to 1.081 kOe and 1.056 kOe for (a) and (b) respectively.

From the spectrum shown in Figure 68 it is possible to estimate the half width at half maximum (HWHM) by the distance between the maximum and minimum points of the derivative spectra. The relation between them is shown as follows

$$\Delta H = \frac{\sqrt{3}}{2} \Delta H_{pp}.$$

The increase in the FMR linewidth observed for the Py/SnTe(7 nm)/InP sample allows the estimation of the effective spin-mixing conductance as follows

$$g_{\uparrow\downarrow}^{\text{eff}} = \frac{4\pi M t_{\text{Py}}}{\hbar\omega} \delta H,$$

where $4\pi M = 1.76$ kG is the saturation magnetization at room temperature of Py[[170], t_{Py} is the Permalloy thickness with a nominal value of 20 nm, $\hbar = 1.0545718 \times 10^{-34} \text{ J} \cdot \text{s}$ is the plank's constant, $\omega = 5.906 \times 10^{10} \text{ s}^{-1}$ is the angular resonance frequency using $f = 9.4$ GHz, and $\delta H = 5$ Oe is the additional linewidth caused by the spin pumping that is obtained by the dP/dH versus H spectrum [170]. With those values, the estimated³

³we used $1 \text{ G} \cdot \text{Oe} = 7.956 \times 10^{-3} \text{ T} \cdot \text{A/m}$ for the SI conversion.

value of the spin-mixing conductance is

$$g_{\text{eff}}^{\uparrow\downarrow} = 1.4 \times 10^{18} \text{m}^{-2}.$$

This value is of the same order of magnitude as that reported for Py/Pt interface, where Pt is known as one of the most efficient spin sinks [175, 174], and the well-known Ti Bi₂Se₃/YIG interface, in which the authors claim that the spin-to-charge current conversion is governed by the IREE [221]. Previous values of $g_{\text{eff}}^{\uparrow\downarrow}$ reported for sputtered SnTe/YIG interfaces are $g_{\text{eff}}^{\uparrow\downarrow} \approx 3 \times 10^{17} \text{m}^{-2}$ [222], about one order of magnitude lower than that obtained in this work. This enhancement may be attributed to the high-quality epitaxial growth of the SnTe layer and to the state-of-art structural quality of the in situ grown Py/SnTe interface, which favors more efficient spin injection under SP-FMR conditions.

Moreover, this $g_{\text{eff}}^{\uparrow\downarrow}$ value is of the same order of magnitude as that reported for the topological insulator Bi₂Se₃ with six quintuple layers grown on a YIG substrate, for which the authors claim that the spin-to-charge current conversion is governed by the IREE [221].

4.3.6 SP-FMR in SnTe

Using the same parameters and setup configuration employed in the previous section, we performed the SP-FMR measurements. The line shape $V(H)$ is composed of one symmetric term related with spin pumping spin-to-charge conversion and other asymmetric term is related to galvanomagnetic effects such as the anisotropic magnetoresistance (AMR) [40]. The voltage in function of the external magnetic field $\vec{H}$ measured between two electrodes at the edges of the sample is given by

$$V(H) = V_{\text{sym}} L(H - H_R) + V_{\text{asym}} D(H - H_R),$$

in which the $L(H - H_R)$ term is a symmetric Lorentzian function,

$$L(H) = \frac{\Delta H^2}{[(H - H_R)^2 + \Delta H^2]}$$

meanwhile $D(H - H_R)$ represents the antisymmetric Lorentzian,

$$D(H) = \frac{\Delta H (H - H_R)}{(H - H_R)^2 + (\Delta H)^2}.$$

Remembering that the FMR linewidth is denoted by ΔH . The voltage generated as function of the magnetic field angle ϕ can be fitted by the following expression

$$V(H, \phi) = [V_{CL}^{\text{sym}} L(H - H_R) + V_{CL}^{\text{asym}} D(H - H_R)] \sin 2\phi \sin \phi + V_{SP}^{\text{sym}} L(H - H_R) \cos \phi$$

where CL and SP subscript denotes *classical* and *spin pumping* origin of each term respectively. The classical term is related to anisotropic magnetoresistance or spin reification effects of the Py layer [40, 223]. Observe that $D(H) = 0$ when the amplitude of the magnetic field is equal to H_R .

