

Magnetophotonics for Electromagnetic Surface Waves Sensors

João Pedro Miranda Carvalho

MSc. Engineering Physics

Departamento de Física e Astronomia

2023

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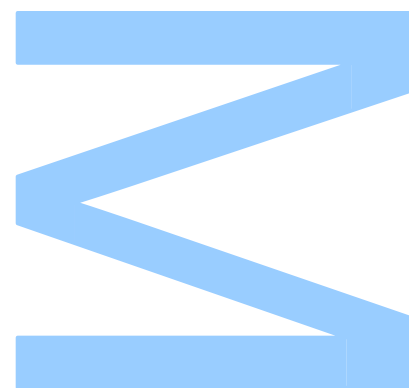
Prof. Doutor Luís Carlos Costa Coelho, Professor Auxiliar Convidado
FCUP, INESC TEC

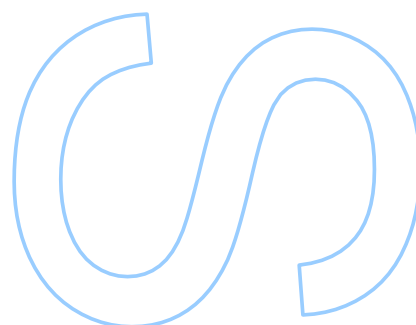
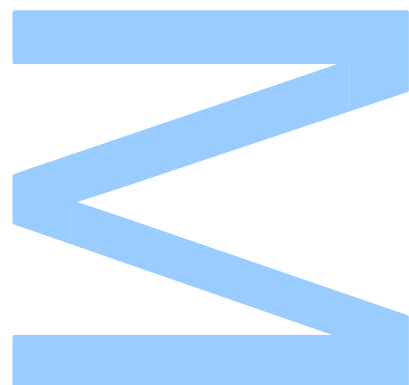
Coorientador

Prof. Doutor José Manuel Marques Martins de Almeida, Professor Associado
UTAD, INESC TEC

Coorientador

Prof. Doutor Pedro Alberto da Silva Jorge, Professor Auxiliar
FCUP, INESC TEC





Acknowledgements

The work described in this dissertation condenses personal efforts of over a year, together with the direct and indirect contribution of numerous people that I would like to acknowledge.

First and foremost, I would like to share a word of gratitude towards my supervisor, Professor Luís Coelho, and my co-supervisors, Professor José Manuel Almeida and Professor Pedro Jorge, for their valuable insight and guidance, encouraging support and ever present availability, without which this dissertation would not be possible. I would also like to leave a kind word to Bernardo Dias for his immense help and work that laid grounds on much of what was there for me to achieve. A felt word of gratitude follows to Dr. João Horta Belo, for his help and availability that was critical to this work.

I would like to direct a kind word of friendship to my dear friends Andreia, João, Maria, Miguel, Miguel Trovão, Nuno, Rafael and Tiago, that formed a close welcoming space to vent frustrations and rejoice achievements.

To my dear parents and sister, I would like to thank for their sacrifices and for being a Home, always supportive of my decisions and certain of my capacities; and to make a promise that the financial burden should be over soon.

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Abstract

Faculdade de Ciências da Universidade do Porto

Departamento de Física e Astronomia

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Magnetophotonics for Electromagnetic Surface Waves sensors

by João Pedro Miranda Carvalho

Electromagnetic Surface Waves (ESWs) are electromagnetic (EM) modes that propagate along an interface, with exponential decay of field intensity into each medium. The evanescent field penetration renders sensitivity of these modes to the optical properties of surrounding media. Surface Plasmon Polaritons (SPPs) and Bloch Surface Waves (BSWs) are examples of ESWs that can be excited at the interface of layered media, and exhibit high sensitivities to changes in the dielectric properties of external media. The optical properties of a medium can too be modulated by the incidence of magnetic fields in a magnetic layer. In some magnetophotonic structures, the magnetization of a thin layer induces a modulation of its dielectric properties, capable of tuning the coupling conditions of ESWs. In return, the excitation of ESWs is known to enhance magneto-optical (MO) effects perceived in the thin magnetic layer.

A complete simulation study of MO effects in ESWs based on Berreman's matrix formalism is carried out to simulate reflectance spectra of optimized magnetophotonic structures, both in planar substrates and multimode optical fibers. All selected thin films structures are fabricated and the spectra of relative reflected intensity for opposite magnetizations are determined. Refractometric characterization of such films is carried out to experimentally verify configurations of enhanced reflectance sensitivity to refractive index (RI) by MO modulation relative to the non-modulated signal.

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Resumo

Faculdade de Ciências da Universidade do Porto

Departamento de Física e Astronomia

Mestrado Integrado em Engenharia Física

Magnetofotónica em sensores de Ondas Eletromagnéticas de Superfície

por [João Pedro Miranda Carvalho](#)

Ondas eletromagnéticas de superfície são modos eletromagnéticos que se propagam ao longo de uma interface, com decaimento exponencial da intensidade de campo no interior dos meios na interface. A propagação da onda evanescente num meio atribui sensibilidade das ondas de superfície às propriedades óticas dos meios envolventes. Plasmões de superfície e Ondas de Bloch são dois exemplos de ondas eletromagnéticas de superfície que demonstram elevada sensibilidade às propriedades dielétricas do meio externo. As propriedades óticas de um material magnético podem também ser moduladas pela incidência de campos magnéticos. Em algumas estruturas magnetofotónicas, a magnetização de um filme fino ferromagnético induz alterações às propriedades dielétricas do meio, capazes de modular as condições de excitação de ondas eletromagnéticas de superfície. Adicionalmente, a excitação de ondas eletromagnéticas de superfície possibilita uma otimização de efeitos magneto-óticos em filmes finos.

A simulação de efeitos magneto-óticos em espectros de ondas de superfície é levada a cabo pelo método da matriz de transferência de Berreman, em estruturas planares e de geometria axial, como em fibras óticas. Todas as configurações otimizadas foram fabricadas e o espectro de intensidade relativa de reflectâncias para estados de magnetização opostos são determinados. A caracterização em índice de refração de todos os filmes é levada a cabo de forma a demonstrar uma otimização da sensibilidade em reflectância ao índice de refração por modulação magneto-ótica face ao sinal original.

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Chapter 1

Electromagnetic Surface Waves in Magnetophotonics

1.1 Maxwell's Equations and Optical Properties of Magnetic Media

The propagation of light in matter forms the core of electromagnetism, through which many interesting phenomena can occur and be accounted for by imposing conditions to the fundamental theory behind classical electrodynamics: the Maxwell's Equations. In their differential form, the Maxwell's equations can be represented as [1]

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\epsilon_0} \quad (1.1)$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \quad (1.2)$$

$$\nabla \cdot \mathbf{B} = 0 \quad (1.3)$$

$$\nabla \times \mathbf{B} = \mu_0 \mathbf{J} + \epsilon_0 \mu_0 \frac{\partial \mathbf{E}}{\partial t} \quad (1.4)$$

being $\mathbf{E}$ the electric field, $\mathbf{B}$ the magnetic induction field, ϵ_0 and μ_0 the electric permittivity and magnetic permeability of free space, respectively, and t represents time.

An auxiliary electric displacement field, $\mathbf{D}$, and the magnetic field, $\mathbf{H}$, can be defined expressing a direct relation of $\mathbf{E}$ with the polarization, $\mathbf{P}$, and $\mathbf{B}$ with the magnetization, $\mathbf{M}$, by

$$\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P} \quad (1.5)$$

$$\mathbf{H} = \frac{1}{\mu_0} \mathbf{B} - \mathbf{M} \quad (1.6)$$

In linear, homogeneous, isotropic materials, equations 1.1 and 1.4 can be expressed as

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\epsilon} \quad (1.7)$$

$$\nabla \times \mathbf{B} = \mu \mathbf{J} + \mu \epsilon \frac{\partial \mathbf{E}}{\partial t} \quad (1.8)$$

with ϵ and μ being the permittivity and permeability of the media, given by $\epsilon = \epsilon_r \epsilon_0$ and $\mu = \mu_r \mu_0$. Here, ϵ_r and μ_r are the relative permittivity and permeability, that scale the permittivities and permeabilities of free space to those of a given medium. ϵ_r and μ_r are adimensional and characterize on their own the optical properties of media, so that from now on, the relative permittivity and permeability will be simply referred to as permittivity, ϵ , and permeability, μ .

In the more general case, ϵ and μ are tensorial quantities, represented in three-dimensional space by a 3×3 matrix. Within the macroscopical view of light-matter interactions, the properties of a medium are both given by the ϵ and μ tensors. At optical frequencies, of particular interest in this work, μ is close to unity [2], so that the optical properties of a medium can be accurately described by the ϵ tensor.

The optical properties of isotropic non-magnetic materials or in the absence of an external magnetic field are formally described as a diagonal matrix with $\epsilon_{ij} = \epsilon_{ji}$ and $\epsilon_{ii} = \epsilon_r$, although the dielectric tensor can be simplified to a complex number $\epsilon = \epsilon_{ii}$ with no risk of mischaracterization.

The formal description of magneto-optical effects, however, needs to account the effects of the applied magnetic field to the dielectric tensor of the medium. A symmetric dielectric tensor $\epsilon_{ij} = \epsilon_{ji}$ for absent magnetic field becomes non-symmetric under the influence of a magnetic field, adding off-diagonal elements to the dielectric tensor ($\epsilon_{ij}(\mathbf{H}) = \epsilon_{ji}(-\mathbf{H})$).

Considering the general case of a medium under the incidence of a static magnetic field, the dielectric tensor takes the form

$$\epsilon = \begin{pmatrix} \epsilon_x & 0 & 0 \\ 0 & \epsilon_y & 0 \\ 0 & 0 & \epsilon_z \end{pmatrix} + ig(M) \begin{pmatrix} 0 & -m_z & m_y \\ m_z & 0 & -m_x \\ -m_y & m_x & 0 \end{pmatrix} + b(M) \begin{pmatrix} m_y^2 + m_z^2 & -m_x m_y & -m_x m_z \\ -m_x m_y & m_x^2 + m_z^2 & -m_y m_z \\ -m_x m_z & -m_y m_z & m_x^2 + m_y^2 \end{pmatrix} \quad (1.9)$$

For the case of an initially isotropic ferromagnetic material, then magnetized by a magnetic field, $\epsilon_x = \epsilon_y = \epsilon_z$ is the complex permittivity, $m_i = \mathbf{M}_i/M$ are unitary vectors of magnetization and g is the complex magneto-optical gyration constant. The third term

of 1.9 relates to second order magneto-optical effects of low expression, and are often disregarded. For the characterization of magneto-optical effects, it is usually defined a complex magneto-optical Voigt parameter, Q , as

$$Q = \frac{g}{\epsilon_i + b} \quad (1.10)$$

For non-magnetic media, Q is governed by the magnetic field, H , whereas for ferromagnetic media, the magnetization M is the order parameter. For the case of an isotropic ferromagnet, g is linear with magnetization and can be given by $g = aM$, with a being a complex frequency dependent magneto-optical parameter and M being the magnetization magnitude. The magneto-optical coefficient can thus be decomposed as

$$a(\lambda) = a'(\lambda) + ia''(\lambda) \quad (1.11)$$

This magneto-optical coefficient couples the properties of the medium with those of light, implementing the spectral dependence of magneto-optical effects into the dielectric tensor.

1.2 Magneto-Optical Effects

Magneto-optics is the field of study of the interaction of light with matter under the influence of a magnetic field or magnetization. The magneto-optical phenomena can be separated into two main classes. The first class describes effects resultant from the direct action of the magnetic field on the orbital motion of the electrons, with dielectric tensor elements as a function of $\mathbf{H}$ [3]. All diamagnetic transparent solids have magneto-optical effects that originate from these direct effects of the magnetic field on the orbital-electronic motion. Ferromagnetic materials and nonmetallic paramagnetic materials at low temperatures represent the other class of magneto-optical materials. In these, the direct effect of the magnetic field on the electron orbital motions is negligible compared to the effects of spin-orbit coupling. For such materials, it is more appropriate to express ϵ as a function of $\mathbf{M}$, instead of $\mathbf{H}$.

Michael Faraday (1791-1867) pioneered the field of magneto-optics with the discovery of the self titled Faraday Effect, in 1845 [4]. In the experiment, Faraday noticed a rotation of the plane of polarization of plane polarized light when transmitted through a glass under the influence of a magnetic field parallel to the direction of propagation of light. This first demonstration of the merge of light and magnetism started the research

of an interesting branch of physics whose fundamentals would not be understood until almost 100 years later, with the quantum mechanical approach to magnetism. A set of other distinct magneto-optical effects can occur depending on factors such as the relative direction of the incident magnetic field, the polarization state of the light or its interaction with matter. Such are the cases of the Magneto-Optical Kerr Effect (MOKE) and the Voigt Effect. The latter effect is very similar to MOKE apart from its quadratic dependence on the magnitude of the magnetic field (or magnetization) and it's of very small expression.

In the context of this work, only the MOKE was involved. The MOKE can enhance and be enhanced by simultaneous excitation of ESWs, and its functioning by reflection off a sample (instead of transmission, as in the Faraday effect) is convenient given the need of total internal reflection of light for ESW excitation.

1.2.1 Magneto-Optical Kerr Effect

The magneto-optical Kerr effect (MOKE) manifests itself in the influence a magnetized medium exerts over light reflected off its surface. The effect was discovered in 1888 by John Kerr (1824-1907), that first observed that when plane-polarized light is reflected from the polished surface of an electromagnet, it becomes elliptically polarized, with its major axis rotated with respect to the plane of polarization of the incident beam [4].

Both the spin-orbit interaction and the exchange splitting of the electronic structure coexist in the MOKE. A strong linear relation between the spin-orbit interaction parameter and the imaginary part of the permittivity has been linked. The relationship between the exchange splitting, closely related to the magnetization, and the intensity of magneto-optical effects is not as clear, as the change in exchange splitting can induce non-linear changes in the spectral features of the optical conductivity of a medium, and thus on the complex permittivities [5].

Depending on the relative orientation of the plane of incidence of light to the direction of the magnetization vector and the sample surface plane, different contributions of the MOKE can be present. Three segregate configurations of the effect can be distinguished: the polar, longitudinal and transverse Kerr effect. An illustrative representation of the three configurations of MOKE is depicted in Figure 1.1

In the polar MOKE (PMOKE), the magnetization vector is normal to the sample surface plane and parallel to the incidence plane. The reflection of light results in the rotation of the plane of polarization and appearance of ellipticity on prior linearly polarized light.

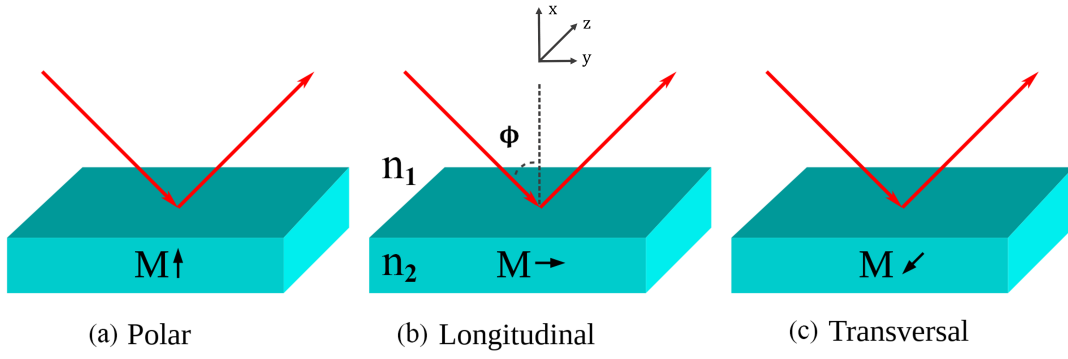


FIGURE 1.1: MOKE configurations of (a) Polar MOKE, (b) Longitudinal MOKE and (c) Transverse MOKE.

The longitudinal MOKE (LMOKE, sometimes referred as meridional MOKE) occurs for samples with the magnetization vector parallel to their surface and the plane of incidence of light. Upon reflection, the previously linearly polarized light gets its polarization plane rotated while obtaining ellipticity. The transverse MOKE (TMOKE, sometimes referred as equatorial MOKE), in contrast to aforementioned effects, does not suffer polarization rotation of the incident light and can only be observed for absorbing materials. When linearly polarized light is incident on a sample magnetized in a plane parallel to the sample surface, but normal to the plane of light incidence, the reflected beam suffers intensity variations and a phase shift. The expressions for the rotation angle for the polar and longitudinal MOKE, as well as the relative change in reflected light intensity for transverse MOKE, are expressed in the following equations:

1. Polar Kerr effect:

$$\Phi_K^{s,p} = \text{Re} \frac{\mp r_{sp}}{r_{ss} (r_{pp})} = \text{Im} \frac{\eta^2 \left[(\eta^2 - \sin^2 \phi)^{1/2} \mp \sin \phi \tan \phi \right]}{(\eta^2 - 1) (\eta^2 - \tan^2 \phi)} Q \quad (1.12)$$

2. Longitudinal Kerr effect:

$$\Phi_K^{s,p} = \text{Re} \frac{(\pm r_{sp})}{r_{ss} (r_{pp})} = \text{Im} \frac{\left[\sin \phi \eta^2 \left(\sin \phi \tan \phi \pm \sqrt{\eta^2 - \sin^2 \phi} \right) \right]}{(\eta^2 - 1) (\eta^2 - \tan^2 \phi) (\eta^2 - \sin^2 \phi)^{1/2}} Q \quad (1.13)$$

3. Transverse Kerr effect:

$$\delta_p = \frac{I(M) - I(-M)}{I(0)} = -\text{Im} \frac{4 \tan \phi \eta^2}{(\eta^2 - 1) (\eta^2 - \tan^2 \phi)} Q \quad (1.14)$$

Being ϕ the incidence angle and $\eta = n_2/n_1$ the relative refractive index of the media at the interface, as seen in Figure 1.1. The upper sign in $\pm$ or $\mp$ corresponds to s -polarized light, while the lower sign corresponds to p -polarized light. These expressions assume a linear Q with magnetization [3], and for $\mu = 1$, the reflection and transmission coefficients r_{ss} , r_{pp} and r_{sp} present in 1.12 and 1.13 for polarization crossing terms [6] are detailed in appendix B.

The transverse MOKE, of all three effects, will be extensively approached throughout this work. Despite not being the configuration that yields the most noticeable magneto-optical effects, the TMOKE is optimal for simultaneous ESW excitation. Additionally, the orientation of the magnetic field being normal to the plane of light incidence assures that there is no contribution of the Faraday effect in the coupling prism to the observed effect.

1.2.2 Transverse MOKE

Consider the transmission of light through an interface between a magnetic layer of refractive index n_2 , and a non-magnetic layer of refractive index n_1 . The light is incident at the interface in the xy plane, and the magnetization vector of the magnetic material is aligned in the z axis. It is assumed that the magnetization can be characterized with only first-order linear terms [3] and stands at its saturated value ($m_i = 1$). In these conditions, the magnetic medium is described by the dielectric tensor

$$\epsilon = \begin{pmatrix} \epsilon_r & 0 & 0 \\ 0 & \epsilon_r & 0 \\ 0 & 0 & \epsilon_r \end{pmatrix} + \begin{pmatrix} 0 & -ia(\lambda)M & 0 \\ +ia(\lambda)M & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

Light is incident from the non-magnetic medium n_1 to the magnetic medium n_2 at an angle ϕ relative to the normal to the interface, and with electric field vector parallel to the plane of incidence (p polarization). Considering the projections on the coordinate axes, the electric fields of incident, $\mathbf{E}_i$, and reflected, $\mathbf{E}_r$, waves can be described by

$$\begin{aligned} \mathbf{E}_i &= E_i \begin{pmatrix} -\sin \phi \\ -\cos \phi \\ 0 \end{pmatrix} e^{-i\omega t + i(\omega/c)\mathbf{n}_i \cdot \mathbf{r}} \\ \mathbf{E}_r &= E_r \begin{pmatrix} -\sin \phi \\ \cos \phi \\ 0 \end{pmatrix} e^{-i\omega t + i(\omega/c)\mathbf{n}_r \cdot \mathbf{r}} \end{aligned}$$

where ω is light frequency, c is the speed of light in vacuum and

$$\mathbf{n}_i = n_1 (-\cos \phi, \sin \phi, 0), \quad \mathbf{n}_r = n_1 (\cos \phi, \sin \phi, 0) \quad (1.15)$$

When the p -polarized wave reaches the magnetic medium, it excites a forward propagating wave with refraction vector $\mathbf{n}_t = n_t (-\cos \phi_t, \sin \phi_t, 0)$ with $n_t = n_2 (1 - Q^2) \approx$

n_2 . Applying Snell's law, it proceeds that $\sin \phi_t = (n_1/n_2) \sin \phi$ and $\cos \phi_t = \left(1 - (n_1/n_2)^2 \sin^2 \phi\right)^{1/2}$. The electric field of this mode is described by

$$\mathbf{E}_t = E_t \begin{pmatrix} -\sin \phi_t + iQ/\cos \phi_t \\ -\cos \phi_t \\ 0 \end{pmatrix} e^{-i\omega t + i(\omega/c)(\mathbf{n}_t \cdot \mathbf{r})}$$

This wave is elliptically polarized in the plane of incidence and contains an additional longitudinal component in the wave-vector direction proportional to Q , and thus to the magnetization magnitude. This component is responsible for the magneto-optical effect.