Figure 69 (a) shows the dc voltage measured for $\phi = 0^\circ$ and 180° . For both angles, the data are fitted with excellent agreement using the above equation. Figure 69 (b) shows the symmetric part for both curves. The voltage peak inverts its sign when the magnetic field is inverted by 180° . This behavior is characteristic of the ISHE or the IREE, responsible to the spin-to-charge conversion [223]. The voltage $V_{SP}^{\text{peak}} = 1.71 \mu\text{V}$ is associated with the maximum value of spin-to-charge conversion process in SnTe and is achieved at an input power of $P_i = 110 \text{ mW}$.

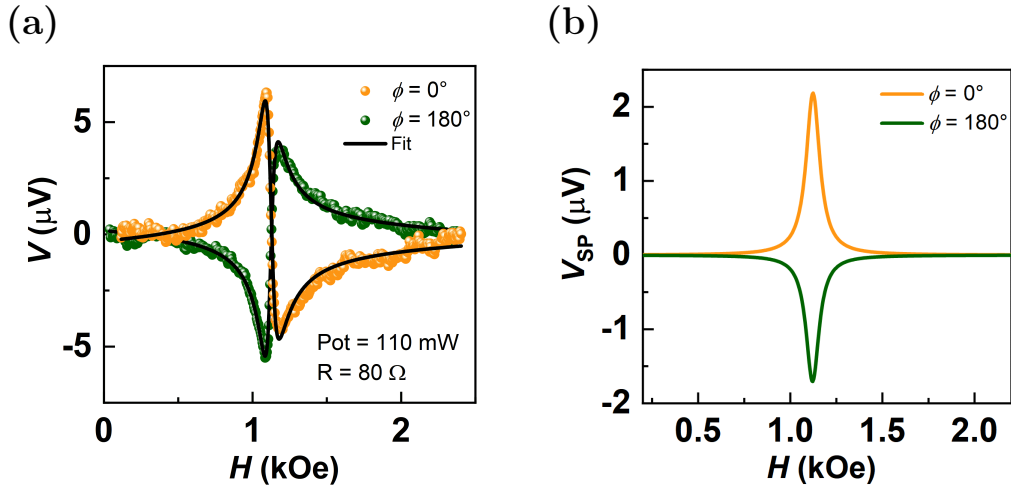


Figure 69 – Measured dc voltages obtained for (a) Py(20 nm)/SnTe(7 nm)/InP sample with the magnetic field oriented at $\phi = 0^\circ$ and 180° with the resonance magnetic field $H_R = 1.056 \text{ kOe}$. The fit shows very good agreement with the data, describing a superposition of symmetric and antisymmetric components. Panel (b) shows the symmetric contribution, which is exclusively associated with a spin to charge conversion mechanism. The inversion of the voltage with the field direction is a characteristic feature of the ISHE or IREE.

This measurement procedure was repeated for all samples and for different microwave powers, as shown in Figure 70 (a). By extracting the symmetric contribution,

Figure 70 (b) was obtained showing the direct relationship between the microwave power and the voltage generated by the spin-to-charge conversion. As shown in the inset of Figure 70 (b), the V_{SP} scales linearly with the applied microwave power.