The formulae for reflection and transmission coefficients in the Q linear approximation are given by [7]:

$$\tilde{t}_{12}^p = \frac{E_t}{E_i} = t_{12}^p + i\delta t_{12}^p = t_{12}^p \left(1 + i\tau_{12}^p\right) \quad (1.16)$$

$$\tilde{r}_{12}^p = \frac{E_r}{E_i} = r_{12}^p + i\delta r_{12}^p = r_{12}^p \left(1 + i\rho_{12}^p\right) \quad (1.17)$$

with correction factors δt_{12}^p , δr_{12}^p , τ_{12}^p and ρ_{12}^p equal to

$$\delta t_{12}^p = \frac{2 \sin^2 \phi \cos \phi_t}{(\eta \cos \phi + \cos \phi_t)^2} Q \quad \delta r_{12}^p = -\frac{2 \sin \phi \cos \phi}{(\eta \cos \phi + \cos \phi_t)^2} Q \quad (1.18)$$

$$\tau_{12}^p = \frac{(1 + r_{12}^p) Q \sin \phi}{2 (\eta^2 - \sin^2 \phi)^{1/2}} \quad \rho_{12}^p = \frac{r_{12}^p Q \sin \phi}{2 (\eta^2 - \sin^2 \phi)^{1/2}} \quad (1.19)$$

being t_{12}^p and r_{12}^p the conventional Fresnel coefficients

$$t_{12}^p = \frac{2 \cos \phi}{\eta \cos \phi + \cos \phi_t} \quad r_{12}^p = \frac{\eta \cos \phi - \cos \phi_t}{\eta \cos \phi + \cos \phi_t} \quad (1.20)$$

A measure for the magnitude of the TMOKE is the relative change in reflected light intensity for the magnetized and non-magnetized states of the magnetic media, determined by

$$\delta_p = \frac{I - I_0}{I_0} = 2\text{Im}\rho_{12}^p \quad (1.21)$$

where I and I_0 are the intensities of reflected light in the magnetized and non-magnetized states, respectively.

For s-polarized light, only the z aligned component is non-zero and both the reflected and propagating waves, $\mathbf{E}_r$ and $\mathbf{E}_t$, are s-polarized and do not contain linear Q corrections, and therefore no magneto-optical effects are present. It is important to note that this is only true under the $\mu = 1$ assumption. If that is not the case, then the transverse

Kerr effect for s-polarized waves is proportional to the magneto-optical parameter Q in the off-diagonal entries of the permeability tensor. However, the magnitude of μ is very small and any attempt to attribute it any physical significance has no real justification at optical frequencies [3]. Besides, including the effects of μ would complicate the analytical formulation beyond its actual contribution to measurable effects.

1.3 Surface Plasmon Resonance

The excitation of ESWs in structures containing magnetic films has been reported to enhance magneto-optical effects, while the magnetic modulation of such magnetic films can in turn manipulate the excitation conditions of ESWs [8].

Surface plasmon resonances (SPR) are ESWs triggered in metal-dielectric interfaces. Depending on the wave vector magnitude of incident light, the free electrons at the interface are known to resonate along the EM field oscillations of the radiation. This resonance creates charge density waves that propagate parallel to the interface called surface plasmon polaritons. The *polariton* term at the end refers to the coupling of light to the polarization within the media, an effect that influences the properties of the surface wave.

In order to demonstrate the physical origin of SPRs, one can start from the wave equations, that can be derived from the Maxwell's equations introduced in 1.1

$$\nabla^2 \mathbf{E} - \mu_0 \epsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} = 0, \quad \nabla^2 \mathbf{H} - \mu_0 \epsilon_0 \frac{\partial^2 \mathbf{H}}{\partial t^2} = 0 \quad (1.22)$$

Here, the absence of external magnetic fields incident on an isotropic layer will void the need for the more complex tensorial formalism. Instead, the dielectric properties of a medium can be readily described by a complex number. The solutions to these wave equations take the form of monochromatic plane waves that can be described as

$$\mathbf{E}(\mathbf{r}, t) = E_0 e^{\mathbf{k} \cdot \mathbf{r} - \omega t}, \quad \mathbf{H}(\mathbf{r}, t) = H_0 e^{\mathbf{k} \cdot \mathbf{r} - \omega t} \quad (1.23)$$

with E_0 and H_0 being the electric and magnetic field amplitudes, respectively, ω the angular frequency of the field, $\mathbf{r}$ the position vector and $\mathbf{k}$ the wave vector.

Consider the incidence of polarized light in an interface between two layers of permittivities ϵ_1 and ϵ_2 , as illustrated in 1.2, and convention the superscript $+$ ($-$) to indicate propagation in the positive (negative) z direction, and the subscript to indicate the medium, 1 and 2. This way, an incident wave of amplitudes (E_1^+, H_1^+) is partially transmitted and reflected with amplitudes (E_2^+, H_2^+) and (E_1^-, H_1^-) , respectively.

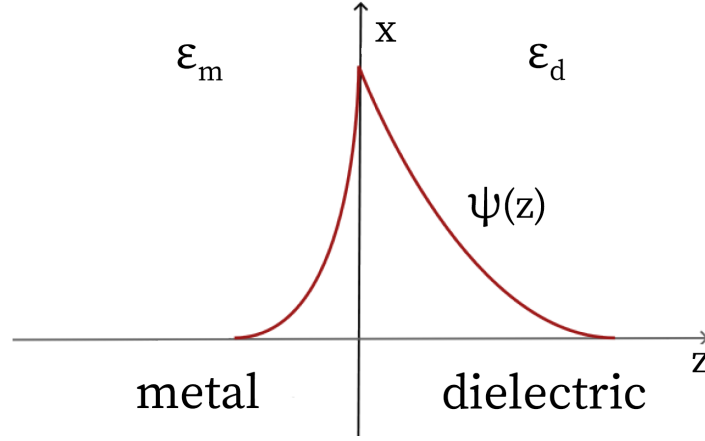


FIGURE 1.2: Schematic representation of field decay at a metal-dielectric interface. Notice the different decay rates for each side.

For light of p polarization, the electric field vector has two components, in x and z , whereas the magnetic field contains the y component, normal to the plane of incidence.

$$\mathbf{E} = (E_x, 0, E_z), \quad \mathbf{H} = (0, E_y, 0) \quad (1.24)$$

The tangential components of the EM field at each side must be continuous, rendering

$$E_{1x}^+ + E_{1x}^- = E_{2x}^+, \quad H_{1y}^+ + H_{1y}^- = H_{2y}^+ \quad (1.25)$$

Combining equations 1.25, 1.1-1.4, 1.23, 1.5 and 1.6, one obtains

$$\mathbf{k} \times \mathbf{H} = -\frac{\epsilon_1 \omega}{c} \mathbf{E} \quad (1.26)$$

and using 1.24, it results that

$$\epsilon_1 \omega E_{1x}^+ = k_{1z} H_{1y}^+, \quad \epsilon_1 \omega E_{1x}^- = -k_{1z} H_{1y}^- \quad (1.27)$$

together with an analogous relation for the second medium. Inserting last equation in 1.25 yields

$$\frac{\epsilon_1 \omega}{k_{1z}} E_{1x}^+ - \frac{\epsilon_1 \omega}{k_{1z}} E_{1x}^- = \frac{\epsilon_2 \omega}{k_{2z}} E_{2x}^+ - \frac{\epsilon_2 \omega}{k_{2z}} E_{2x}^- \quad (1.28)$$

Consider the case of a plane wave incident on a planar metal-dielectric interface. The electric field intensity of the surface wave is maximum at this interface, and decays exponentially with the distance to the interface, through the dielectric and metal media of electric permittivities ϵ_d and ϵ_m , respectively, as represented in 1.2. The EM field in each region can be described as

$$\psi_m(z) = \psi(0)e^{-ik_{mz}z}, \quad z < 0 \quad (1.29)$$

$$\psi_d(z) = \psi(0)e^{ik_{dz}z}, \quad z > 0 \quad (1.30)$$

where k_{mz} and k_{dz} are the perpendicular components of the wave vectors to the interface in media ϵ_m and ϵ_d . Given the exponential decay of the fields, these components can be written as

$$k_{mz} = ik_m \quad \text{and} \quad k_{dz} = ik_d, \quad k_m, k_d \in \mathbb{R} \quad (1.31)$$

Accounting for the continuity condition of 1.28 and equation 1.31, the dispersion relation for both p and s polarization, considering $\mu = 1$, can be represented as

$$\frac{k_m}{\epsilon_m} = -\frac{k_d}{\epsilon_d}, \quad p - \text{polarization} \quad (1.32)$$

$$k_m = -k_d, \quad s - \text{polarization} \quad (1.33)$$

Upon inspection of equations 1.32 and 1.33, one notices 1.32 implies that the sign of the permittivities at each side of the equation must be opposite. On the other hand, 1.33 shows no valid solutions. Therefore, excitation of surface plasmon resonances is not possible for s -polarized light. Additionally, the sign restriction to a solution of equation 1.32 confirms that the excitation of this kind of surface wave is only possible for an interface of a metal (or a heavily doped semiconductor), of negative permittivity, and a dielectric, of positive permittivity. Recasting 1.32 using $k_x = \sqrt{k^2 - k_z^2}$, the dispersion relation of this resonant quasi-particle is given by

$$k_x = \frac{\omega}{c} \sqrt{\frac{\epsilon_m \epsilon_d}{\epsilon_m + \epsilon_d}} \quad (1.34)$$

This is the dispersion relation of an SPP, and it creates another restriction to the excitation of such surface wave. In order for the parallel component of the wave vector to be a real solution, then

$$\frac{\epsilon_m \epsilon_d}{\epsilon_m + \epsilon_d} > 0 \quad (1.35)$$

Given that the permittivity of a metal is negative and the permittivity of a dielectric is positive, then $\epsilon_m \epsilon_d < 0$, and therefore $\epsilon_m + \epsilon_d$ must be negative. As a result,

$$|\epsilon_m| > \epsilon_d \quad (1.36)$$

is also a necessary condition for the excitation of SPRs between two media.

1.3.1 A Drude model Approach to Surface Plasmon Resonance

The mobility of electrons in metals is a central object of study in solid-state physics. A complete, up to date description of electron behavior within condensed matter will ultimately resort to a quantum mechanical approach. Nonetheless, a characterization of the behavior of surface plasmon resonances can be achieved resorting to the much simpler, classical Drude-Lorentz model. This approach is admissible given that the high density of free carriers in metals results in very short gaps in the energetic levels when compared to thermal excitations at room temperature [9]. In this model, the motion of electrons is modeled as a driven damped harmonic oscillator, coupled to the metallic ions by a spring-like force, and driven by the electric field of incident EM radiation. It assumes, however, that electrons in the last electronic layer are free to move within the metal, permitting the excitation of SPPs.

This apparently simplistic model allows for the study of the dispersion relation of light in different metals, and therefore evaluate the possibility of SPP excitation and its properties. One can start with the Lossy Drude model for metals, with a dispersion relation described by

$$\epsilon = 1 - \frac{\omega_p^2}{\omega(\omega - i\gamma_p)} \quad (1.37)$$

where ω_p is the plasma frequency of the metal and γ_p is the damping factor. The plasma frequency, ω_p , of a metal refers to the natural frequency electrons oscillate around metal ions, and sets a boundary on the interaction of light with matter. For frequencies below plasma frequency, the metal is highly reflective, stopping light propagation in the medium. For higher frequencies than plasma frequency, the metal becomes transparent [10], allowing light transmission. The damping factor quantifies the incidence of absorption processes in the metal, mainly losses due to electron scattering.

The dispersion relation of different metals can be studied using the Drude model. Figure 1.3a shows the dispersion relation of SPPs for different Drude model metals forming an interface with air ($n \approx 1$), according to 1.34. The permittivity of Ag, Au, Fe and Ni metals were determined by 1.37 for referenced values of ω_p and γ_p [11] [12], shown in Table 1.1 (in wavelength units, $\lambda_p = 2\pi/\omega_p$). At low ω and k_x , the dispersion curve of the SPP converges to a light-like behavior for a medium of permittivity ϵ_1 , with $k_x \approx \omega/c\sqrt{\epsilon_1}$. For higher wave vectors k_x , the curve converges to an asymptote. This condition occurs

for equal in magnitude, but opposite in sign values of ϵ_m and ϵ_d , and the limit can be calculated from 1.34 to give

$$\omega_s = \frac{\omega_p}{\sqrt{\epsilon_1 + 1}} \quad (1.38)$$

which is the cutoff frequency of the SPP. For low k_x , the dispersion curves tend to the light line, and for higher k_x approach an asymptotic, cutoff frequency. This cutoff frequency is a function of the plasma frequency, and therefore is different for the different metals.

TABLE 1.1: Plasma frequency (λ_p) and damping factor (γ_p) for Ag, Au, Fe and Ni metals.

	Ag	Au	Fe	Ni
λ_p (nm)	146.73	163.14	303.03	253.81
γ_p (nm)	25830	23393	68027	28409

Other parameters can be derived from the Drude model that can be useful for characterization of structures bearing SPR. The resonant wavelength, the propagation length along the interface and the penetration depth in a medium, for instance, are particularly relevant parameters for implementation of SPR structures in optical sensors.

Starting from the SPP dispersion relation, described by 1.34, the SPP wavelength can be defined as

$$\lambda_{SPP} = \lambda_0 \sqrt{\frac{\epsilon_1 + \epsilon_2}{\epsilon_1 \epsilon_2}} \quad (1.39)$$

with λ_0 being the free space wavelength. Figure 1.3b illustrates the dependence of λ_{SPP} on λ_0 for an interface of different metals with air. It is clear that for higher wavelengths, the SPP wavelength converges to that of free space light. This regime corresponds to the converging low k_x limit seen in Figure 1.3a. In the near ultraviolet, however, the SPP wavelength approaches $\lambda_s = 2\pi c/\omega_s$ – defined from 1.38 as cutoff wavelength – becoming plasma-like and confining light in sub-wavelength structures. The plasma-like behavior for Ag and Au metals occurs for close together wavelengths, owing to the similar optical properties of these noble metals. The higher cutoff wavelength of Fe and Ni films limits the range of possible SPP excitations for different RIs of external media, higher than that of air.

The intensity of the EM fields decays exponentially not only in the normal direction to the interface, as shown in Figure 1.2, but also in the direction of SPP propagation. The optical losses along the interface originate from material absorption in interface media, particularly the metal. The propagation length of an SPP can be calculated from 1.34 by

including the complex component of the permittivities, linked to optical losses. If we define the propagation length as the distance covered until $1/e$ of the intensity is left, and defining $k_x = k'_x + ik''_x$, the propagation length can be calculated by

$$L_{SPP} = \frac{1}{2k''_x} \quad (1.40)$$

Figure 1.3c depicts the propagation length of different metals as a function of the free space wavelength. Notice that for lower wavelengths, near the plasma regime, the propagation length drops sharply, due to higher values of k''_x in the region. The low optical losses known for noble metals are in display here, with times the obtained propagation lengths of more lossy Fe and Ni metals.

The penetration depth can be defined as the distance covered until the EM field intensity exponentially decays by $1/e$ in the direction normal to the interface. The penetration depth is different for each side of the interface, being significantly shorter in metals than dielectrics. The penetration depth in metals is given as $\delta_m = 1/2k_m$, and $\delta_d = 1/2k_d$ for the dielectric counterpart. Resourcing to the dispersion relation 1.34, the penetration depth for the metal and dielectric layers can be calculated by

$$\delta_d = \frac{\sqrt{\epsilon_m}}{k_x \epsilon_d}, \quad \delta_m = \frac{\delta_d \epsilon_d}{\epsilon_m} \quad (1.41)$$

The penetration depth is higher for near infrared wavelengths due to lower incidence of electron scattering, a clearly visible tendency in Figure 1.3d, depicting the penetration depth dependence on the free space wavelength. The penetration depth in the dielectric medium is also significantly higher than in the metallic medium, with both showing an abrupt drop in the near UV, plasma-like regime.

Regarding the studied metals, it is clear that the application of noble metals, as Ag and Au, in SPP excitation is in every way preferable than ferromagnetic metals, as Fe and Ni. Between the two, Ag shows both larger propagation lengths and penetration depths, rendering Ag-based plasmonic structures higher RI sensitivity. The ferromagnetic Fe and Ni show significantly higher optical losses that translate to shorter propagation lengths and penetration depths that halt their utility in sensing applications. Despite the lower losses observed for Ni, its magnetic properties are far reduced when compared to those of Fe, making it of no use in this work.

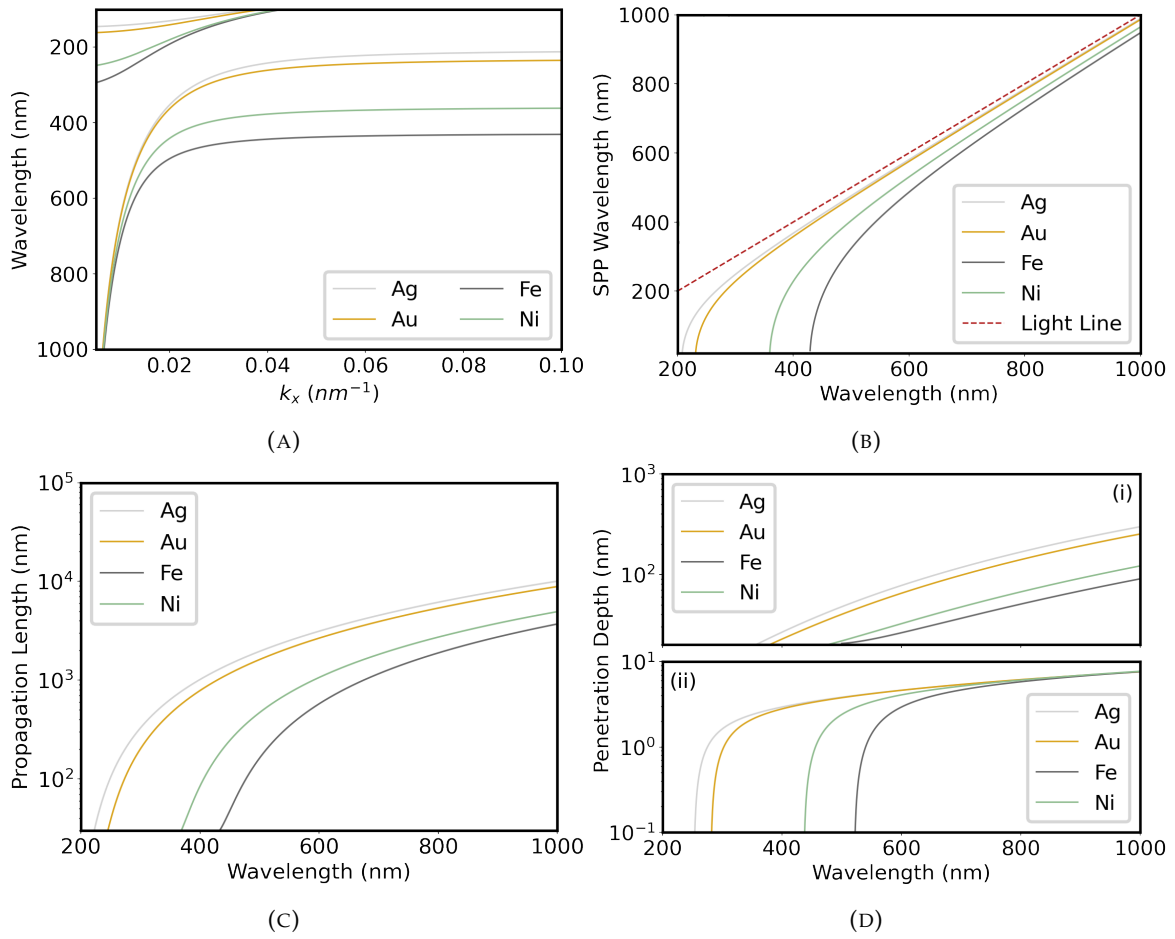


FIGURE 1.3: SPP physical properties for metal-air interfaces: (A) SPP dispersion relation for different lossless metals, according to Drude's model; (B) SPP wavelength for different metals; (C) Propagation length of SPP mode along different metal-air interfaces; (D) Penetration depth of the decaying fields into the (i) air (dielectric) and the (ii) metal, for different metals.

1.3.2 Surface Plasmon Resonance Interrogation

The three described parameters are particularly relevant for the application of SPP bearing structures in optical sensors. The resonant wavelength, given by λ_{SPP} , determines the spectral region where optical interrogation is to be set, which can determine its viability. By being dependent on the optical properties of both media, namely permittivities ϵ_m and ϵ_d , this resonant wavelength can be tracked for variations of these parameters. As detailed in the last section, the intensity of the EM fields decays exponentially away from the interface, and its penetration depth is higher in the dielectric layer, capable of reaching distances well over hundreds of nanometers. If an interface is fabricated with a dielectric medium of inferior thickness to that of the penetration depth, λ_{SPP} will not be dependent exclusively on ϵ_d , but on the external medium surrounding the interface as well, defining

an effective permittivity seen by the decaying EM wave.