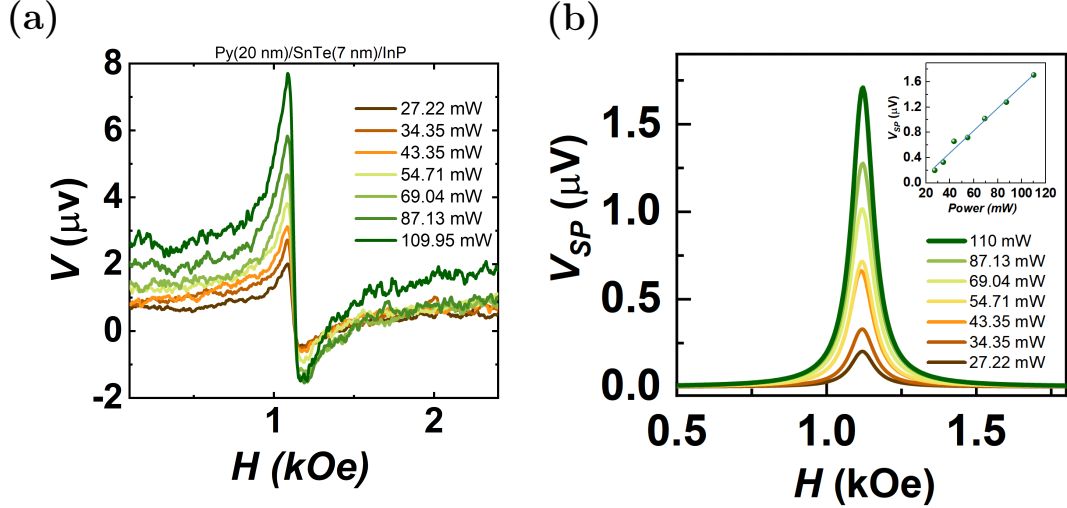


Figure 70 – Panel a) shows the dependence of the dc voltage line shapes as a function of the incident microwave power for Py(20 nm)/SnTe(7 nm)/InP sample at $\phi = 0^\circ$. Panel (b) shown the dependence of the symmetric component of the spin pumping dc voltage as a function of microwave incident power. The inset shows peak values of the dc spin pumping voltage as a function of the incident microwave power. The data points exhibit a linear behavior, as expected for ISHE or IREE.

Tin telluride is a topological crystalline insulator, it means this material is robust against an external magnetic field, until certain level [54, 55, 53]. Moreover, the in-plane external magnetic field and the magnetization of the permalloy layer are expected only to shift the Dirac cone along the momentum direction, while preserving its linear dispersion, making spin-to-charge conversion via the IREE possible [223].

4.3.6.1 Charge current density

The bi-dimensional charge current density J_c is generated in the non-magnetic material or at an interface. With units of charge per unit length, its origin can be related to quantum phenomena such as the IREE or the ISHE. Its magnitude depends on the spin-to-charge conversion efficiency, characterized by θ_{SH} or λ_{IREE} . The charge density J_c is estimated by

$$J_c = \frac{V_{SP}^{\text{peak}}}{Rw}.$$

The therm $V_{\text{SP}}^{\text{peak}}$ corresponds to the peak value of the measured spin pumping DC voltage associated to exclusively to spin pumping $V_{\text{SP}}^{\text{peak}} = 1.71 \mu\text{V}$. The electrical resistance is $R = 180 \Omega$. The sample width, defined as the distance between the electrical contacts, is given by $w = 2.5 \text{ mm}$. With this, we estimated

$$J_c = 3.80 \times 10^{-6} \text{ A/m}.$$

The obtained value is consistent in order of magnitude with that reported for MBE-grown SnTe, in which the authors claim that the spin-to-charge current conversion is governed by the ISHE [224].

4.3.6.2 Spin current density

The Three-dimensional spin current density J_s represents the angular momentum flux of injected spins per unit area and time. This flux occurs from a magnetic material into a non-magnetic material in a layer or interface, as in FMR experiments. The spin current density J_s can be estimated by

$$J_s = \frac{e\omega p g_{\text{eff}}^{\uparrow\downarrow}}{8\pi} \left(\frac{h_{\text{rf}}}{\Delta H} \right)^2 L(H - H_R)$$

where the $L(H - H_R)$ is given by

$$L(H) = \frac{\Delta H^2}{[(H - H_r)^2 + \Delta H^2]}$$

and is equal to 1 when in the resonance regime ($H = H_R$). In the J_s equation, $p = 0.30$ is the precession ellipticity factor of the Py layer [15]. The term h_{rf} corresponds to the amplitude of the microwave magnetic field. It is related to the microwave power applied to the sample by $h_{\text{rf}} = 1.776\sqrt{P_i}$ relationship, where P_i is given in watts. In this work, the maximum applied power was $P_i = 110 \text{ mW}$. With this parameters, we found the spin current density of

$$J_s = 4.74 \times 10^4 \text{ A/m}^2.$$

This spin current value is less than one order of magnitude lower than that reported for the topological insulator $(\text{Bi}_{0.22}\text{Sb}_{0.78})_2\text{Te}_3$, ($J_S = 2.3 \times 10^5 \text{ A/m}^2$), which is recognized as one of the most robust and efficient topological insulators [71].