Variations on the permittivity of the external medium can thus be tracked upon interrogation of λ_{SPP} , rendering these kind of SPR-bearing structures viable for application in refractive index sensors. Further functionalization of a plasmonic structure to bind to specific molecules enables molecule concentration detection in more complex sensors. The penetration depth, thus, is critical to the sensitivity of the sensor by determining the interaction of the SPP EM field with the analyte. The propagation length ultimately dimensions the sensing region at the interface, a condition that can also raise technical limitations for SPP characterization.

In this work, the prism coupling approach for wave vector matching was considered, using spectral interrogation to track changes in resonant wavelength and reflectance (or absorbance) of captured spectra. As such, light is incident at a fixed angle and a broadband light source and spectrometer are required.

A. Prism Coupling Excitation of Surface Plasmon Resonances

A scheme for the considered Kretschmann configuration is represented in Figure 1.4. White light from a broadband source is coupled into the prism and is incident on the top surface at an angle θ , reflecting with the same angle by attenuated total reflection (ATR) at the interface between the prism and the plasmonic film. The light is then collected by a spectrometer and a peak in the reflectance spectra is noticed due to light being absorbed in the film for SPP excitation.

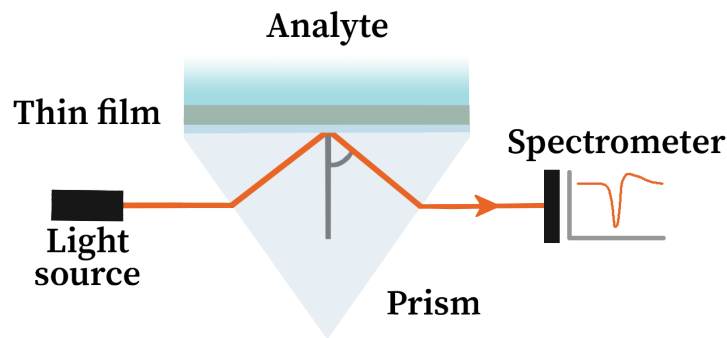


FIGURE 1.4: Kretschmann configuration scheme for surface wave excitation. A prism of high RI is required for wave vector matching.

The SPR sensing structures are thin films of thicknesses frequently smaller than the penetration depth. The interaction of the SPP decaying field with both the media outside the interface, as mentioned before, will rise modifications in the dispersion relation. In

the metal side of the interface, the prism will cause a slight variation on the dispersion relation. In this case, the parallel component of the wave vector k_x^{SPP} can be described by

$$k_x^{SPP} = k_x^{SPP_0} + \Delta k_x = \frac{2\pi}{\lambda} \sqrt{\frac{\epsilon_d \epsilon_m}{\epsilon_d + \epsilon_m}} + \Delta k_x \quad (1.42)$$

where $k_x^{SPP_0}$ is the dispersion relation given by 1.34 and Δk_x is the correction term originating from the prism media interaction with the SPP. In the particular case of prism coupling excitation, the surface plasmon resonance will occur for the wavelength that solves $k_x^{SPP} = (2\pi/\lambda)n_p \sin \theta$, with n_p being the RI of the prism and θ being the angle of light incidence. This equation describes the spectral position where the propagation constants of the SPP and the prism match.

In the dielectric side of the interface, the dispersion relation of the SPP will be influenced, as previously mentioned, by the RI of the external medium. An effective index, generally defined as $n_{eff} = k_x \lambda / (2\pi)$, can be derived to account for the combined effect of both the dielectric medium at the interface and the external medium. In the case of SPP excitation in Kretschmann configuration, the effective index is given by

$$n_{eff} = n_p \sin \theta = \Re \left[\sqrt{\frac{\epsilon_d \epsilon_m}{\epsilon_d + \epsilon_m}} \right] + \Delta n_{eff}^{SPP} \quad (1.43)$$

where $\Delta n_{eff}^{SPP} = \Re \{ \Delta \beta \lambda / 2\pi \}$, with β being the propagation constant. It is therefore noticeable that not only the interface materials, but also the incidence angle and prism refractive index affect the SPR wavelength.

1.4 Bloch Surface Waves

One other kind of surface wave that can be explored for magneto-optical sensitivity are Bloch Surface Waves (BSW). These waves differ fundamentally from plasmonic waves, although some aspects for their excitation and manipulation remain similar.

1.4.1 Photonic Crystals

A BSW is excited at the interface between a photonic crystal (PhC) and a dielectric medium. A PhC is defined as a periodic array of dielectric materials, producing a periodic modulation of the RI that is known to enable the appearance of allowed and forbidden bands of light propagation. The smallest periodic structure, that is then repeated throughout the PhC, is called a unit cell. Different choices of unit cell are possible, and the theoretical

formalism developed around any choice of unit cell is equivalent. Consider the PhC and a possible unit cell depicted in Figure 1.5, composed by materials $n_1 = 2.5$ and $n_2 = 1.45$, and thicknesses $d_1 = 200nm$ and $d_2 = 100nm$. Such unit cell can be thought of as a waveguide, confining light in the media of higher RI with some field penetration in the external medium of lower RI. The dispersion relation of this unit cell is plotted in Figures 1.6(a) and 1.6(b) for p and s -polarized light, respectively.

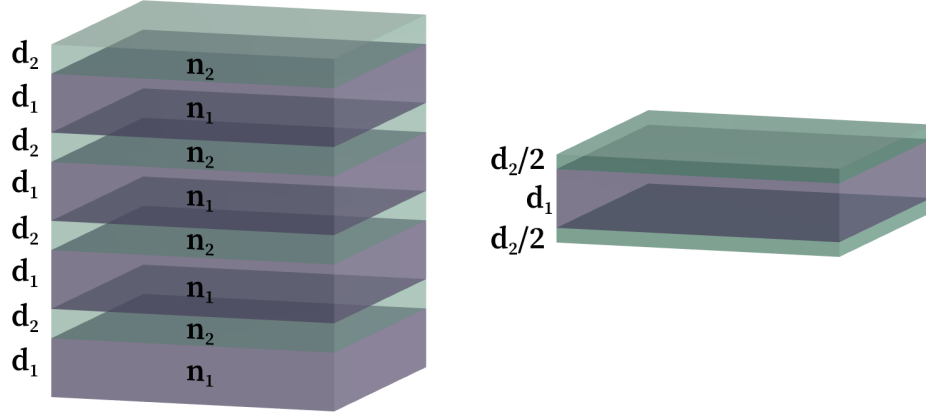


FIGURE 1.5: Illustration of a PhC composed of layers of n_1 and n_2 with thicknesses d_1 and d_2 (left), and representation of a possible unit cell (right).

In this configuration, multiple bound states are found, corresponding to the guided modes seen in a typical waveguide. The different EM modes are identified according to the waveguide nomenclature for transverse modes, describing the polarization state, TM (p-polarization) or TE (s-polarization), and the number of nodes of each mode, subscripted [13]. In fact, this unit cell structure behaves similarly to a waveguide, confining TE and TM modes in the higher index waveguiding layer, while the lower external medium index changes the effective index of the modes, deviating from the single slab waveguide line.

If additional unit cells are stacked contiguously, additional modes are expected to surge. Consider the dispersion relations (Figure 1.7) of the PhC depicted in Figure 1.5. Plenty of guided modes are found at low values of k , that seem to converge down to few concise modes at higher k . The narrowing down actually translates to a stronger confinement of the fields inside the higher index layers, impeding the different EM modes to interact with each other. At low k , such confinement is lifted and mode interference leads to mode splitting into four new non-degenerate modes. Since these modes arise from degeneracy lifting of fundamental modes, Yeh [14] proposed the insertion of an additional digit in the nomenclature, distinguishing between different modes. In this nomenclature,

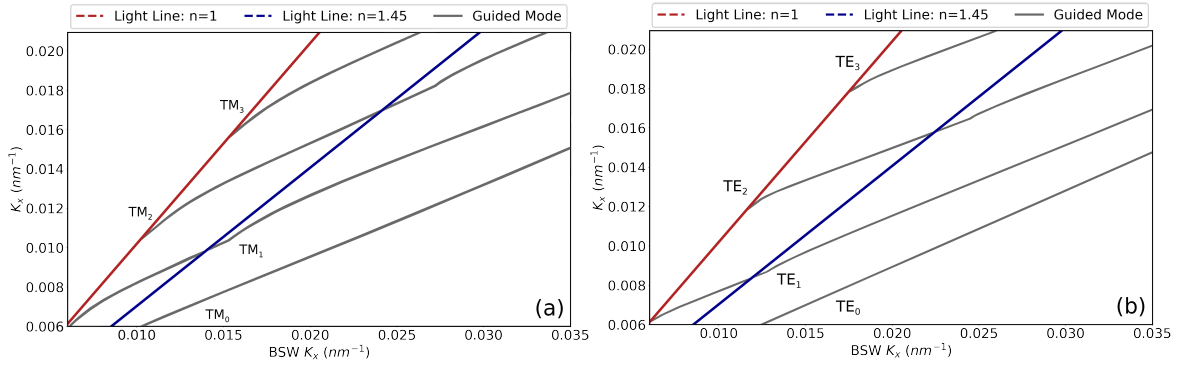


FIGURE 1.6: Dispersion relation of unit cell of Figure 1.5 for (a) p-polarization, displaying modes TM_0 to TM_3 and (b) s-polarization, displaying modes TE_0 to TE_3 .

a PhC of N unit cells will hold modes $TM_{00}, TM_{01}...TM_{0N-1}, TM_{10}, TM_{11}...TM_{1N-1}$, and so on.

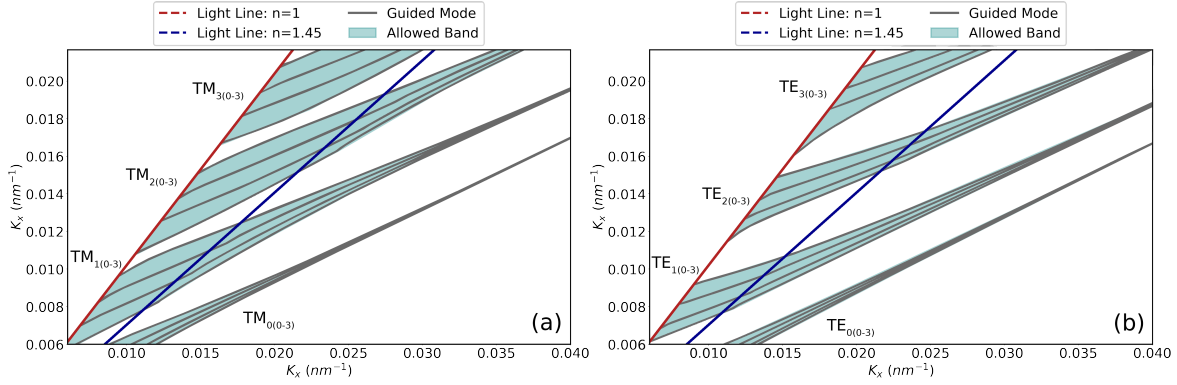


FIGURE 1.7: Dispersion relation of PhC of Figure 1.5 for: (a) p-polarization and (b) s-polarization. Notice how the stacking of N unit cells raises N additional modes around each unit cell mode.

Taking the limit of an infinite number of unit cells, the periodic modulation of the RI can be written as

$$n(z) = \begin{cases} n_1, & m\Lambda < z < m\Lambda + d_1 \\ n_2, & m\Lambda + d_1 < z < (m+1)\Lambda \end{cases}, \quad m \in \mathbb{N} \quad (1.44)$$

where $\Lambda = d_1 + d_2$ is the unit cell length, and over the course of which the RI is periodic, with $n(z + \Lambda) = n(z)$.

Proceeding from the wave equation (1.22) and respective monochromatic plane wave solution (1.23), one can obtain

$$(\nabla^2 - \mu_0 \epsilon \omega^2) \mathbf{E} = 0 \quad (1.45)$$

Given the periodic nature of the permittivity in 1.45, as inferred from the periodic RI in 1.44, an analysis of this equation renders it very similar to Schrodinger's equation for an electron in a crystal, and therefore passive to be applied to a Phc, with EM fields inside governed by Bloch's theorem:

$$\Psi(z) = \Psi_K(z) e^{iKz} \quad (1.46)$$

where $\Psi_K(z)$ is a periodic function of same period as the 'lattice' and K is Bloch's number, a constant in which $\Psi_K(z)$ also depends. The Bloch's number also describes the dispersion relation of a BSW propagating inside a PhC, given by

$$K(k_x, \omega) = \frac{1}{\Lambda} \arccos \frac{1}{2} (A + D) \quad (1.47)$$

A and D are obtained by transfer matrix propagation of the considered structure and its derivation was thoroughly demonstrated by Yeh [14]. A brief analysis of Equation 1.47 allows us to notice the existence of two regimes. For $|1/2(A + D)| < 1$, $K \in \mathbb{R}$ and the equation corresponds to a Bloch wave as described by 1.46; for $|1/2(A + D)| > 1$ and $K = m\pi/\Lambda + iK_i$, with $m \in \mathbb{Z}$ and $K \in \mathbb{C}$, and K_i being the imaginary part, the solutions correspond to evanescent waves in the Phc and therefore do not constitute propagating modes in the structure. These two regimes describe the allowed and forbidden bands of light propagation in PhC, that are ultimately defined by the Bloch's number, that will be real in allowed bands and complex in forbidden bands. As predictable, the degeneracy lifting of infinite modes leads to the appearance of continuous bands of allowed states for light propagation, as visible in Figure 1.7, and forbidden states that reflect light off of the PhC.

These allowed and forbidden bands in PhCs are widely applied in the fabrication of optical filters and anti-reflection coatings. However, the fabrication of infinite PhCs can be time-consuming, so that for all practical applications, structures with a finite number of unit cells are usually considered.

1.4.2 Truncated Photonic Crystals

A particular configuration of PhCs involves topping off a regular PhC of N unit cells with a capping layer of the high index material n_1 of smaller thickness, $d_s = 100nm$, as represented in 1.8. Such truncated layer can be added to excite a BSW by breaking the symmetry of the periodic modulation of the RI. The symmetry breaking is a result of a

phase that is added to the forward and backward propagating waves. These forward and backward propagating waves are represented as

$$\Psi(z) = a_s e^{ik_{2z}z} + b_s e^{-ik_{2z}z} \quad (1.48)$$

where a_s and b_s are the amplitudes of the forward and backward propagating waves, respectively. In the truncated layer, the added phase leads to a modulation of a_s and b_s given by

$$\begin{pmatrix} a_s \\ b_s \end{pmatrix} = \frac{1}{2} \begin{pmatrix} B e^{-2ik_z d_s} \\ e^{iK\Lambda} - A \end{pmatrix}$$

The added phase in the truncated layer will thus modulate the BSW dispersion relation. In fact, taking in account the exponential decay of the fields outside of the PhC,

$$\Psi(z) = \alpha e^{q_0 z} \quad (1.49)$$

with $q_0 = \sqrt{k_x^2 - k_{z0}^2}$, and taking the continuity conditions of the fields and respective derivatives in 1.48 and 1.49 at the interface of the PhC with the external medium, it can be noticed that

$$q_0 = -ik_1 \frac{e^{iK\Lambda} - A - B e^{-2ik_z d_s}}{e^{iK\Lambda} - A + B e^{-2ik_z d_s}} \quad (1.50)$$

Considering the truncated PhC with 4 unit cells with a capping layer of 100nm, as represented in Figure 1.8, and in contact with an external medium of index $n_0 = 1$, the dispersion relation, given by 1.50, can be calculated numerically and plotted, as shown in Figure 1.9. One can now visualize the dispersion lines of BSW modes within the forbidden bands. Notice also how the BSW dispersion lines lie below the dispersion line of light, resulting that direct excitation of BSW is also not possible, and a coupling prism, grating or waveguide will be needed for BSWs excitation. For the dispersive medium of a silica prism ($n \approx 1.45$), the excitation of both guided and BSW modes is attainable at optical frequencies and can be estimated from Figure 1.9 through the intersection of guided and BSW mode curves with the dispersive curve of light propagating through the prism, being the wavelength of excitation given simply by $\lambda = 2\pi/k_x$. Upon close inspection, a BSW mode is attainable in a prism coupling scheme around $k_x = 0.007nm^{-1}(897nm)$ for p-polarization, and two modes around $k_x = 0.0055nm^{-1}(1142nm)$ and $k_x = 0.0182nm^{-1}(345nm)$.

The excitation of BSW modes is a result of both symmetry breaking of the PhC and the interaction of fields in the layers of higher RI, that behave as waveguides. For large

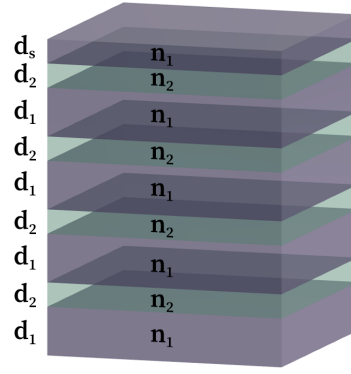


FIGURE 1.8: Illustration of a truncated PhC composed of layers of n_1 and n_2 with thicknesses d_1 and d_2 . At the top, a capping layer of n_1 with thickness d_s breaks RI symmetry of the PhC.

waveguide separations, these waveguide modes are degenerate, but when the separation is short enough for the fields to interfere with each other, this degeneracy is lifted, and an allowed band is formed. This band supports the N modes of the original degenerate state, and the non-degenerate mode associated to the truncated layer can only propagate within the forbidden band. From 1.47, a mode propagating in the forbidden band has a complex K , and as such, there is an exponential decay of the field inside the PhC. The coexistence of exponentially decaying fields inside and outside of the PhC corresponds to the surface wave solution known to BSWs. Figure 1.10 shows the reflectance spectrum of the structure depicted in Figure 1.8. As expected, 4 non-degenerate guided modes are seen in the allowed bands, and a BSWs are seen in forbidden bands, or bandgaps, at matching wavelengths of those estimated in Figure 1.9

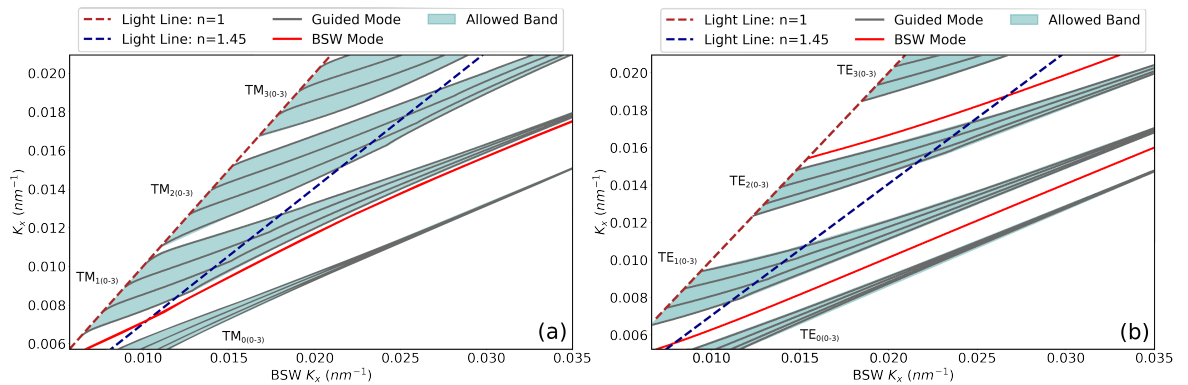


FIGURE 1.9: Dispersion relation of truncated PhC of Figure 1.8 for: (a) p-polarization and (b) s-polarization. Notice the propagation of BSW modes outside the allowed bands

The symmetry breaking leads to a significant field amplification in the truncated layer,

and the subsequent exponential decay both inside and outside of the PhC, enabling sensing capabilities. The sensing capabilities of BSWs can be characterized by the same physical parameters seen for SPPs: mode wavelength, propagation length and penetration depth. In the case of BSWs, however, these properties have to be determined numerically, making it harder to determine explicit dependencies of such physical parameters on the PhC design.

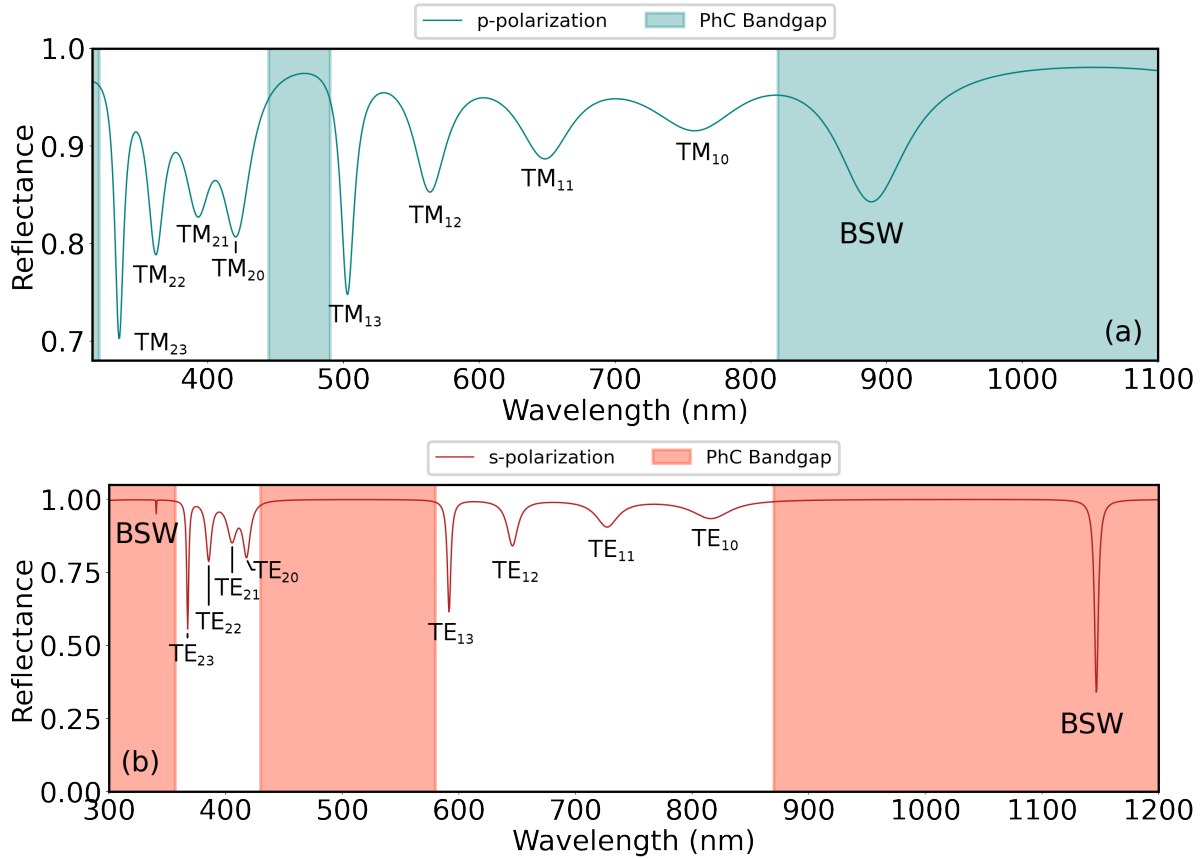


FIGURE 1.10: Reflectance spectra of truncated PhC in 1.8 for $n_1 = 2.5$, $d_1 = 200nm$, $n_2 = 1.45$, $d_2 = d_s = 100nm$ for (a) p-polarized and (b) s-polarized light and external medium of air ($n = 1$).

The propagation length of conventional BSW modes in a PhC does not face the constraints present in SPP propagation given the absence of a metal layer. PhCs are virtually lossless: their all-dielectric nature renders them little to no absorption, and the interfaces are usually smooth enough to avoid significant scattering losses. In the particular case of BSW excitation by the Kretschmann configuration, it was found that most losses are due to field leakage into the prism, that still allowed a propagation length over $3mm$ [15].

The penetration depth of a BSW is different for decay into the PhC or into the external dielectric. The decay into the dielectric has been treated before 1.49, being given by

$$L_{BSW}^d = \frac{\lambda}{4\pi\sqrt{n_{eff}^2 - n_0^2}} \quad (1.51)$$

In the case of decay into the PhC, analysis of 1.46 for a BSW ($K \in \mathbb{C}$) renders that the penetration depth can be defined simply as

$$L_{BSW}^{PhC} = \frac{1}{2K_i} \quad (1.52)$$

The all dielectric composition of truncated PhCs and their inherent lower losses can be useful in magnetophotonics, as larger thicknesses of magnetic materials, usually of high optical losses, can be inserted within controlled broadening of spectral bands.

1.5 Magneto-Optical Effects in Electromagnetic Surface Waves

Magneto-optical effects are small, and their minute expression prevents widespread application of magneto-optics in applied magnetophotonics. The relative change of the reflected light intensity in TMOKE, for instance, is in the order of 10^{-3} . The enhancement of magneto-optical effects is possible, and has been achieved by different approaches. The optimization and development of new materials such as ferromagnetic dielectric garnets has enabled increased magneto-optical responses, either by increased magnetic permeabilities or reduced optical losses, that allow scaling of magneto-optical structures, something challenging for ferromagnetic metals. The use of nanostructured materials can also enhance magneto-optical effects by geometrical resonances instead of electronic ones.

One other interesting approach to boost magneto-optical effects involves the use of surface plasmon resonances to increase the efficiency of magneto-optical interactions, by concentrating EM fields near an interface with a magnetized medium. In fact, the merge of plasmonics with magneto-optics seems to be symbiotic, as not only SPR excitations can be used to enhance the magneto-optical effects, but magnetic fields can also be used to control the properties of an SPP.

1.5.1 Magnetoplasmonics

The first reports on the interaction between SPPs and magneto-optics were observed for SPP propagation along the surface of a ferromagnetic metal or semiconductor film under an external magnetic field. An increased TMOKE was reported for long range SPPs in nickel films [16]. Bilayers of noble and ferromagnetic layers were observed to bear SPPs

at the interface, enhancing the TMOKE taking place at the ferromagnetic surface. The replacement of the ferromagnetic metal layer by ferromagnetic dielectric layers was observed to permit higher enhancement of TMOKE, however still short to the enhancement attained by tailored nanostructuring of noble metal and ferromagnetic dielectric garnets. In such structures, gratings are used to excite EM fields near zones of intense TMOKE.

As demonstrated previously, the transverse magnetization does not change the polarization state of light reflecting off a magnetic film, in contrast to other possible magnetization directions. The TMOKE signal of relative reflected intensity of a magnetoplasmonic structure, combining plasmonic and magneto-optical properties, can be determined in an analogous fashion with 1.14, by

$$\frac{\Delta R_{pp}}{R_{pp}} = \frac{R_{pp}(+M) - R_{pp}(-M)}{R_{pp}(-M)} \quad (1.53)$$

where $R_{pp}(+M)$ and $R_{pp}(-M)$ are the reflectance spectra of the magnetoplasmonic structure for positive and negative saturation magnetizations of the ferromagnetic layer, respectively. This signal oftentimes depicts a sharp peak in intensity due to a shift in the resonance wavelength of the SPP, that is in turn a direct consequence of the modulation of the dispersion relation of the SPP for transverse magnetic field incidence.

The obtained spectra from these operation enables a visualization of the magneto-optical effect in a plasmonic structure, usually displaying a sharp peak in intensity near the SPR wavelength. This modulation of SPR spectra, from now on referred as magneto-optical SPR – MOSPR – spectra, can be applied in the sensing of small refractive index variations by controlling the reflectance changes.

As the external refractive index changes, the SPR band will shift, altering the measured reflectance at a given referenced wavelength. The spectral sensitivity of the change in reflectance for external index variations for SPR and MOSPR measurements can be defined as

$$\eta_{SPR} = \frac{\partial R_{pp}}{\partial n_d} = \frac{\partial R_{pp}}{\partial \lambda} \frac{\partial \lambda}{\partial n_d} \quad (1.54)$$

$$\eta_{MOSPR} = \frac{\partial(\Delta R_{pp}/R_{pp})}{\partial n_d} = \frac{\partial(\Delta R_{pp}/R_{pp})}{\partial \lambda} \frac{\partial \lambda}{\partial n_d} \quad (1.55)$$

The plot for the variations of η_{SPR} and η_{MOSPR} with interrogated wavelength will return a peak of highest sensitivity, corresponding to the ideal interrogation wavelength. For certain configurations, the high slope of the MOSPR modulation will render a higher

sensitivity than the SPR counterpart, thus increasing the capacity of a given sensing structure [17].

This change in light-SPP coupling conditions reflects a non-reciprocity effect in SPP propagation, induced by the magnetization of a ferromagnetic layer [18]. The dispersion relation is dependent on the materials and the geometry of the SPP carrying structure. Neglecting the effect of field leakage into the prism, introduced in 1.3, the dispersion relation of an SPP propagating along a transversely magnetized medium is given by

$$k_x^{SPP}(M) = k_x^{SPP_0} + \Delta k_x(M) \quad (1.56)$$

where $k_x^{SPP_0}$ is the SPP wave vector for a non-gyrotropic interface, as described by 1.34, and $\Delta k_x(M)$ is the magnetization induced modulation of the surface plasmon wave vector [19]. An exact analytical description of this term depends on the materials and respective geometry at the interface, and is sometimes impossible to obtain. For an interface composed of a plasmonic metal and a dielectric, with a thin layer of a ferromagnetic metal embedded within the plasmonic metal, the magneto-optical modulation can be approximated to [20]

$$\Delta k_x(\epsilon_{xz}) = \frac{2t_{fm}(k_0\epsilon_d\epsilon_m)^2 i\epsilon_{xz}}{(\epsilon_d + \epsilon_m)(\epsilon_d^2 - \epsilon_m^2)\epsilon_{fm}} e^{-2h \cdot k''_{z,m}} \quad (1.57)$$

Considering $|\epsilon_m| \gg |\epsilon_d|$, one can simplify the expression to

$$\Delta k_x(\epsilon_{xz}) \approx \frac{i2t_{fm}k_0^2\epsilon_d^2\epsilon_{xz}}{-\epsilon_m\epsilon_{fm}} e^{-2h \cdot k''_{z,m}} = \frac{i2t_{fm}k_0^2\epsilon_d^2\epsilon_{xz}}{-\epsilon_m\epsilon_{fm}} e^{-h/\delta_m}, \quad (1.58)$$

where $k''_{z,m}$ is the imaginary part of the transverse component of the SPP wave vector in the plasmonic metal layer, δ_m is the penetration depth of the SPP EM fields in the plasmonic metal, t_{fm} is the thickness of the ferromagnetic layer, h is the distance of the ferromagnetic layer to the interface, k_0 is the wave vector of the incident light in vacuum, ϵ_m is the isotropic permittivity of the noble metal, ϵ_d is the permittivity of the dielectric layer, and ϵ_{fm} and ϵ_{xz} are the diagonal and off-diagonal entries of the isotropic tensorial permittivity of the ferromagnetic layer, respectively.

In the limit where the ferromagnetic layer is adjacent to the interface ($h \rightarrow 0$), the magnetic field modulation to the dispersion relation of the propagating SPP can be approximated by

$$\Delta k_x(\epsilon_{xz}) \approx \frac{i2t_{fm}k_0^2\epsilon_d^2\epsilon_{xz}}{-\epsilon_m\epsilon_{fm}} \quad (1.59)$$

Figure 1.11 depicts the dispersion relation of a magnetoplasmonic structure, evidencing the non-reciprocity of the wave vector of opposite saturation magnetizations, according to 1.59. This magneto-optical modulation is noticeably small, and its spectral dependence can be vaguely perceived by regions of higher or lower divergence of the curves for positive and negative magnetization. Upon close inspection, the wave vector shift introduced by the modulation of 1.59 equals a wavelength shift close to $3nm$ around the resonance wavelength of $818nm$. As will be shown further in Chapter 3, these values are well within the range of obtained results.

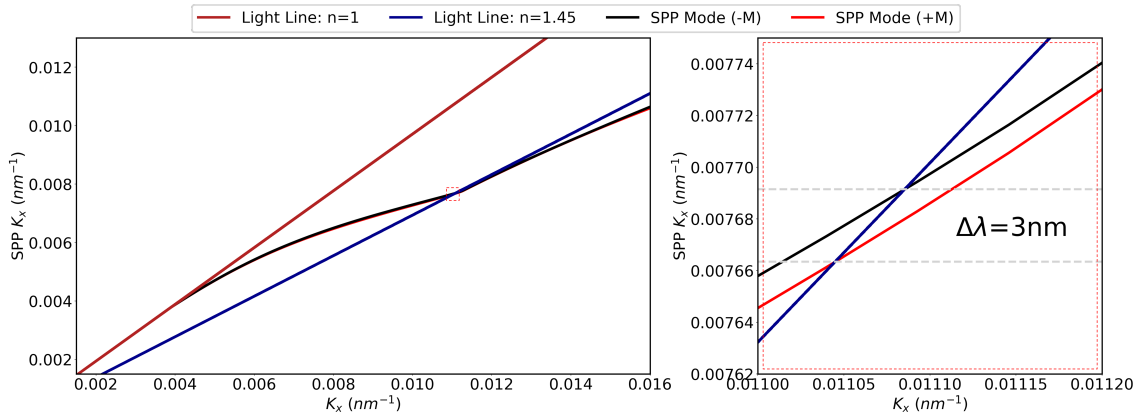


FIGURE 1.11: Dispersion relation of magnetoplasmonic structure 30nm Ag/10nm Fe/40nm TiO₂ for $n = 1$ and p-polarized light. Notice the splitting of the curves for opposite state magnetizations, that ultimately leads to a shift of resonance frequency.

The increase in TMOKE by underlying SPP excitation is due to a shift of the SPP resonance wavelength in transverse magnetic fields, and such shift is only significant for propagating SPPs [21]. For localized SPPs, the resonance frequency is hardly influenced by the magnetic field.

1.5.2 Bloch Surface Waves in Magnetophotonic Crystals

As seen before, the periodic constitution of PhCs affects the propagation of light, providing a new mechanism for light control. These structures are sensitive to variations of external dielectric medium, and this sensitivity can be tuned by PhC design of layered materials and respective thicknesses. If the constitutive materials of a PhC (or an inserted perturbative layer) are magnetic, the resultant PhC can additionally be sensitive to magnetic fields and magnetizations, and exhibit magneto-optical responses [22].

In fact, the strong confinement of BSWs in a PhC with a magnetic defect has been used to enhance magneto-optical effects, with modulation of both light polarization and intensity. As introduced in 1.2.1, for the case of transverse incidence of magnetic fields to light propagation (TMOKE), no change in the polarization plane is induced for p-polarized light, and only a modulation on reflected intensity is noticeable.

The study of magneto-optical effects in conventional PhCs with a planar ferromagnetic metal layer is not well documented, probably due to the high losses induced by the metallic ferromagnetic layer. Additionally, the field confinement in BSWs is smaller than in plasmonic films, limiting the enhancement of magneto-optical effects for sensing applications. Not surprisingly, the study of magneto-optical effects in PhCs is more reported for PhCs with dielectric ferromagnetic garnets or in PhCs put together with a plasmonic metal layer, such as Ag or Au, exciting another kind of surface wave called Tamm Plasmon Polariton, that although being of interesting properties, are beyond the scope of this work.

Nonetheless, an experimental study on the change in reflected intensity can still be made, as defined in 1.5.1. The highest definition of Bloch modes compared to SPR bands allows for the use of larger thicknesses of metallic ferromagnetic layers that may compensate for decreased magneto-optical enhancement.

Chapter 2

Materials and Methods

Along the following chapter, all methods and techniques addressed in simulation and experimental work are introduced. In section 2.1, the numerical implementation of Berreman's transfer matrix method is detailed, and some considerations on setup limitations and optimization of input parameters to account for particularities of planar substrate and multimode fiber geometries are described. In section 2.2, a complete guide on the setup used for thin film fabrication is described, addressing the sputtering deposition setup and the calibration process for film thickness precision. A description of all the characterization techniques is then addressed in section 2.3. These characterization techniques unveil the magnetic and magneto-optical properties of the fabricated films, and their working principle and setup description are presented.

2.1 Simulation Work

2.1.1 Numerical Implementation

A Python language based numerical implementation of the 4×4 Berreman's transfer matrix method [23][24] was carried out for the simulation of reflectance spectra of multilayer thin film stacks starting from the tensorial permittivities of desired materials, extracted from literature.

Considering a stack of N layers normal to the z axis and invariant in the xy plane. Each layer i is bounded by two interfaces at z_i and z_{i+1} , separated by a layer thickness of h and described by a permittivity tensor ϵ_i . The layer stack is bounded between two semi-infinite isotropic media, n_{in} and n_{out} , from where light enters and leaves the stack, respectively. A diagram of such configuration is depicted in Figure 2.1.

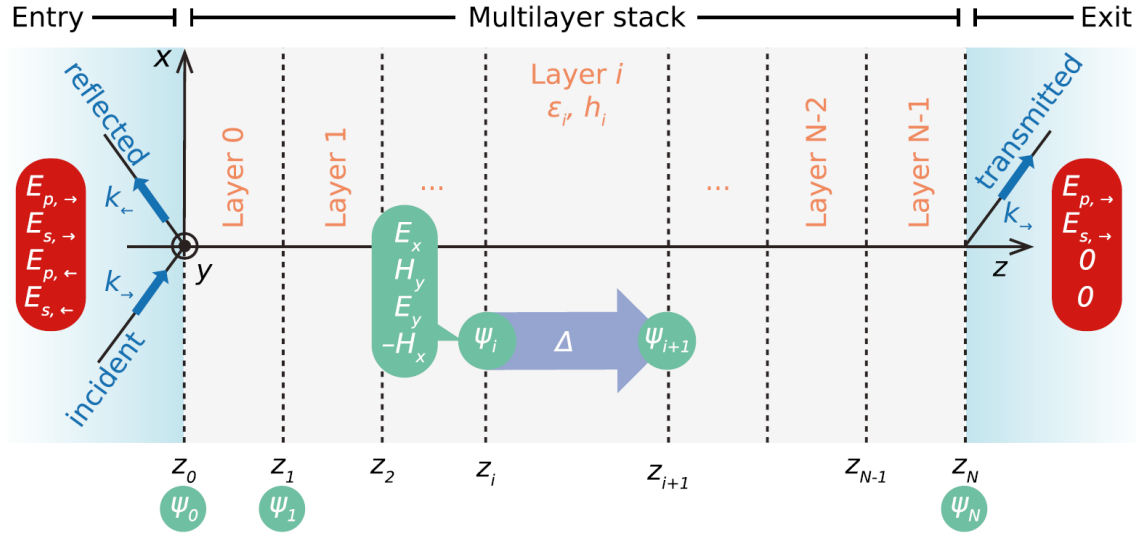


FIGURE 2.1: Diagram of forward and backward propagating partial waves through a stack of N layers along z direction [24].

A plane-wave of frequency ω is incident on this multilayer stack with a θ_{in} angle from the surface normal, propagating through the stack in the xz plane. The wavevector $\mathbf{k}$ of this incident plane wave is given by

$$\mathbf{k} = \begin{bmatrix} \vec{k}_x \\ \vec{k}_y \\ \vec{k}_z \end{bmatrix} = k_0 \begin{bmatrix} K_x \\ K_y \\ K_z \end{bmatrix}$$

being $k_0 = 2\pi/\lambda$, $K_x = n_{in} \sin \theta_{in}$ and $K_y = 0$ are constant throughout the stack.

In isotropic layers, plane waves can be decomposed into s and p polarisations. The p -polarised waves have its electric field parallel (and its magnetic field perpendicular) to the plane of incidence xz , whereas s -polarized waves have its electric field perpendicular (and its magnetic field parallel) to the plane of incidence.

Maxwell's equations in gaussian units and rectangular coordinates may be written in the 6×6 matrix form

$$\begin{bmatrix} 0 & 0 & 0 & 0 & -\frac{\partial}{\partial z} & \frac{\partial}{\partial y} \\ 0 & 0 & 0 & \frac{\partial}{\partial z} & 0 & -\frac{\partial}{\partial x} \\ 0 & 0 & 0 & -\frac{\partial}{\partial y} & \frac{\partial}{\partial x} & 0 \\ 0 & \frac{\partial}{\partial z} & -\frac{\partial}{\partial y} & 0 & 0 & 0 \\ -\frac{\partial}{\partial z} & 0 & \frac{\partial}{\partial x} & 0 & 0 & 0 \\ \frac{\partial}{\partial y} & -\frac{\partial}{\partial x} & 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \\ H_x \\ H_y \\ H_z \end{bmatrix} = \frac{1}{c} \frac{\partial}{\partial t} \begin{bmatrix} D_x \\ D_y \\ D_z \\ B_x \\ B_y \\ B_z \end{bmatrix}$$

being c the velocity of light in vacuum and $\vec{E}$, $\vec{H}$, $\vec{D}$ and $\vec{B}$ the electric, magnetic, electric displacement and magnetic induction electromagnetic fields, respectively. The permittivities ϵ , permeabilities μ , and optical activity tensors ρ and ρ' can be inserted to relate $\vec{D}$ and $\vec{B}$ to $\vec{E}$ and $\vec{H}$ fields, yielding

$$\begin{bmatrix} 0 & 0 & 0 & 0 & -\frac{\partial}{\partial z} & 0 \\ 0 & 0 & 0 & \frac{\partial}{\partial z} & 0 & -iK_x \\ 0 & 0 & 0 & 0 & iK_x & 0 \\ 0 & \frac{\partial}{\partial z} & 0 & 0 & 0 & 0 \\ -\frac{\partial}{\partial z} & 0 & iK_x & 0 & 0 & 0 \\ 0 & -iK_x & 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \\ H_x \\ H_y \\ H_z \end{bmatrix} = -\frac{i\omega}{c} \mathbf{M} \begin{bmatrix} E_x \\ E_y \\ E_z \\ H_x \\ H_y \\ H_z \end{bmatrix}$$

with

$$M = \begin{bmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} & \rho_{xx} & \rho_{xy} & \rho_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} & \rho_{yx} & \rho_{yy} & \rho_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} & \rho_{zx} & \rho_{zy} & \rho_{zz} \\ \rho'_{xx} & \rho'_{xy} & \rho'_{xz} & \mu_{xx} & \mu_{xy} & \mu_{xz} \\ \rho'_{yx} & \rho'_{yy} & \rho'_{yz} & \mu_{yx} & \mu_{yy} & \mu_{yz} \\ \rho'_{zx} & \rho'_{zy} & \rho'_{zz} & \mu_{zx} & \mu_{zy} & \mu_{zz} \end{bmatrix}$$

The matrix M can describe effects in the ϵ and μ tensors, together with optical activity introduced by the ρ and ρ' tensors. Throughout this work, only modulations in the ϵ will be considered, whereas all contributions introduced by the remaining tensors are neglected and taken as null.