4.3.6.3 The IREE conversion length

The IREE conversion length λ_{IREE} is a parameter that characterizes the IREE, connecting the three dimensional spin current density J_s to the two-dimensional charge current density J_c through the following relationship:

$$\lambda_{\text{IREE}} = \frac{J_c}{J_s}$$

This parameter quantify the spin-to-charge efficiency conversion in a interface or surface with high SOC. It has units of length and is proportional to the magnitude of the SOC [225, 226]. In this work we estimated $\lambda_{\text{IREE}} = 0.080 \text{ nm}$. This λ value is slightly higher than those reported from SP-FMR, and longitudinal SSE measurements (0.076 nm) in $\text{Bi}_{0.22}\text{Sb}_{0.78}\text{Te}_3/\text{Py}$ heterostructures [71]. This value is also and one order of magnitude higher for that of $\text{Py}/\text{SnTe}/\text{YIG}$ heterostructures with sputter-deposited SnTe [222].

4.3.6.4 Gilbert Damping

The Gilbert damping parameter α is a phenomenological, dimensionless quantity that describes the rate at which the precessing magnetization relaxes toward the equilibrium direction in the LLG equation. The FMR linewidth parameter is related to α by the following expression

$$\Delta H = \frac{\alpha\omega}{\gamma},$$

where

$$\gamma = \frac{g\mu_B}{\hbar}$$

ω being the angular frequency, μ_B the Bohr magneton, $\hbar$ the reduced Planck constant, and g spectroscopic splitting factor (g -factor). In this sense, a higher values of α lead to a broader the FMR linewidth, indicating increased energy loss. Using $\gamma = 2,94 \text{ GHz/kOe}$, $\omega = 5.906 \times 10^{10} \text{ s}^{-1}$, and the obtained value of the FMR linewidth from Py/SnTe and Py structures, we have

$$\alpha_{\text{Py}} = 1.44 \times 10^{-3}$$

$$\alpha_{\text{Py/SnTe}} = 1.69 \times 10^{-3}$$

with this results, we have a $\Delta\alpha = 0.25 \times 10^{-3}$. A lower Gilbert damping value is desirable for reducing power consumption in magnetic memories [227]. The estimated value in this work is smaller that those reported from Py with the same thickness [40]. A more accurate estimation can be obtained by evaluating the linewidth as a function of frequency [228].

4.3.6.5 Spin Hall Angle

The spin Hall angle (θ_{SH}) is a parameter defined as the ratio between the spin current density J_s and the charge current density J_c . The final equation form in function of V_{SP} is

$$\theta_{SH} = \frac{4V_{SP}}{Rw\lambda efg_{eff}^{\uparrow\downarrow}p \tanh\left(\frac{t_{SnTe}}{2\lambda_{sd}}\right) \left(\frac{h_{rf}}{\Delta H}\right)^2}.$$

The measured DC voltage generated by spin pumping is denoted by $V_{SP} = 1.71 \mu V$. The electrical resistance of the layer where the spin-to-charge conversion occurs is given by $R_{py} = 180 \Omega$. The sample width, defined as the distance between the electrical contacts, is denoted by $w = 2.5$ mm. The spin diffusion length of the non-magnetic layer is represented by λ_{sd} . The effective spin-mixing conductance of the magnetic/non-magnetic interface is given by $g_{eff}^{\uparrow\downarrow} = 1.4 \times 10^{18} m^{-2}$. The electron charge $e = 1.602176634 \times 10^{-19}$ C. The frequency of the driving microwave magnetic field is represented by $f = 9.4$ GHz. The ellipticity factor of the magnetization precession is denoted by $p = 0.3$. The thickness of the SnTe layer is represented by $t_{SnTe} = 7$ nm. The amplitude of the exciting microwave field is given by $h_{rf} = 0,348$ Oe. The FMR linewidth is denoted by $\Delta H = 34$ Oe.

$$\theta_{SH} \approx \frac{0.24 \text{ nm}}{\lambda_{sd}}$$

This parameter quantifies the spin-to-charge conversion efficiency, meaning that materials with high values of θ_{SH} (such as Pt, Ta, W) are crucial for spin-orbit torque magnetic memories, as they allow magnetization manipulation using small electric current densities. In our experiments, we found a value of the spin Hall angle multiplied by the spin diffusion length of $\theta_{SH}\lambda_{sd} = 0.24$ nm. Based on the reported spin diffusion length of (≈ 6 nm) well-known topological insulator Bi_2Se_3 [229], as reference [222] did, we performed a rough estimation of $\theta_{SH} = 0.04$ for our material. This magnitude is usually found in good spin-sink materials such as Pt [230].