The normal components in z can be expressed as a function of invariant x and y components. Some quite complex algebra [23] allows us to take out the third and sixth rows:

$$\frac{ic}{\omega} \frac{\partial}{\partial z} \begin{bmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & -1 & 0 \\ 0 & -1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ H_x \\ H_y \end{bmatrix} = \mathbf{S} \begin{bmatrix} E_x \\ E_y \\ H_x \\ H_y \end{bmatrix}$$

Since the matrix on the left is its own inverse, multiplying both sides by that matrix eliminates it from the left. The entries of matrix $\mathbf{S}$ are of tedious calculation, and are shown in Appendix A. Rearranging the matrices to a 4×4 matrix ordinary differential equation:

$$\frac{\partial}{\partial z} \begin{bmatrix} E_x \\ H_y \\ E_y \\ -H_x \end{bmatrix} = \frac{i\omega}{c} \begin{bmatrix} S_{41} & S_{44} & S_{42} & -S_{43} \\ S_{11} & S_{14} & S_{12} & -S_{13} \\ -S_{31} & -S_{34} & -S_{32} & S_{33} \\ S_{21} & S_{24} & S_{22} & -S_{23} \end{bmatrix} \begin{bmatrix} E_x \\ H_y \\ E_y \\ -H_x \end{bmatrix}$$

that can be simplified to

$$\frac{\partial \psi(z)}{\partial z} = \frac{i\omega}{c} \Delta(z) \psi(z) \quad (2.1)$$

where

$$\psi = \begin{bmatrix} E_x \\ H_y \\ E_y \\ -H_x \end{bmatrix}$$

describes the field at position z . The Δ entries are dependent on K_x , ϵ , μ , ρ and ρ' , and relate to $\mathbf{S}$, as shown in Appendix A.

Considering now an arbitrary homogeneous layer bounded between z_i and z_{i+1} , with Δ_i constant along layer thickness. According to 2.1,

$$\psi(z_{i+1}) = \exp(ik_0 h_i \Delta_i) \psi(z_i) = R_i \psi(z_i) \quad (2.2)$$

where R_i describes the propagation from z_i to z_{i+1} . The propagation of the plane wave through a layer can be calculated by decomposing the wave inside a layer in four partial waves. These four partial waves can involve forward and backward propagating waves in different polarization states.

Lets define a matrix $\mathbf{P}$ as a matrix containing the eigenvectors p_j ($j = 0, 1, 2, 3$) of Δ as

$$\mathbf{P} = [p_0 \ p_1 \ p_2 \ p_3]$$

where

$$\mathbf{p}_j = \begin{bmatrix} E_{j,x} \\ H_{j,y} \\ E_{j,y} \\ -H_{j,x} \end{bmatrix}$$

represents the partial wave field components in the homogeneous layer.

The diagonal matrix $\mathbf{Q}$ depends on the corresponding eigenvalues q_j of Δ as

$$\mathbf{Q} = \begin{bmatrix} e^{ik_0 h q_0} & 0 & 0 & 0 \\ 0 & e^{ik_0 h q_1} & 0 & 0 \\ 0 & 0 & e^{ik_0 h q_2} & 0 \\ 0 & 0 & 0 & e^{ik_0 h q_3} \end{bmatrix}$$

$\mathbf{Q}$ matrix thus describes the propagation of partial waves in a layer, where q_j represents the z component of the corresponding wave vectors $K_{j,z}$.

By application of 2.2 for a layer i , it is possible to relate the tangential components of the electric and magnetic field at the interface z_{i+1} to those at z_i . The continuity of these tangential components at each interface of a stack of N layers leads to the generalization of $\mathbf{R}$ as

$$\mathbf{R} = \prod_{i=0}^{N-1} R_i = \prod_{i=0}^{N-1} P_i Q_i P_i^{-1} \quad (2.3)$$

so that

$$\psi_N = \mathbf{R} \psi_0 \quad (2.4)$$

The electromagnetic field can be expressed in terms of s and p polarization of the incident and exiting light. The backward and forward propagating s and p components of the electric and magnetic electromagnetic fields are related to the tangential components of the fields by

$$\begin{bmatrix} E_x \\ H_y \\ E_y \\ -H_x \end{bmatrix} = \mathbf{L} \begin{bmatrix} E_{p,\rightarrow} \\ E_{s,\rightarrow} \\ E_{p,\leftarrow} \\ E_{s,\leftarrow} \end{bmatrix}$$

with

$$\mathbf{L} = \begin{bmatrix} \cos \theta & 0 & \cos \theta & 0 \\ n & 0 & -n & 0 \\ 0 & 1 & 0 & 1 \\ 0 & n \cos \theta & 0 & -n \cos \theta \end{bmatrix}$$

in which n is either n_{in} or n_{out} and θ is either θ_{in} or θ_{out} , referring to entry and exit media, respectively. With both $\mathbf{L}_{in}$ and $\mathbf{L}_{out}$, it is now possible to define a transfer matrix $\mathbf{T}$ of the multilayer stack relating ingoing and outgoing waves at z_0 and z_N as

$$\begin{bmatrix} E_{p,\rightarrow}(z_N) \\ E_{s,\rightarrow}(z_N) \\ E_{p,\leftarrow}(z_N) \\ E_{s,\leftarrow}(z_N) \end{bmatrix} = \mathbf{T} \begin{bmatrix} E_{p,\rightarrow}(z_0) \\ E_{s,\rightarrow}(z_0) \\ E_{p,\leftarrow}(z_0) \\ E_{s,\leftarrow}(z_0) \end{bmatrix}$$

with

$$\mathbf{T} = \mathbf{L}_{out}^{-1} \mathbf{R} \mathbf{L}_{in} \quad (2.5)$$

The reflection and transmission coefficients, r_{kj} and t_{kj} , respectively can be calculated by

$$(2.6) \quad \begin{aligned} r_{pp} &= \frac{T_{30}T_{23} - T_{20}T_{33}}{T_{22}T_{33} - T_{32}T_{23}} & t_{pp} &= T_{00} + T_{02}r_{pp} + T_{13}r_{sp} \\ r_{ps} &= \frac{T_{31}T_{23} - T_{21}T_{33}}{T_{22}T_{33} - T_{32}T_{23}} & t_{ps} &= T_{01} + T_{02}r_{ps} + T_{03}r_{ss} \\ r_{sp} &= \frac{T_{20}T_{32} - T_{30}T_{22}}{T_{22}T_{33} - T_{32}T_{23}} & t_{sp} &= T_{10} + T_{12}r_{pp} + T_{03}r_{sp} \\ r_{ss} &= \frac{T_{21}T_{32} - T_{31}T_{22}}{T_{22}T_{33} - T_{32}T_{23}} & t_{ss} &= T_{11} + T_{12}r_{ps} + T_{13}r_{ss} \end{aligned} \quad (2.7)$$

where j indicates the polarization of the incident wave and k indicates the polarization of the reflected or transmitted wave, respectively. For stacks containing anisotropic layers, the cross-terms r_{ps} , r_{sp} , t_{ps} and t_{sp} are usually non-zero. From these coefficients, the reflectance or transmittance of a given multilayer stack can be simply calculated by

$$R_{kj} = |r_{kj}|^2, \quad T_{kj} = |t_{kj}|^2 \frac{\cos \theta_{in}}{\cos \theta_{out}} \quad (2.8)$$

being θ_{in} and θ_{out} the angle of incidence and exit of light in the first and last layer, respectively.

In the implemented numerical routine, these rather complex calculations are carried out for a multilayer stack at any arbitrary wavelength. Calculating R or T for a defined array of wavelengths, it is possible to obtain the respective reflectance or transmittance simulated spectra of a given structure.

2.1.2 Optimization of Simulation Parameters

The use of a dielectric tensor in the numerical implementation of the transfer matrix method enables the simulation of light propagation in stacks of both isotropic and anisotropic media. The simulation of magneto-optical effects in structures carrying ESWs containing ferromagnetic, plasmonic metals and dielectrics is one of the possible applications: the diagonal permittivity tensor of the dielectric and plasmonic metal layers can be stacked together with the non-diagonal permittivity tensor of the ferromagnetic layer to return the reflectance spectra of a given structure under the effect of different layer magnetizations.

To form a spectrum, a permittivity tensor is generated for each desired wavelength, and spectral permittivity data (diagonal and off-diagonal) for a given material is extracted from literature and interpolated to fit the desired spectral range and wavelength step. The MOKE configuration can be chosen by the position of the off-diagonal elements in the dielectric tensor. As only the TMOKE configuration was used in this work, the tensor was set as specified in [1.2.2](#).

A. Planar substrate

The simulation of the magneto-optical response of a planar thin film stack will be as good as the more measures of experimental limitations it accounts for. The light collection and injection through optical fibers, even with the use of collimators, will lead to slight divergence of the beam due to the width of the fiber tip, that is not a point-like source. This deviation propagates through the optical circuit up to the prism, where light is incident at slightly different angles. These rays of different incidence angles will trigger ESWs of slightly different peak wavelength, that in practice would result in band broadening. To account for this effect, the broadening of the spot size has been previously measured and determined to be approximately 2° [\[25\]](#). This way, the average spectrum of several spectra simulated for light rays ranging from 73° to 75° is taken.

B. Multimode fiber

The excitation of ESWs is also possible in optical fibers, whose core have a high enough refractive index to enhance light's wave vector to wave vector matching values. The same simulation routine can be applied if light propagation follows the paraxial approximation, where light rays can be thought of as propagating in a straight line. This is the case for the

used optical fibers, with a fiber core of $600\mu m$. In this case, light is incident inside the fiber in angles varying from the critical angle, θ_c , up to 90° . Additionally, each ray can reflect off the fiber–film interface several times. For a film deposited over a length L of a fiber with a core diameter D , each light ray reflects $N(\theta) = L/(D \tan \theta)$ times on the interface.

Each mode, propagating for a given θ , does not carry an equal share of optical power. For this work, an angular distribution of gaussian profile was assumed, being maximum when parallel to the fiber ($\pi/2$) and with a standard deviation of around 6° [26]. All these specifications of axial geometry are taken in account in the simulation work, allowing accurate simulation of surface wave spectra in multimode optical fibers.

2.2 Thin Film Fabrication

The simulation of ESWs in planar or MM fiber matrix allows narrowing the study of all possible configurations of number of layers, materials and thicknesses down to a diminished set of optimized ones. This dissection allows a more incisive study of particular configurations and saves time on the fabrication process. However, the fidelity of the magneto-optical properties of fabricated samples to the simulated counterparts is reliant on thoroughly controlled and calibrated processes.

2.2.1 RF Magnetron Sputtering

All the thin film structures fabricated throughout this work were deposited by radio frequency modulated magnetron assisted sputtering deposition (RF magnetron sputtering).

Sputtering deposition is among the most widely applied thin film fabrication techniques, either for industrial or research and development applications. The process of sputtering deposition consists on the kinetic ejection of atoms out of a target material and their subsequent deposition and nucleation into a thin film on a substrate [27]. The kinetic energy required for the removal of the atoms from the target material is supplied by the impact of accelerated ions of heavy inert gases, such as argon or krypton.

In the case of RF sputtering deposition, the gas atoms are ionized and accelerated by feeding a high RF voltage to a low pressure atmosphere of the working gas, sustaining a plasma. The RF modulation of the power source requires plasma impedance matching circuitry that allows for maximum energy transfer while also enabling efficient sputtering

of insulating materials, since the alternating polarity avoids charge build-up on the target surface that could compromise sputtering efficiency.

The use of a magnetron source in RF magnetron sputtering makes use of the strong magnetic fields from a permanent magnet to confine secondary electron near the target surface in a helical path, extending their residence time in the plasma, thus increasing ionization rates of working gas and deposition rates [27].

A. RF magnetron sputtering setup

The deposition takes place inside a stainless steel sputtering chamber at low pressures. The low pressures are ensured by a vacuum system composed of two primary rotary vane pumps and a secondary turbomolecular pump. The sputtering chamber is firstly evacuated down to $\approx 2.5 \times 10^{-2}$ mbar with a primary pump (Alcatel Adixen 2010 C2), while another (Edwards RV12) evacuates the chamber of the turbomolecular pump, whose functioning requires a starting rough vacuum of $\approx 1 \times 10^{-2}$ mbar. These two chambers are initially isolated by a gate valve (MDC EGV-6000M). Once the latter reference pressure is reached, the turbomolecular pump (Alcatel 5400) is started on its controller (Alcatel CFF 450 Turbo) and accelerates up to the working speed 27000 rpm. When working speed is reached, the gate valve is slowly and carefully opened* and the sputtering chamber pressure starts to decrease rapidly due to the action of the turbomolecular pump, that draws particles from the sputtering chamber and through its chamber to then be evacuated by the primary pump. A Pirani pressure gauge (Adixen ACC 2009) is set on the way out between the turbomolecular pump and the primary rotary pump, and a reference maximum pressure of 1×10^{-1} mbar is assigned when opening the gate valve.

A Pirani-Penning pressure gauge is set inside the sputtering chamber. This gauge consists of two sensing gauges — Pirani and Penning type — that automatically switch to one another for higher or lower pressures, respectively, in the range from $\approx 1 \times 10^2$ mbar down to $\approx 1 \times 10^{-6}$ mbar, and will be used to control the plasma pressure during the deposition process.

*The turbomolecular pump consists of multiple layers of rotating tilted blades that transfer momentum to the gas molecules upon collision, directing them out of the chamber, reducing the pressure on the way in and increasing it on the way out. If the pressure difference between the top and bottom of the pump rises uncontrollably, the blades may bend, increasing the risk of violent blade crash. To avoid this catastrophic scenario, the gate valve must be opened very carefully.

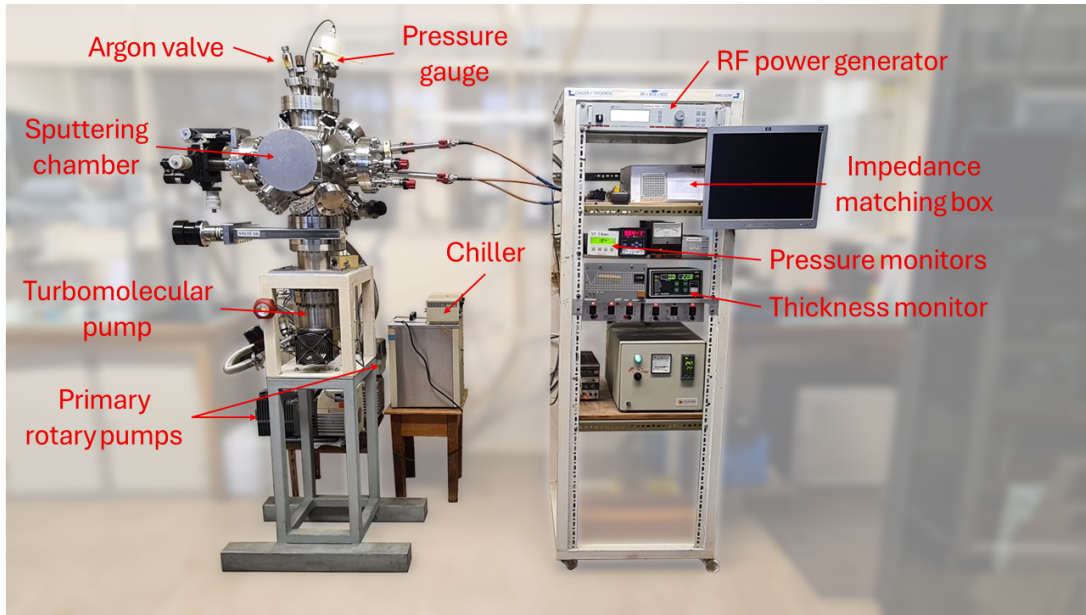


FIGURE 2.2: Overview of the RF magnetron sputtering setup at INESC TEC facilities.

In all depositions, argon gas was used to form the plasma. The argon gas is injected in the sputtering chamber through a needle valve that allows precise control of the pressure inside the chamber, and is connected to an external argon gas cylinder (99.99% pure).

A cooling system supported by an external water chiller (Endocal RTE-110) set to 10°C is used to cool the sputtering targets, that heat up significantly for high power depositions and due to plasma etching. Additionally, keeping the targets at a stable temperature helps keeping the sputtering conditions under control.

The RF power generator (Advanced Energy CESAR 136) allows for powers up to 600W to be fed into the sputtering guns. The power is forwarded to the plasma through an impedance matching network (Advanced Energy VM600A), connected between the RF power generator and the sputtering guns, that adjusts its internal impedance to match that of the generator and the plasma to the industry standard pure load of 50Ω [28]. The matching network adjusts automatically for power changes in the generator or pressure and temperature variations in the plasma, ensuring that at all times, all the power is transmitted from the generator to the plasma, and none is reflected back.

The connection of the matching network to the sputtering guns is made through a selector, that allows to switch between sputtering guns, and therefore the target material to be deposited. Although the sputtering chamber can bear three sputtering guns (Ferrotec MEIVAC MAK 2.0"), only two were functional (arbitrarily named M1 and M2), so that only two targets could be used in a single vacuum cycle.

During deposition, the film thickness is controlled by a quartz crystal microbalance (QCM). This device tracks resonant frequency changes of a quartz crystal according to the mass of deposited material over a known exposed area. Providing the density and acoustic impedance of the depositing material, the QCM controller (Edwards FTM7) returns the real-time thickness of the depositing film. The resonant frequency is also dependent on the temperature, meaning that a variation of its temperature would induce errors in the thickness reading. To prevent such errors, the QCM is cooled by the chiller, keeping it at a steady temperature.

An overview of the RF magnetron sputtering deposition setup used throughout this work at INESC TEC facilities is pictured in [2.2](#).

B. Planar substrate deposition calibration

The sputtering chamber includes supports for planar substrates and optical fibers. The planar substrate holder can rotate in the axis normal to the film surface by action of an external motor driven by a voltage source. Given the relative position of the sputtering targets to the substrate and the angular profile of sputtering deposition, a still deposition would result in non uniform film thicknesses along the substrate. The rotation of the substrate holder allows for the averaging of film thicknesses across substrate area.

The relative position of the QCM to the sputtering guns is also a source of error. Although the sputtering guns are symmetrically placed around the substrate, the same does not happen for the QCM, that is skewed to a corner of the substrate holder. This disposition results in unreliable thicknesses of the deposited film that do not match those read on the QCM controller. To overcome this problem, a correction factor needs to be accounted for. This correction factor was determined by depositing films of a certain thickness as read by the QCM monitor, and then comparing it to the actual thickness as measured from SEM (FEI Quanta 400 FEG ESEM) images. The ratio between these two thicknesses is called the tooling factor and should be exclusive to each sputtering gun, regardless of target material. However, the registered low depositions rate of Fe films – even at high power – led to a suspicion that the magnetic field created by the magnetron could be deflecting material away from the QCM, compromising the calibration. To account for this possible effect, one of the sputtering targets was calibrated for Fe deposition.

In this process, commercial high purity targets of Ag, TiO_2 and Fe were used for film deposition. Accounting for the SEM resolution, a bilayer film with 50nm Fe (M1) and

50nm TiO_2 (M2), and a multilayered film of alternating 100nm Ag (M1) and 50nm TiO_2 (M2) layers were deposited. The alternating metallic and dielectric layers allow for a better contrast, while the relatively large thicknesses of the films help reduce tooling factor error. The substrates with the deposited film were then carefully cleaved with a diamond tip glass cutter to expose a transversal section of the film stack before SEM inspection.