5 FINAL CONSIDERATIONS

Spin transport and spin-to-charge conversion in PANI heterostructures were investigated using SSE and SP-FMR experiments. Spin-coated PANI films with thicknesses of approximately 100 nm exhibited good interfacial conformity and induced an increase in RMS surface roughness from 0.8 nm (YIG) to about 2.65 nm, which was transferred to the Pt top layer. Electrical measurements revealed ohmic contacts and tunable resistivity (via secondary doping), with resistance values decreasing from 14.13 k Ω to 6.39 k Ω . FMR measurements demonstrated linewidth broadening in PANI/YIG and Pt/PANI/YIG structures, evidencing efficient spin pumping across the interfaces. The extracted spin-mixing conductance are $g_{\text{eff}}^{\uparrow\downarrow \text{PANI/YIG}} = 0.56 \cdot g_{\text{eff}}^{\uparrow\downarrow \text{Pt/YIG}}$ and $g_{\text{eff}}^{\uparrow\downarrow \text{PANI/Pt/YIG}} = 1.6 \cdot g_{\text{eff}}^{\uparrow\downarrow \text{Pt/YIG}}$, indicating that most of the injected spin current is transmitted through the PANI layer and absorbed in Pt. Spin Seebeck measurements confirmed spin-current transmission through PANI, with V_{SSE} values approximately one order of magnitude smaller than in Pt/YIG due to spin losses in the polymer. Direct detection of spin-to-charge conversion in PANI yielded a linear dependence of V_{SSE} on ΔT , from which a spin Hall angle of $\theta_{\text{SH}} = (1.70 \pm 0.06) \times 10^{-5}$ was extracted. This value is several orders of magnitude smaller than those typically reported for heavy metals and topological insulators, such as Pt and Bi₂Se₃, reflecting the intrinsically weak spin-orbit coupling in organic polymers [230, 229]. These results demonstrate that, despite its weak SOC, PANI supports long-range spin transport and measurable spin-to-charge conversion, establishing conductive polymers as viable platforms for organic spintronic and flexible devices.

Atomic force microscopy measurements show that PEDOT:PSS films deposited by drop-casting exhibit thicknesses of approximately 2.20 μm and 2.54 μm for deposited volumes of 20 μL and 40 μL , respectively, indicating the onset of thickness saturation at higher solution volumes. The films present a granular morphology with an RMS roughness of about 5.11 nm, reflecting the characteristic core-shell structure induced by excess PSS. Raman spectroscopy confirms the chemical integrity of the PEDOT:PSS films through the observation of characteristic vibrational modes associated with the polymer backbone, in good agreement with literature reports. Optical transmittance and absorbance measurements performed on quartz substrates reveal well-defined π - π^* transitions and subgap absorption features related to bipolaron states, evidencing the highly

conductive nature of the polymeric films. Tauc analysis yields optical band gap energies of 2.44–2.55 eV for direct transitions and 2.00–2.06 eV for indirect transitions, placing PEDOT:PSS within the typical range of organic semiconductors suitable for optoelectronic and photovoltaic applications. The pseudo-four-probe measurements revealed linear I – V characteristics, confirming the formation of ohmic contacts directly in the PEDOT:PSS surface. The ISHE voltage exhibited a clear dependence on the in-plane magnetic field orientation (0° , 90° , and 180°) and reversed its sign upon field inversion, evidencing spin-to-charge current conversion generated by ISHE. Using a linewidth broadening in the FMR measurements, we found $g_{\text{PEDOT:PSS/YIG}}^{\uparrow\downarrow} = 1.46 \times 10^{18} \text{ m}^{-2}$, and with this, we obtained $g_{\text{PEDOT:PSS/YIG}}^{\uparrow\downarrow} = 1.46 g_{\text{Pt/YIG}}^{\uparrow\downarrow}$. This reveals a highly efficient spin transfer across the interface, as the $g_{\text{eff}}^{\uparrow\downarrow}$ of the PEDOT:PSS/YIG system is of the same order of magnitude as that of Pt/YIG [12, 28].