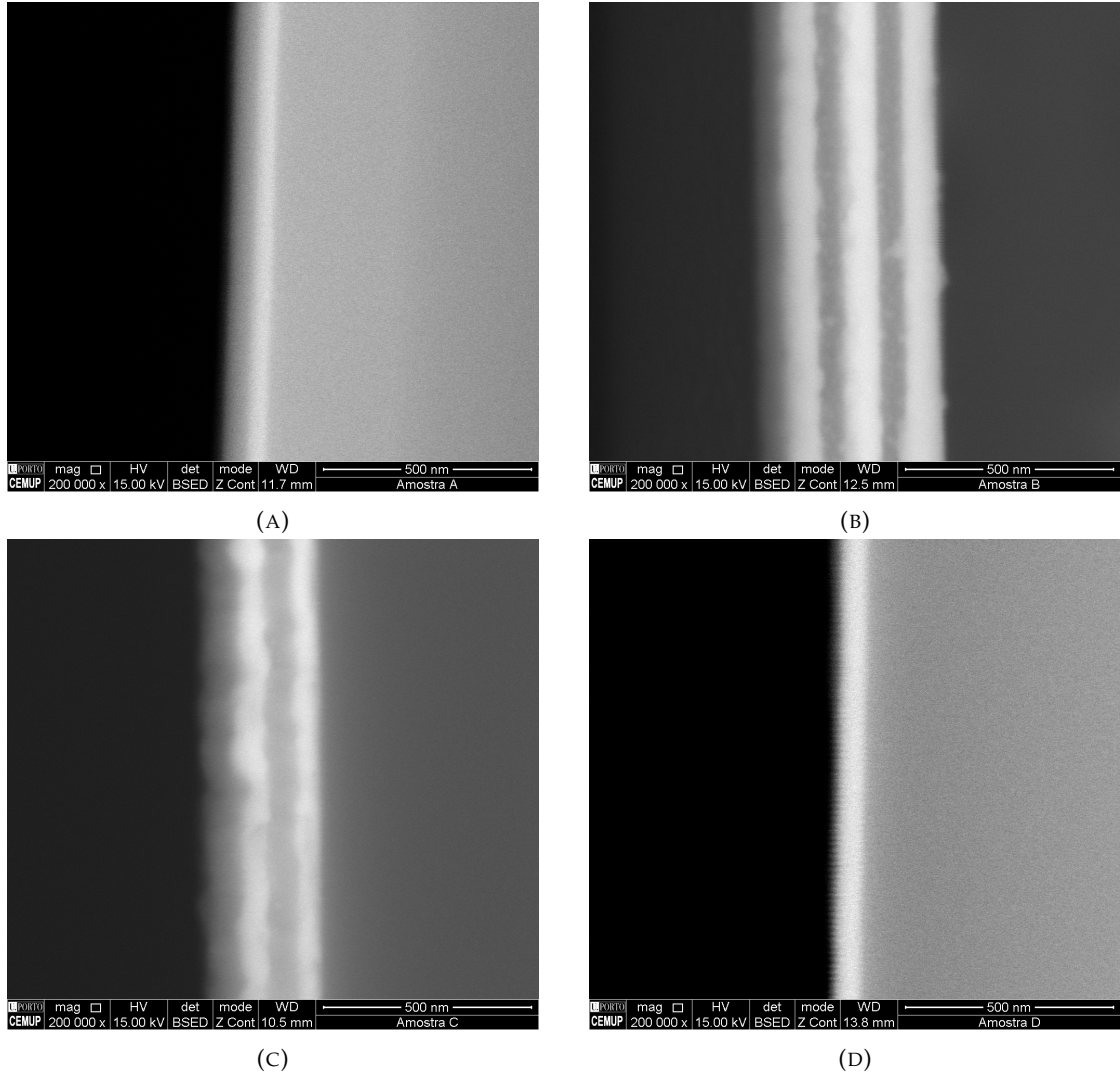


FIGURE 2.3: SEM images of planar structures: (A) 50nm Fe/50nm TiO_2 and (B) 3×100 nm Ag/50nm TiO_2 ; Optical fibers: (C) 2×200 nm Ag/150nm TiO_2 and (D) 130nm Fe. All images taken at $\times 200.000$ magnification.

The SEM images were analyzed for layer thickness with ImageJ software. Many images were recorded at different regions of the exposed film, and multiple measurements were made in each image, according to the scale provided by the SEM. The SEM images for the planar substrate depositions are pictured in 2.3 (A) and (B), and the thicknesses for all measured layers are plotted in 2.4.

The Fe and Ag layers were deposited using the same sputtering gun (M1), closer to the QCM than the substrate, resulting in films actually thinner than the monitor value. The opposite occurs for the TiO_2 layers, that were deposited with the other sputtering gun (M2), closer to the substrate than to the QCM. The average measured thickness was calculated for the three deposited materials, and the tooling factors $t = t_{QCM}/t_{SEM}$ were determined and are shown in table 2.1.

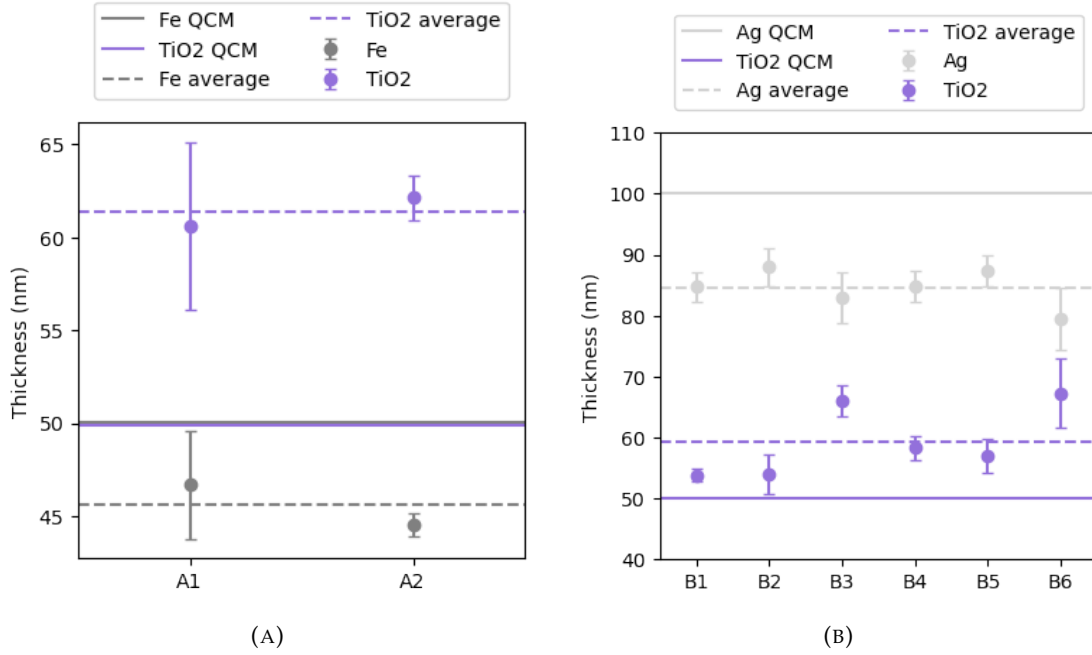


FIGURE 2.4: SEM thickness measurements for planar substrates: (A) 50nm Fe/50nm TiO_2 and (B) 2x100nm Ag/50nm TiO_2 .

C. Multimode fiber deposition calibration

The optical fiber holder is designed for concentric deposition around an exposed segment of the optical fiber core or cladding. It consists of a rotating SMA905 connector (driven by a motor powered by an external another voltage source) that sits parallel to the target's surface, around 5cm away. The central region of exposed optical fiber was set to an equidistant position to both the sputtering guns, regardless of the fiber length or exposed section length. Although this setup grants uniform axial deposition at each point around the optical fiber, it does not grant uniform deposition along the exposed segment in the fiber axis, given that different points on the exposed segment will be at different distances to a particular sputtering gun. The uneven deposition will cause band broadening and is

therefore undesirable. Keeping the exposed segment short will help limit the uneven deposition effects, minimizing band broadening. However, a short segment leads to fewer electromagnetic modes within the fiber interacting with the sensitive section, limiting the FWHM of ESW bands. A compromise on exposed section length was settled to around 1cm.

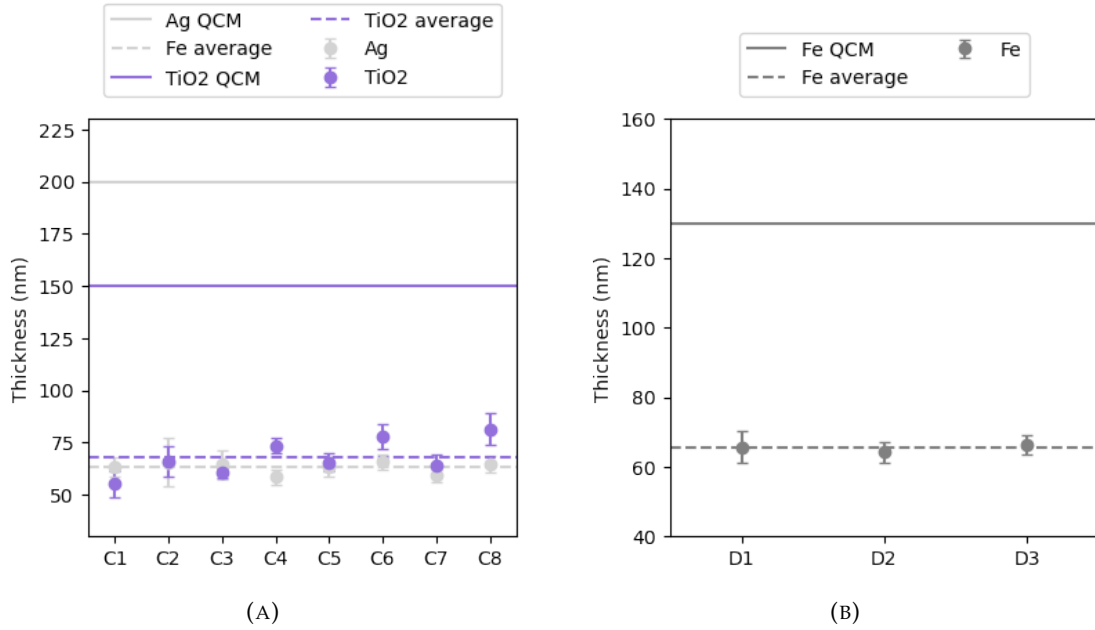


FIGURE 2.5: SEM thickness measurements for optical fibers: (A) 200nm Ag/150nm TiO_2 and (B) 130nm Fe.

The calibration process for optical fiber deposition was analogous to that of planar substrates, holding only slight differences in the fabrication process, including layer thicknesses. Two samples were fabricated, one with two unit cells of 200nm Ag (M1) and 150nm TiO_2 (M2), and the other made up of a single 130nm layer of Fe (M1). The SEM images are shown in 2.3 (C) and (D). The SEM measurements are plotted in 2.5 and the respective tooling factors for M1 and M2 sputtering guns for optical fiber deposition are also shown in table 2.1.

For both the case of planar substrate and optical fiber deposition, it is noticeable that the tooling factors are higher for M1 than M2 sputtering gun, fact that was expected given the closer relative position of the QCM to M1 sputtering gun. The overall higher tooling factors for optical fibers is also expected, since the axial fiber rotation in the plane parallel to the target surface means that only a fraction of the fiber is in the line of sight of depositing material at all times.

TABLE 2.1: Tooling factors of M1 and M2 sputtering guns.

Sputtering Gun	Planar substrate	Optical Fiber
M1	1.18 ± 0.06	3.16 ± 0.33
M1 (Fe)	1.09 ± 0.06	1.99 ± 0.11
M2	0.83 ± 0.03	2.23 ± 0.32

D. Thin film sample deposition

All the planar substrate samples were deposited on glass substrates cut from microscope slides (Labbox SLIU-005-050) to $2\text{cm} \times 2\text{cm}$ dimensions. Being of amorphous structure and different composition than crystalline SiO_2 , a slight mismatch of refractive index at the prism-substrate interface is expected. However, such mismatch has not been found to compromise accuracy of ESWs bands significantly.

The substrates were cleaned in the clean room environment of the MNTEC unit of CEMUP following an established protocol. Each substrate was cleaned for five minutes in a ultrasound bath of acetone, isopropanol, ethanol and deionized water, before being blow dried with a N_2 gun. The organic solvent ultrasound bath allows cleaning of adsorbed organic residues that would be harder otherwise, while the final deionized water bath eases an efficient removal of residue with the N_2 gun due to the high surface tension of water.

The optical fiber segments were cut from commercially available ThorLabs FT600EMT multimode optical fiber. Around 3mm of coating at each end were removed and polished with $3\mu\text{m}$ grain silicon carbide and $1\mu\text{m}$ grain aluminum oxide sandpaper disks. The sensitive area to be deposited had its covering cladding and protective coating layers mechanically removed with a sharp razor blade to a desired extent. The polymeric nature of the cladding and protective coating allows for its removal without damaging the SiO_2 core. Both the fiber tips and the exposed section were carefully cleaned with ethanol before being placed inside the sputtering chamber for deposition.

All depositions were carried out under a argon plasma pressure of $\approx 7 \times 10^{-3}$ mbar. The sputtering power delivered to the sputtering guns varies according to the material. Dielectric materials generally need higher sputtering powers than metals to achieve a practical deposition rate. Amongst all the available materials, the Ag, Fe, TiO_2 and SiO_2 targets were the most resourced, with deposition rates of around 0.11nm/s , 0.02nm/s , 0.04nm/s and 0.04nm/s for powers of 13W, 110W, 110W and 110W, respectively.

2.3 Thin Film Characterization

Throughout this work, some characterization techniques were employed to unveil the properties of the fabricated samples. This information is of great importance in order to optimize and understand the structure's functional response. Samples of different properties had their magnetic response profiled by a superconducting quantum interference device (SQUID) and ultimately their magneto-optical properties characterized by spectral interrogation in a modified Kretschmann configuration setup, where the effects of magnetization and the sensitivity to external refractive indices in ESWs are quantified.

2.3.1 Optical Characterization - Kretschmann Configuration Setup

The magneto-optical properties of the fabricated structures undergo spectral characterization in a modified Kretschmann configuration. As detailed in 1.3, the excitation of surface waves requires a matching wave vector condition possible by the use of a high-index prism to couple light into the fabricated films through ATR. One of the possible setup configurations for ESWs is the Kretschmann configuration. This configuration eases the optical interrogation of ESWs and the manipulation of external refractive indices. This section details the components and the assembly of a Kretschmann configuration setup for planar substrates and optical fibers, and the adaptations needed for applying controlled magnetic fields.

A. Planar substrates

A schematic representation of the modified Kretschmann configuration setup for planar substrates is shown in Figure 2.6.

The light is emitted from a broadband VIS-NIR tungsten-halogen light source (Sarspec LS-W) and coupled to a MM optical fiber patch cord (ThorLabs M29L). The light beam exiting the optical fiber is collimated by a collimating lens (Avantes COL-UV/VIS) before entering the prism. A broadband polarizer (Thorlabs LPNIR050-MP2) is inserted before the prism, allowing the manipulation of the polarization of the light interacting with the films. The light proceeds to the fused silica prism (Edmund Optics TECHSPEC UV Fused Silica Right Angle Prisms, $25 \times 25\text{mm}$), reflecting off the thin film structure on top. The thin films are deposited on glass substrates that are placed on the top surface of the prism. Due to close proximity of the refractive indices, the glass substrates behave as

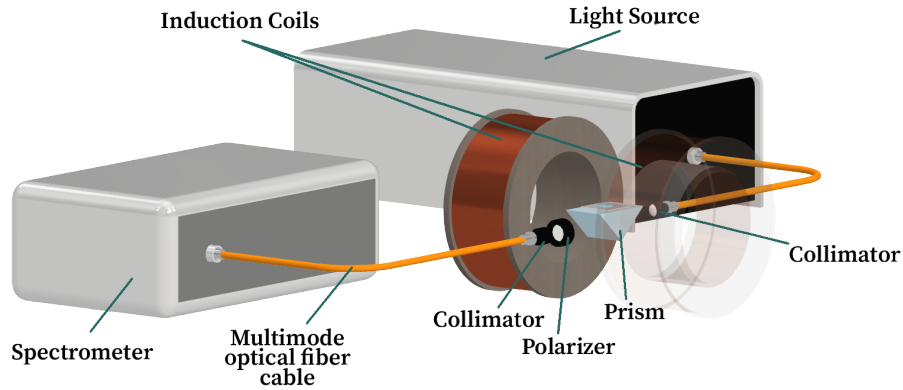


FIGURE 2.6: Illustration of the modified experimental Kretschmann configuration setup. Notice the induction coils for transverse magnetic field incidence.

an extension of the prism. An index-matching oil is applied between the prism and the substrate, ensuring close contact between the surfaces, mitigating reflection losses linked to the SiO_2 –air–glass interface. The light is collected back into an optical fiber patch cord by another collimator, and proceeds to a spectrometer. Two spectrometers of complementary wavelength ranges were used: Avantes AvaSpec-ULS2048CL-EVO, spanning from 190 to 1100nm, and OceanOptics NIRQuest, from 900 to 1700nm. The two spectrometers could be used simultaneously using a bifurcated fiber bundle (ThorLabs BFY600HS02), that collects light from a single source off the prism and splits the optical beam into two optical fiber ends, as depicted in Figure 2.7 along with the optical characterization setup.

A central block was 3D printed to fit the prism, the polarizer and the collimators in a single, compact piece. An optical fiber connector was placed in each side of the piece, to couple the incoming and outgoing light. A through in the middle allows for the prism to sit vertically. A teflon piece was also designed to tightly fit the top of the prism for manual application of different analytes over the film. This piece can be bolted to the main block, pressing against the film with an O-ring, ensuring that the analyte does not leak outside the central region of the film. The polarizer was also embedded in a 3D printed handle, allowing for easy rotation of polarization between s and p state at any time.

An induction coil is placed at each side of the prism, being both connected so to induce magnetic field lines parallel to the film surface and perpendicular to the plane of light incidence. The two coils were designed and fabricated in order to achieve saturation

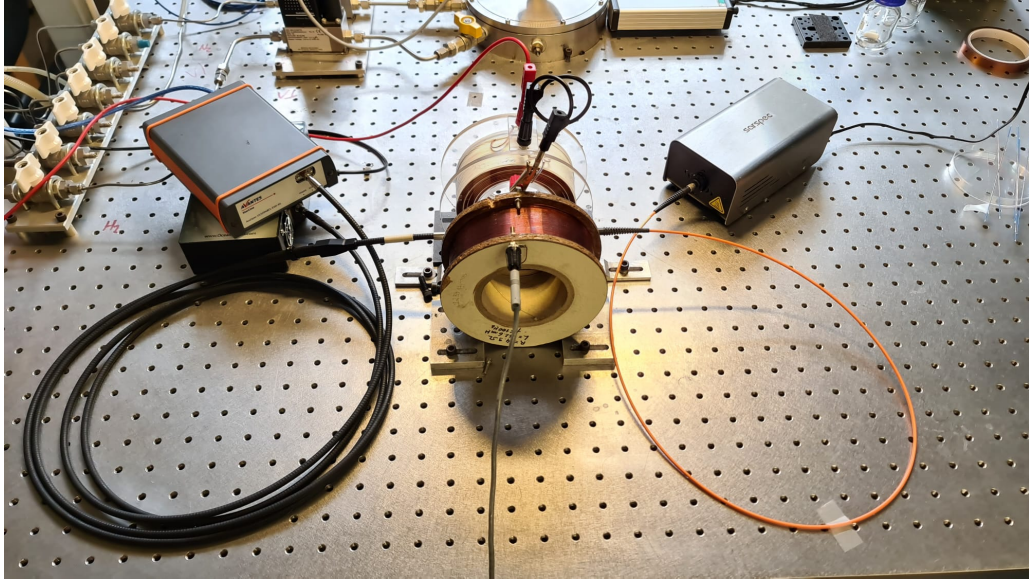


FIGURE 2.7: Experimental Kretschmann configuration setup for optical characterization of planar thin films. Notice de optical fiber bundle for simultaneous characterization of VIS-NIR spectra.

magnetization in Fe thin films, and its dimensions (average radius, winding number, separation distance) were determined to grant a close to uniform magnetic field in the region between them (Helmholtz configuration). The two coils were connected in series and to a DC voltage source (Univolt DT305AD), and a multimeter (Hewlett Packard 34401A) was set in series to measure the electric current.

B. Multimode fibers

A similar setup was designed for the characterization of MM optical fibers (Figure 2.8) with deposited films, albeit no coupling prism is needed, since the SiO_2 fiber core assures a high enough RI for ESW excitation. The same broadband light source was used to couple light through an optical fiber patch cord, a collimator, a polarizer and finally into the sensitive optical fiber segment. The optical fiber segment is connected to the other components with a connector through two bare fiber terminators (Thorlabs BFT1 Bare Fiber Terminator with a B10125A Multimode SMA905 Connector). The fiber is carefully inserted through a connector at each end and fixed in place with the magnets in the terminators. The light finally proceeds through another optical fiber patch cord into a spectrometer. A manual translation stage to adjust for fiber segment length and a base with a channel for analyte flow were also 3D printed for optical fiber characterization of magneto-optical properties.

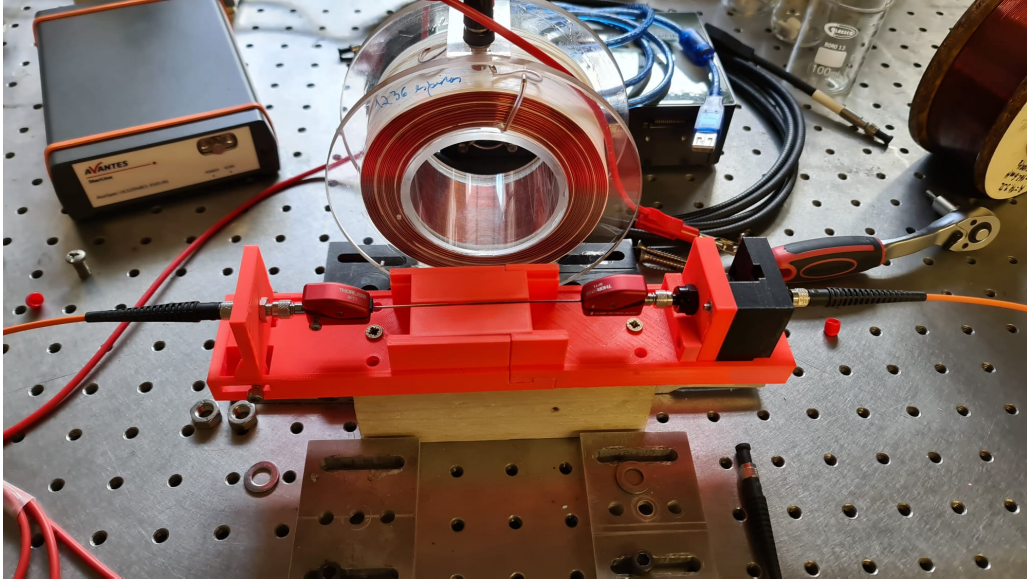


FIGURE 2.8: Experimental setup for optical characterization of thin films in multimode fibers. Notice the deposited fiber segment, in the middle, and the terminators at each end, coupling light in and out of the segment.