Under a thermal gradient applied perpendicular to the layers, a measurable SSE signal was detected in the PEDOT:PSS layer. The magnetic-field dependence of V_{SSE} for different temperature differences ΔT shows the expected ISHE–SSE behavior, confirming that thermally generated spin currents in YIG are efficiently converted into electrical signals in the polymer layer. Consequently, we obtained a spin Hall angle of $\theta_{\text{SH}} = (3.43 \pm 0.06) \times 10^{-5}$ that agrees well with the spin Hall angles reported for similar polymeric materials [202, 12, 172]. Combined with spin-to-charge conversion measurements obtained by SSE and SP-FMR, these results demonstrate that PEDOT:PSS films exhibit appropriate structural quality, optical properties, and spin-transport functionality, supporting their potential for organic spintronic devices. Although the spin-to-charge conversion efficiency in PEDOT:PSS remains lower than that observed in inorganic systems such as Pt or topological insulators, its performance is comparable to other conductive polymers reported in the literature, highlighting the relevance of interface engineering for enhancing spin–orbit-related phenomena in organic materials [227].

Atomic force microscopy measurements reveal that uncapped SnTe films exhibit a triangular surface features, characteristic of grown along the (111) direction, and low roughness, with an RMS value of approximately 0.55 nm. The presence of a Te capping layer preserves the surface uniformity, yielding comparable roughness values of about 0.58 nm. In contrast, the incorporation of SnTe beneath the Py layer can be induced

a pronounced modification of the top AlO_x surface morphology, leading to an increase in RMS roughness from 0.33 nm to 1.49 nm and to the formation of larger granular agglomerates. This alteration can also be attributed to the relatively large thickness of the Py layer (≈ 20 nm). Raman spectroscopy confirms the characteristic A_{1g} and E_{TO} phonon modes at 125.33 cm^{-1} and 142.04 cm^{-1} , respectively, in agreement with literature values. In heterostructures, the partial attenuation of the Raman signal reflects the masking effect of the overlayers, while the emergence of the disorder-related mode near 104 cm^{-1} indicates structural imperfections in thinner films. A systematic downshift of the A_{1g} mode from 125.3 cm^{-1} to 121.6 cm^{-1} is observed as the thickness decreases from 20 nm to 7 nm, revealing enhanced compressive strain induced by the substrate. The concomitant changes in the relative intensities of the M2, M3, and M4 modes further support the presence of strain and confinement effects. These results demonstrate that film thickness strongly influences the vibrational and structural properties of SnTe.

High-resolution X-ray diffraction confirms the epitaxial growth of single-phase SnTe(111) films on InP(111), as evidenced by well-defined (111) and (222) reflections and the presence of Laue fringes. The narrow rocking-curve width of 0.080° for the 30 nm SnTe layer demonstrates high crystalline quality and low mosaicity. A weak NiFe(220) reflection at $2\theta \approx 77^\circ$ is related to the Py layer, suggesting its polycrystalline phase. Analysis of the (222) diffraction peak yields out-of-plane lattice parameters of $a_\perp = 6.330 \text{ \AA}$ for 20 nm and $6.370 \text{ \AA}$ for SnTe (7 nm) films, both exceeding the bulk value, revealing compressive in-plane strain induced by the InP substrate. X-ray reflectivity measurements provide quantitative thickness, density, and roughness values for all layers, confirming sharp interfaces. Together, the HRXRD and XRR results demonstrate that high-quality epitaxial SnTe films were obtained and that strain and thickness play a central role in determining the structural properties of the heterostructures. Symmetric reciprocal space mapping around the (222) reflection confirms the high structural coherence and epitaxial alignment between the SnTe(111) films and the InP(111) substrate, as evidenced by the alignment of the SnTe and InP diffraction peaks along the $q_\parallel = 0$ axis. The observed peak broadening reflects a moderate degree of in-plane mosaicity. Asymmetric reciprocal space mapping around the (224) reflection reveals that the 20 nm SnTe layer remains largely strained, exhibiting an in-plane lattice parameter of $5.951 \text{ \AA}$ and a relaxation factor of 0.214, close to the InP lattice constant. For the SnTe (7 nm) layer, pronounced

peak broadening and intensity redistribution indicate increased mosaicity and a higher compressive strain state. Together, these results demonstrate that film thickness strongly controls strain relaxation and crystalline coherence in SnTe/InP heterostructures.

The spin-to-charge conversion experiments performed on the Py/SnTe heterostructures demonstrate efficient interfacial spin transport and conversion. From the analysis of the ferromagnetic resonance linewidth, an effective spin-mixing conductance of $g_{\text{eff}}^{\uparrow\downarrow} = 1.4 \times 10^{18} \text{ m}^{-2}$ ($1.4 \times 10^{14} \text{ cm}^{-2}$) was obtained, indicating high spin-transmission efficiency across the interface. The spin-mixing conductance of SnTe is of the same order of magnitude as that of Pt, indicating a high efficiency for spin-current absorption [175, 174]. This result is particularly significant, since platinum is widely recognized as one of the most efficient spin sinks. Furthermore, this value is comparable to those reported for other topological materials, such as $\text{Bi}_{0.22}\text{Sb}_{0.78}\text{Te}_3$ and Bi_2Se_3 , confirming the robust spin-absorption capability of SnTe-based heterostructures [71, 229]. The Gilbert damping parameter increased from $\alpha_{\text{Py}} = 1.44 \times 10^{-3}$ for the reference Py layer to $\alpha_{\text{Py/SnTe}} = 1.69 \times 10^{-3}$ in the presence of the SnTe layer, corresponding to a damping enhancement of $\Delta\alpha = 0.25 \times 10^{-3}$. This increase is attributed to spin pumping into the SnTe layer. The injected spin current density was estimated as $J_s = 4.74 \times 10^4 \text{ A/m}^2$, which was converted into a transverse charge current density of $J_c = 3.80 \times 10^{-6} \text{ A/m}$.

This spin current density is less than one order of magnitude lower than that reported for highly efficient topological insulators, such as $(\text{Bi,Sb})_2\text{Te}_3$, which typically exhibit J_s values on the order of 10^5 A/m^2 [71]. From these values, an inverse Rashba–Edelstein length of $\lambda_{\text{IREE}} = 0.080 \text{ nm}$ was extracted, evidencing the relevance of interfacial spin–orbit coupling and spin–momentum locking at the Py/SnTe interface. This value is slightly higher than those reported for Py/ $\text{Bi}_{0.22}\text{Sb}_{0.78}\text{Te}_3$ heterostructures and approximately one order of magnitude larger than that obtained in sputter-deposited SnTe films, indicating a superior interfacial quality and influence of high crystallinity in the present MBE-grown samples [71, 222]. A rough estimation of the spin Hall angle of $\theta_{SH} = 0.04$ was done for our material. This magnitude is usually found in good spin-sink materials such as Pt [230]. It is important to note that there is no consensus on whether the ISHE or the IREE is dominant in the spin-to-charge conversion in SnTe. Due to the configuration of our heterostructures, we cannot reach a definitive conclusion regarding which mechanism

is dominant. Overall, these results confirm that SnTe acts as an efficient spin sink and spin-to-charge converter, combining sizable spin-mixing conductance, measurable damping enhancement, and finite interfacial conversion length. Compared to conventional heavy metals and other topological insulators, the Py/SnTe system exhibits competitive spin-transport and conversion efficiencies, reinforcing the potential of topological crystalline insulators for spintronic applications based on spin pumping and interfacial spin-orbit effects.

The results obtained for the PANI-based heterostructures have already been published in <https://doi.org/10.1016/j.jmmm.2021.168635>. The results concerning the PEDOT:PSS polymer films are currently in the final step of submission. Finally, the results related to the SnTe heterostructures are in the final stage of manuscript preparation.

6 PERSPECTIVES

In this study, we will investigate SnTe samples with varying thicknesses, spanning from the three-dimensional to the two-dimensional limit. Our goal is to elucidate the relative contributions of the ISHE and the IREE to the spin-to-charge conversion efficiency across these dimensional regimes.

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