C. Magnetic field profile

The insertion of two induction coils allows for the incidence of controlled magnetic fields, something that would be considerably more difficult with permanent magnets. The spatial profile of the field lines is of extreme importance, given that the relative orientation of the magnetic field to the plane of incidence of light defines the kind of magneto-optical effect present in the film. Diverging field lines in the sensitive area should then be avoided, as they would sum undesired contributions of different magneto-optical effects, compromising the analysis of results. For this reason, the use of a single induction coil in front of the setup is not ideal, since the magnetic field lines start diverging near the inside edge of the coil (edge effects). Uniform magnetic field lines can be produced inside a induction coil long enough to neglect these edge effects. However, the insertion of the characterization setup in such induction coil would not be any practical. A more practical approach requires two identical coils of a given radius set apart by a certain distance, so that the magnetic field lines in the region between them are approximately uniform – Helmholtz configuration.

The magnetic field at the axis from a coil can be obtained starting from the Biot-Savart law for a single wire loop.

$$B(x) = \frac{\mu_0 I}{2} \frac{R^2}{(R^2 + x^2)^{3/2}} \quad (2.9)$$

where $\mu_0 = 4\pi \times 10^{-7} \text{ T} \cdot \text{m} / \text{A}$ is the vacuum permeability constant, I is the DC current, R is the loop radius, and x is the axial distance to the loop's center point. For N turns, the produced magnetic field is obtained by simply multiplying 2.9 by the number of turns.

$$B(x) = \frac{\mu_0 N I}{2} \frac{R^2}{(R^2 + x^2)^{3/2}} \quad (2.10)$$

Notice that this equation does not consider the width of the coil, nor the varying radius of built up windings. Being ω the width, D the largest radius, d the smallest radius and r the distance between the two coils, a more exact profile of magnetic field can be obtained by

$$B(x) = \frac{\mu_0 N I}{\omega(D-d)} \left[Q_{pp} \ln K_{pp} + Q_{np} \ln K_{np} + Q_{pn} \ln K_{pn} + Q_{nn} \ln K_{nn} \right] \quad (2.11)$$

where parameters Q_{pp} , Q_{np} , Q_{pn} , Q_{nn} , K_{pp} , K_{np} , K_{pn} and K_{nn} are functions of the setup dimensions, detailed in Appendix D.

Two coils were available with similar radii ($d_1 = 4.45\text{cm}$, $D_1 = 6.7\text{cm}$; $d_2 = 4\text{cm}$, $D_2 = 6\text{cm}$), widths ($\omega_1 = 4\text{cm}$, $\omega_2 = 4.8\text{cm}$) and identical number of turns ($n = 1235$). When connected in series, they showed a resistance of 28.8Ω , limiting the current to 1.86A . This high current led to Joule heating that increases resistance overtime, diminishing the value of maximum current through the circuit, and consequently the induced magnetic field. To avoid magnetic field deviations over time, a maximum operable current of 1.6A was set.

The optimal distance between the center of the two coils that would produce a uniform magnetic field between them was estimated by 2.11 to be between 6 and 8cm . Unfortunately, the dimensions of the support for the Kretschmann configuration setup did not allow the center of the two induction coils to sit apart less than 8cm , so that this distance between the centers of the induction coils was set.

The magnetic field magnitude in the vicinity of the prism surface was later measured using a gaussmeter (LakeShore 421). The gaussmeter probe is based on the Hall effect, making it very sensitive to the direction of the magnetic field, so that any misalignment from normal incidence can significantly impact the sensed value. The probe was secured with a clamp to a translation stage, fixed to the optical table. Measurements were made in steps of 1cm along the axis distancing the two coils, which contains the plane where the thin films sit for characterization. A comparison of the measured magnetic field with the estimated by 2.11 is plotted in Figure 2.9.

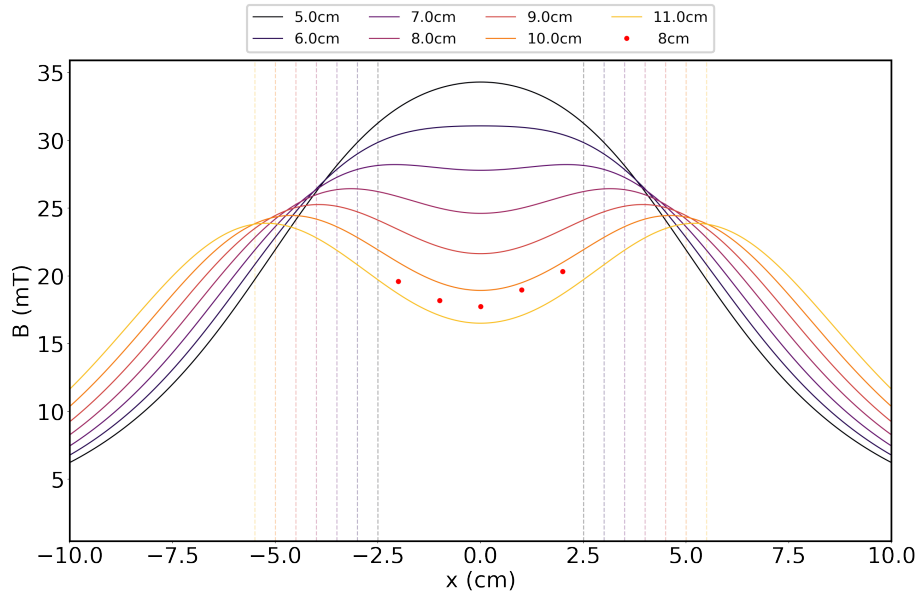


FIGURE 2.9: Simulation and experimental results for the sensed magnetic field in the axis of two induction coils sitting different distances apart. The magnetic field in the central region was measured for a distance of 8cm between the induction coils.

A significant error on the magnitude and profile of the magnetic field was obtained for the estimated and measured magnetic field at a coil separation of 8cm. Instead, the experimental data seems to match, in magnitude and profile, the estimated curves for spacings between 10 and 11cm. Despite the error, the magnetic field profile in the vicinity of the prism is still roughly uniform, assuring dominant unidirectionality of field lines in the region, and therefore the dominant influence of a single, discriminate magneto-optical effect. The magnitude of the maximum obtained field ($\approx 20\text{mT}$) is also high enough to attain saturation magnetization in the fabricated Fe thin films, as confirmed by SQUID measurements.

2.3.2 Materials Characterization

A. Superconducting Quantum Interference Device (SQUID)

A Superconducting QUantum Interference Device, commonly referred by the acronym SQUID, is a very sensitive detector of magnetic flux. A complete description of its principle of operation involves BCS theory of superconductivity, the Josephson effect and Magnetic flux quantization, all quite complex and beyond the scope of this work.

A SQUID consists of two loop halves of superconducting material bound together by two insulating layers, connected in parallel. These insulating layers sandwiched between

superconductors are called Josephson junctions, and the maximum possible current of superconductor Cooper pairs tunneling through these junctions is called the critical current, I_C . By biasing the critical current in each half and applying a magnetic flux through the loop, an additional current I_Φ is induced to the loop, decreasing it in one side, and since it cannot go beyond I_C in the other, an electric potential is developed, creating a voltage across the junctions.

Due to the quantization of magnetic flux in a superconductor loop, as the magnetic flux is increased, the device induces electric currents in each branch to achieve an integer flux quanta. This way, as the magnetic flux increases linearly, an electric current of switching directions is induced, and a periodic measurable voltage is created, being the period that of the flux quantum, $\Phi_0 = h/2e$.

This device can be used as vibrating sample magnetometer (VSM) by periodically driving a sample through the SQUID loop under the influence of a scanning magnetic field. The induced currents create a voltage that can be measured and converted to magnetic moment, m , along the applied magnetic fields. The use of a gradiometer – composed of another, spaced apart SQUID loop – allows for the sample's induced magnetic flux to be isolated from that of the applied external magnetic field. For ferromagnetic materials, an average of the sample's magnetization, M , is easily obtained knowing its volume, V , by

$$M = \frac{m}{V} \quad (2.12)$$

Knowing the magnitude of the magnetization in a given film is helpful in the study of sample properties. A study on the dependence of magnetization on film thickness can help perceive different regimes in the magnetic properties of thin films.

Chapter 3

Simulation and Experimental Results

In this chapter, a complete study towards the enhancement of the magneto-optical response of different structures bearing SPRs and BSWs is carried out. The first section carries a simple characterization of plasmonic and magnetoplasmonic structures for experimental validation of the implemented simulation work for plasmonic and magneto-optical responses. These simple results are followed by a characterization of the magnetic and magneto-optical properties of fabricated films by means of different techniques. By the end, a complete simulation work of multiple configurations of SPR and BSW bearing films is performed, and the optimized structures are fabricated in an attempt to achieve enhanced magneto-optical sensitivity to RI.

3.1 Experimental Validation of Simulation Work

For the experimental validation of the simulation work, both for theoretical inputs and experimental setup constraints, a set of plasmonic films were fabricated. The plasmonic basis of all deposited structures have Ag or Au layers as plasmonic metal, and the dielectric layer of positive permittivity required for SPR excitation consisted of either an external medium, such as air or a water-based solution, or thin film of TiO_2 . The magneto-optical properties of the studied films originated from the deposition of Fe thin layers.

All simulation work was carried out using spectral permittivities found in literature of different sources. To analyze the experimental fidelity of the simulation routine for each material, a set of different plasmonic films were deposited: 50nm Ag, 50nm Au, 40nm Ag/10nm Fe and 50nm Ag/40nm TiO_2 . All films were characterized in the implemented Kretschmann configuration setup with deionized water ($RI = 1.333$) as external medium.

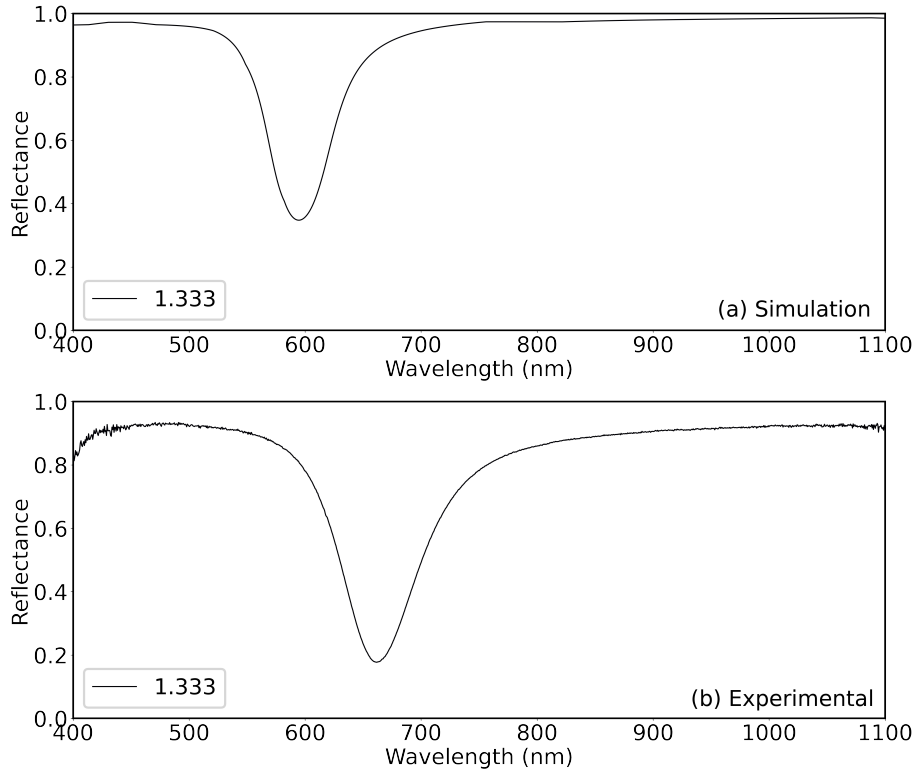


FIGURE 3.1: Reflectance spectra of (a) simulated and (b) experimental films of 50nm Ag for RI=1.333

The reflectance spectra of simulated and experimental films of 50nm of Ag are plotted in Figure 3.1. Although the resonance band is clearly visible for both cases, there is a significant red shift of 67nm from the experimental spectra (661.5nm) relative to the simulated one (594.5nm). The reflectance of both spectra shows a very similar profile, with almost total reflection of light away from the resonance wavelength. At the SPR wavelength, the reflectance drop appears to have a slight mismatch of 17pp. This mismatch can either originate from error in the deposited thickness, as higher thicknesses of Ag lead to higher reflectances, or from slightly different dielectric properties of the deposited films to those extracted from literature [29].

The same film thickness of Au was deposited and characterized for the same external RI of deionized water, and a comparison with the respective simulated spectrum is depicted in Figure 3.2. In contrast with the deposited Ag films, the match between simulated and experimental spectra for the 50nm Au films is almost identical. The experimental minimum of the plasmonic band shows a red shift under 8nm when compared to the simulated spectrum, from 696.7nm to 689.3nm. At resonance wavelength, both spectra differ only by 1.5pp. The fidelity between experimental and simulation spectra for thin

films of Au is, thus, higher than that of Ag.

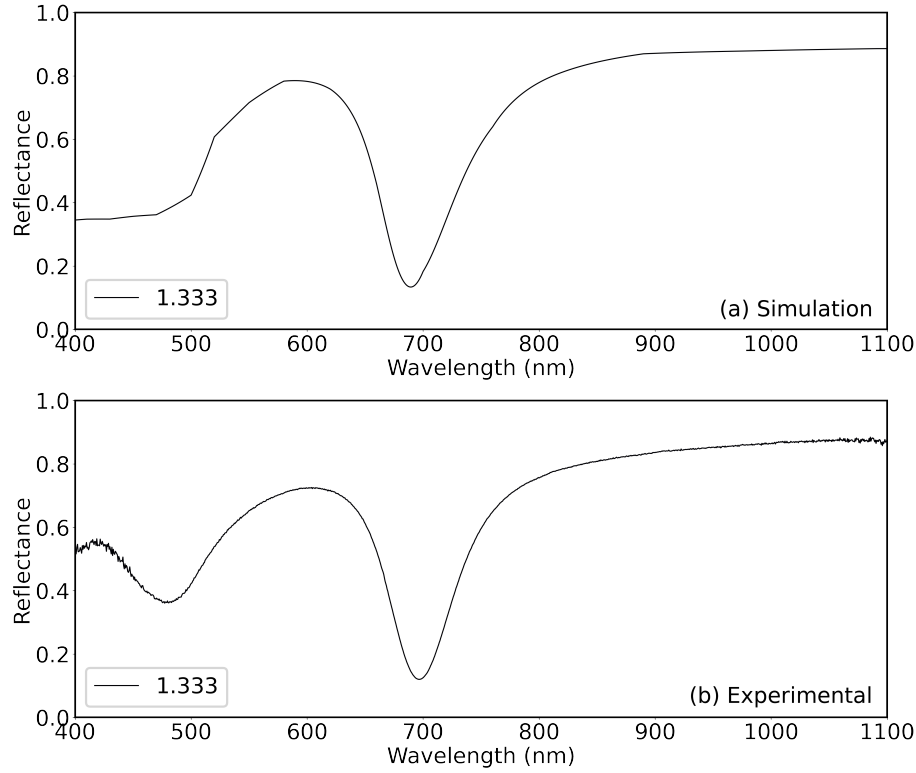


FIGURE 3.2: Reflectance spectra of (a) simulated and (b) experimental films of 50nm Au for RI=1.333

Despite the better match obtained for Au films, much of the work carried out in this chapter was made using Ag thin films, given their higher sensitivity to RI variations and the much higher accessibility of Ag when compared to Au.

Since the rest of the materials resorted to in this work do not show plasmonic behavior, namely TiO_2 that is a dielectric, and Fe that is a metal with too high optical losses, their experimental validation was analyzed in combination with Ag thin films. Figure 3.3 shows the simulation and experimental spectra of a 50nm Ag/40nm TiO_2 bilayer, and a similar observation to that of the 50nm Ag film could be made. There is a significant red shift of 53nm (1280nm to 1333nm) and a small mismatch in the share of light absorbed at SPR wavelength of 3pp.

This mismatch only worsens for a 40nm Ag/10nm Fe bilayer, shown in Figure 3.4. For this configuration, a red shift of resonance wavelength of 78nm and a significant mismatch in peak reflectance of 12pp are obtained. This greater deviance may suggest that both the Ag and Fe layer are contributing to a redshift of the plasmonic band. Additionally, a huge damping of the plasmonic band is notorious, widening the resonance peak considerably

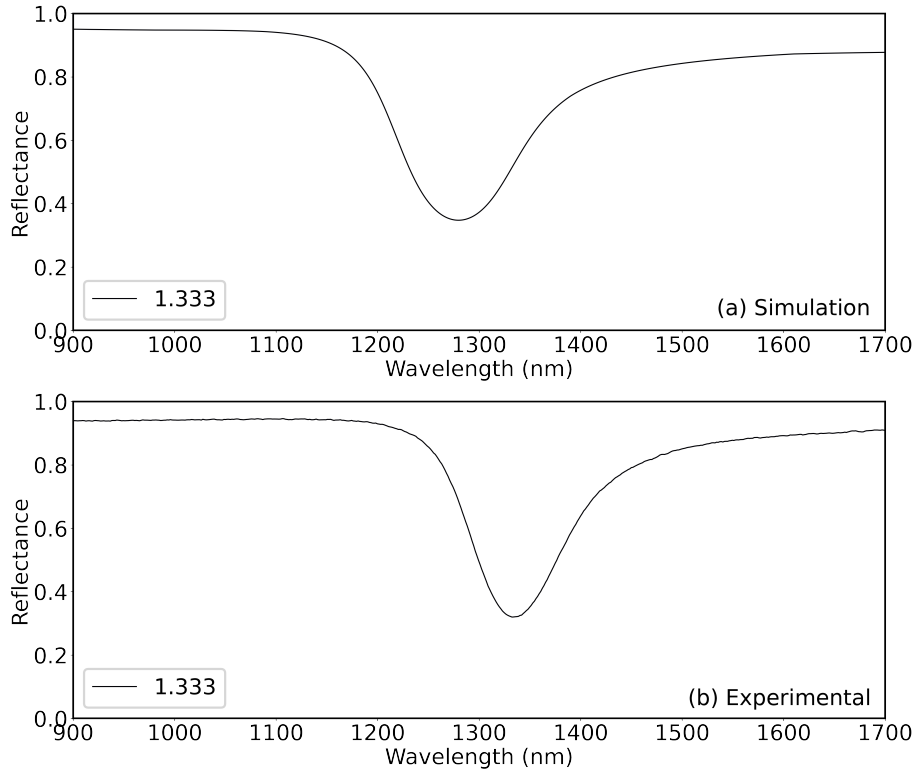


FIGURE 3.3: Reflectance spectra of (a) simulated and (b) experimental films of 50nm Ag/40nm TiO_2 for RI=1.333

when compared to those above shown. This damping is caused by the high optical losses of the Fe layer, and for this reason the thickness of the Ag layer had to be lowered to 40nm Ag, as the contribution of both metals in optical losses would have damped down the SPP peak. This damping also increases the FWHM of the plasmonic band, compromising sensing capacities.

Nonetheless, the excitation of SPPs is clear and the simulations accuracy is overall satisfactory for the study and optimization of plasmonic structures. As mentioned before, the discrepancies found can be related to different dielectric properties of the deposited films to those extracted from literature. The 50nm films of Ag and Au were fabricated by RF magnetron deposition, unlike those referenced in [29] and [30], that were fabricated by thermal evaporation, that tends to lead to less uniform, less compact films with lower grain size, that is not optimal for the conductivity of electrons in plasmonic metals. The enhanced properties of magnetron sputtering deposition may thus explain the lower FWHM of the plasmonics bands. This enhancement is particularly notorious for the 50nm Ag and 40nm Ag/10nm Fe bilayer, although such band definition in the latter may be in part related to Fe oxidation, that leads to iron oxides of lower optical damping.

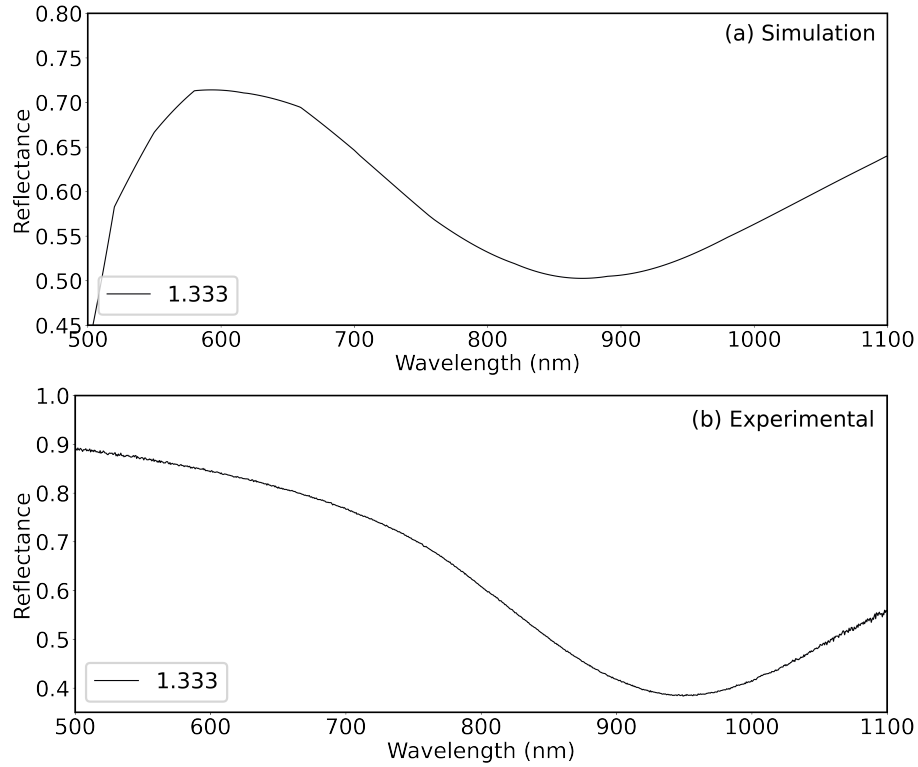


FIGURE 3.4: Reflectance spectra of (a) simulated and (b) experimental films of 40nm Ag/10nm Fe for RI=1.333

Despite the accurate simulation of SPPs in isotropic planar plasmonic structures, the study of magneto-optical properties of layered media requires an accurate description of the influence of magnetic fields or remanent magnetizations in magnetic media. Such work was carried out by adding off-diagonal tensor elements to the dielectric tensor of the simulated structures according to the relative direction of the magnetic field (or magnetization) vector. Since Fe is ferromagnetic and was the only magnetic material used throughout this work, we may, from now on, refer exclusively to the layer magnetization as the source of any magneto-optical modulation.

As seen in 1.1, the off diagonal elements of the dielectric tensor of a magnetic medium show spectral dependence and are a linear function of magnetization for first order effects. Assuming that all the deposited Fe layers share the same constituency and magnetic properties, the magnetization, as an intrinsic physical property, should be independent of Fe layer thickness, and the spectral dependence of the off-diagonal elements is introduced by the complex magneto-optical coefficient, referenced in 1.11. This way, the magnitude of magneto-optical effects is ultimately a function of the spectral magneto-optical coefficients and the thickness of the ferromagnetic layer through which light propagates, while

magnetization remains the same, simply scaling the off-diagonal elements' profile magnitude.

The off-diagonal elements for the dielectric tensor of a Fe layer magnetized transversely to light propagation were extracted from the work of Krinchik and Artem'ev on the magneto-optical properties of Fe, Ni and Co ranging from the ultraviolet to the infrared part of the spectrum [31], and the work of Ferguson and Romagnoli on the TMOKE properties of transition and rare earth alloys [32]. The data considers saturation magnetizations and was inserted into the dielectric tensor together with the complex diagonal permittivities extracted from [30] at equal interpolated wavelengths. Figure 3.5 shows the spectral dependence of the real (ϵ_{xy}^r) and imaginary (ϵ_{xy}^i) parts of the complex off diagonal elements extracted from [31], [32]. Both spectra show a similar profile, although signal magnitudes differ significantly. This difference may be explained by the different nature of the Fe samples, since a bulk sample is used in [31], but an evaporated thin film sample is used in [32].

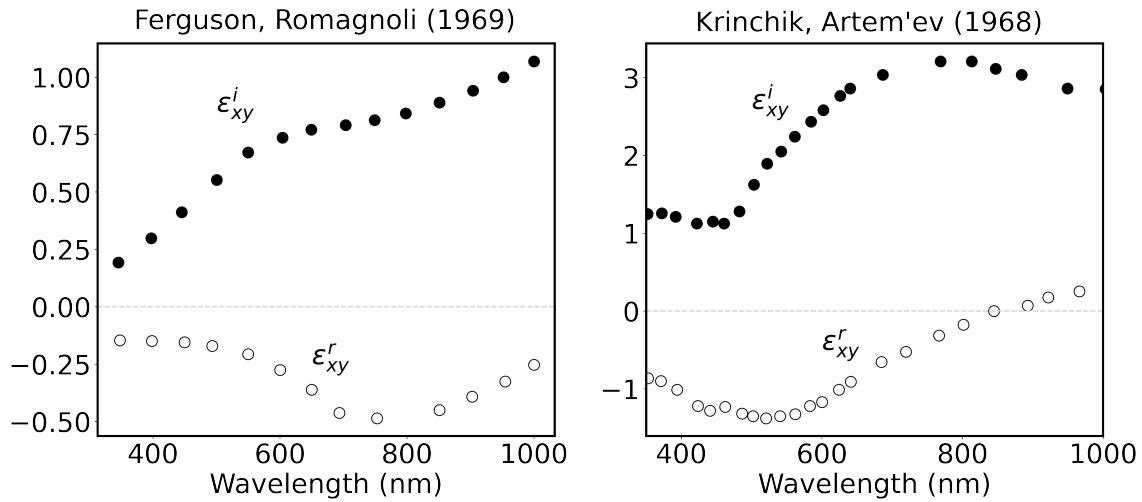


FIGURE 3.5: Spectral real and imaginary components of the off-diagonal elements of the dielectric tensor of Fe for two sets of data: Ferguson, Romagnoli (1969) [32] and Krinchik, Artem'ev (1968) [31].

In all simulation work, only the Fe ferromagnetic layer was considered to be anisotropic, while all other layers were simply described by a diagonal matrix, whose diagonal entries are equal and correspond to the isotropic complex permittivity. A dielectric tensor is generated for each desired wavelength and the reflectance spectrum for a given configuration of layered media is returned. This allows not only the observation of SPPs, but also the band splitting for magnetizations from M_s to $-M_s$, and their dependence on wavelength and Fe layer thickness.

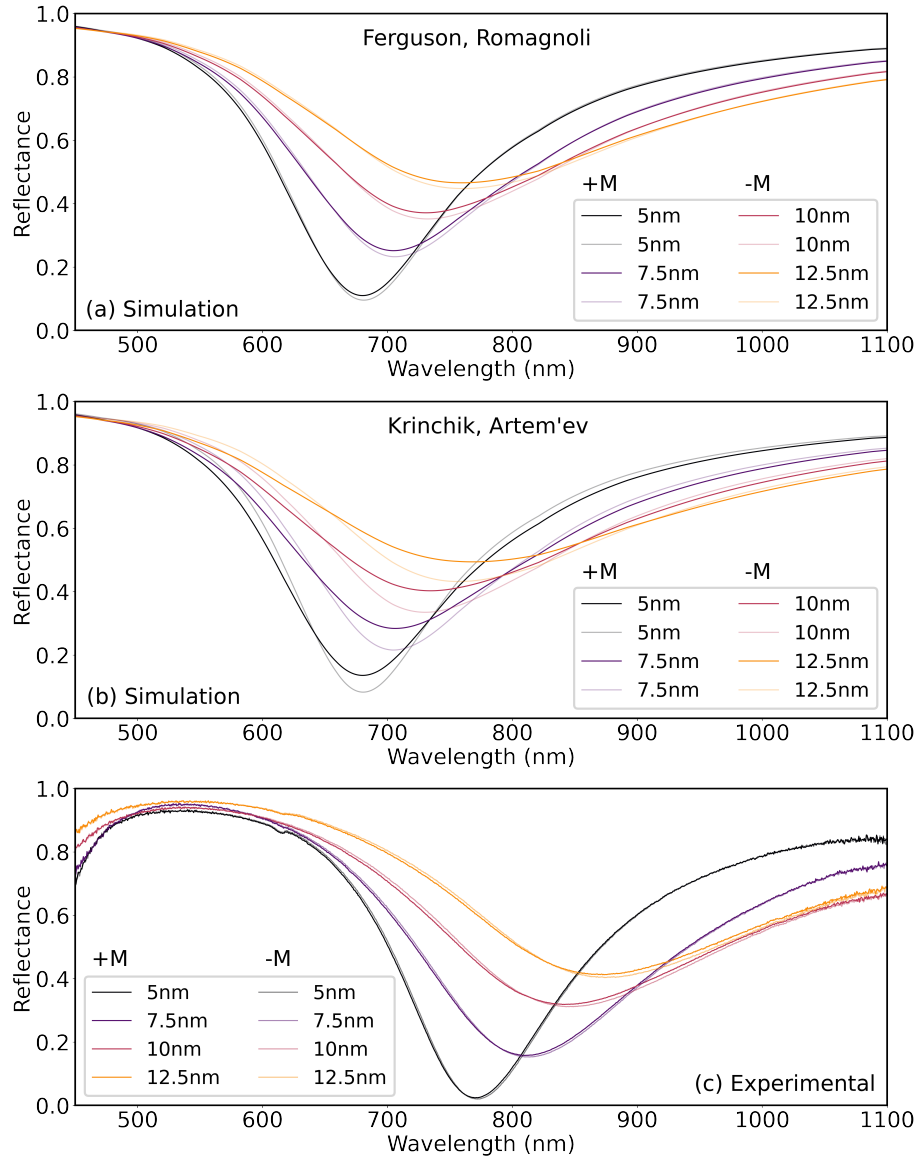


FIGURE 3.6: Reflectance spectra of simulated and experimental films of Ag/Fe/TiO₂ trilayer for 30nm Ag, 40nm TiO₂ and varying thicknesses of Fe ($n=1$): (a) Ferguson, Romagnoli [32], (b) Krinchik, Artem'ev [31] and (c) experimental data.

The magneto-optical response of structures with varying thicknesses of Fe is plotted in Figure 3.6 for the off-diagonal elements from [31] and [32]. The structures are composed of layers with 30nm of Ag and 40nm of TiO₂ sandwiching a layer of Fe with thicknesses of 5, 7.5, 10 and 12.5nm. As previously seen, the experimental spectrum shows a significant red shift compared to the simulated ones and slightly higher plasmonic absorption in the vicinity of the SPR wavelength. The increase in optical damping of the plasmonic bands is also notorious for increasing thicknesses of the Fe layer and in line with simulated results.

The obtained magneto-optical modulation for opposite polarity saturation magnetizations are fairly modest, but definitely noticeable for both simulated and experimental spectra. When compared to the simulated spectra, the experimental spectrum shows smaller magneto-optical modulation, although the profile of the modulation appears to be similar, with an enhanced effect in the proximity of the SPR wavelength that wears off away from the absorption peak. A study on the dependence of the absolute change in SPP wavelength, $\Delta\lambda_{SPP}$, and reflectance, ΔR_{SPP} , in the thickness of the Fe layer is shown in Figure 3.7. Regarding $\Delta\lambda_{SPP}$, the experimental data and the simulated spectrum for Krinchik *et al* off-diagonal elements show an increase along Fe thickness (although much larger in the latter), whereas the data extracted from Ferguson *et al* does not vary significantly. The drop in reflectance for opposite magnetization states does not seem to depend significantly in Fe layer thickness for the simulated spectra, although a small, constant increase is noticed for the experimental data, albeit smaller in magnitude.

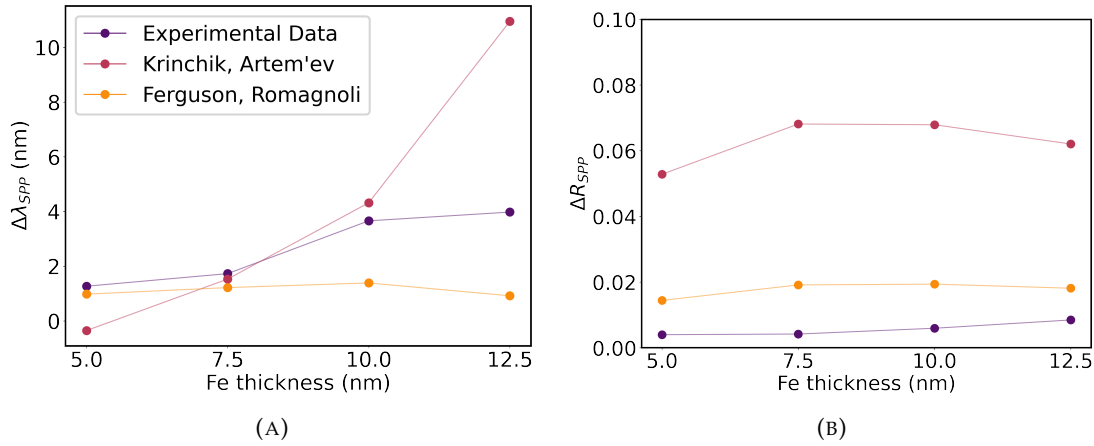


FIGURE 3.7: (A) SPR wavelength peak and (B) reflectance shifts for opposite polarity saturation magnetizations of Ag/Fe/TiO₂ trilayers for varying thicknesses of Fe ($n=1$) in simulated and experimental spectra.

This difference in behavior of resonance wavelength and reflectance shift is related both to the different magnitudes of the real and imaginary parts of the off diagonal elements and respective spectral dependence. The magnetization-induced shift in resonance wavelength depends greatly on the real part of the off diagonal elements, whereas the drop in reflectance is more intimately related to the magnitude of the imaginary part.

A superficial analysis of the data shown in Figure 3.7 could maybe suggest that the off-diagonal elements of the experimental films show a different profile than those extracted from literature, with an enhanced real part compared to [32] but more modest compared to [31], while the complex part would be smaller than those of both referenced

sources. These claims, while not necessarily false, can be excessive, since a direct comparison ignores potential differences in the diagonal dielectric elements of simulated and experimental structures, for instance. In fact, such differences in the values of the diagonal permittivity elements could possibly justify the red shift in the resonance peak, that in turn would also affect the effect of the off-diagonal elements, given their spectral dependence. Inspecting the experimental bands, one can estimate their resonance wavelength to shift from around 770 to 870nm. In said range, the Ferguson off-diagonal permittivity decreases slightly (in module) for the real part, and increases slightly for the imaginary part. In the same range, the real part of Krinchik data diminishes in module, whereas the imaginary part plateaus around a maximum.

The experimental data does not clearly fit any of the references. While the shift in SPR wavelength does increase with the Fe layer thickness, it is uncertain if the increased shift strictly relates to the spectral dependence of the magneto-optical effects, or simply to the cumulative effect of light propagating larger thickness.

In order to make the simulation work more precise and capable of previewing experimental results, a scaling to the magnitude of the off-diagonal elements was made by simply multiplying the entire spectral elements by a scaling factor of 0.275, simulating more accurate spectra while maintaining the same spectral relative composition and relative composition between real and imaginary parts.

The determination of the exact magneto-optical properties of the fabricated films, namely the off-diagonal components, would require magneto-optical ellipsometry measurements, an advanced technique that determines optical constants based on the change of the polarization change and phase difference of light reflected or transmitted in thin films, that was unfortunately out of reach for the time frame of this work. It was however possible to characterize some convenient magnetic properties of the fabricated films, namely the saturation magnetization by SQUID [2.3.2](#) and the saturation magnetic field and magnetic coercivity both by SQUID and simply resorting to the Kretschmann configuration setup.

3.2 Magnetic Characterization of Thin Films

The characterization of magnetic properties of the fabricated films can be useful for a complete study of magneto-optical properties. Knowing the absolute value of magnetization within a sample allows us to better perceive the magnitude of the spectral dependent

magneto-optical coefficients in the off-diagonal elements.

The measurement of net magnetic moment in a series of plasmonic thin films was carried out by a SQUID. The thin films were deposited in small glass substrates, cut to fit inside the SQUID magnetometer sample holder, that consists of a quartz crystal straw that restricts sample dimensions to around $6\text{mm} \times 5\text{mm} \times 4\text{mm}$. The thin films composition was identical to those characterized in 3.6, with the same varying thicknesses of the Fe layer.

The previous SPR characterizations allowed to estimate the saturation magnetic field, according to the magnetic field calibration, to under 30mT. To account for an unexpected outcome, the SQUID measurements were made with a safe margin, ranging from -50mT to 50mT. All measurements were performed at the controlled temperature of 300K, discarding any temperature dependence. The obtained results are plotted in Figure 3.8.

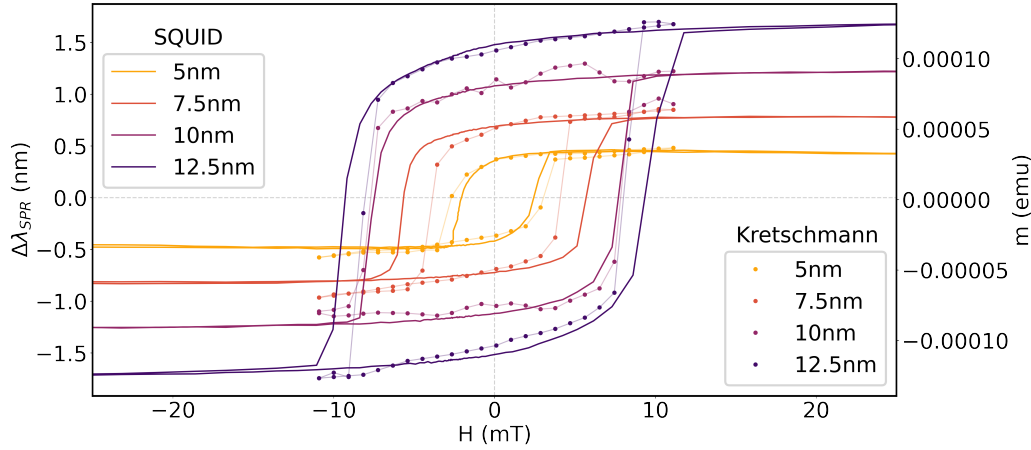


FIGURE 3.8: Hysteresis cycles obtained from SQUID measurements and spectral interrogation in Kretschmann setup.

Identical films to those characterized in the SQUID magnetometer were characterized in the Kretschmann setup for sweeping values of calibrated magnetic field. Since the wavelength shifts due to magnetization changes are small and the spectrometer signal is noisy, five spectra were captured for each magnetic field, and the respective average signal was calculated. These measurements take some time, so that measurements at high currents lead to significant induction coil heating. To avoid any temperature-related problems, the magnetic field sweep measurements were halted when no change in the SPR band was noticeable. For all the spectra, the SPR wavelength (characterized by a minimum in reflectance) was determined and plotted together with the respective magnetic field. Such plot is also depicted in Figure 3.8, for the films with different thicknesses

of the Fe layer.

Upon brief inspection of Figure 3.8, it is clear that magnetic hysteresis – typical of ferromagnetic materials – occurs in both measurements, verifying at least some degree of ferromagnetic behavior from the Fe layers, and the explicit dependence of the magneto-optical effects in the magnetization, and not the magnetic field. A good relative match between the hysteresis cycles of both methods was attained, although such condition wasn't necessarily expected for the hysteresis cycles measured in the Kretschmann setup. As mentioned before, the spectral dependence of the magneto-optical effects, and the sensitivity of the SPR wavelength to layer thicknesses, make it possible for a layer of smaller thickness to display higher shift in resonance wavelength for opposite magnetization polarities than a layer of higher Fe thickness. This condition would result in the layer of smaller thickness to display a 'taller' hysteresis cycle than the thicker layer. For the particular studied configurations, that is not the case, as evidenced in Figure 3.7 and 3.8.

The hysteresis cycles should, however, match in the values of saturation magnetic fields and coercivities, as the same value of saturation magnetic field is responsible for the saturation of net magnetic moment, as measured by the SQUID, and the saturation of the SPR peak, as measured in the Kretschmann setup. A comparison of results for saturation magnetic field, H_s , and coercive field (or coercivity), H_c , is depicted in Figure 3.9 for varying thicknesses of Fe. In this regard, the match is fairly satisfactory, as there is a clear matching tendency between SQUID and Kretschmann setup measurements for increasing thickness of Fe layer. The increase in Fe thickness appears to increase the saturation magnetic field, a behavior not typical for bulk samples but reported for thin films of Fe below a critical thickness [33]. The saturation magnetic field and coercivity of the film with 5nm of Fe for the Kretschmann setup cycle seems to be slightly larger than for the SQUID measurements, a tendency that inverts for the films of higher thicknesses. Such deviations from the SQUID measurements can be attributed to errors in thickness deposition of thin films, to errors in the determination of the SPR wavelength or in the magnitude of the measured and actually sensed magnetic field. The calibration for thin film deposition was performed for films with tens and hundreds of nanometers, but for very thin films, under or around 10nm, the accounted error is more significant. Given the small magnitudes of the SPR peak shifts, and the large FWHM of plasmonic bands for large Fe layer thicknesses, the determination of the SPR wavelength peak can also induce significant errors to the measured results. This difficulty of measurement is clear for the Kretschmann setup

hysteresis cycles, that are visibly more noisy and with more poorly defined profiles than those exhibited by SQUID measurements. All measured values regarding the magnetic properties of the Fe thin films are displayed in Table 3.1.

Nonetheless, an interesting, distinct method for visualizing hysteresis cycles in magneto-optical thin films was tested and found to be useful for good estimated of magnetic properties of such structures, constituting an alternative to MOKE measurements that may require more specialized instruments, such as lasers. The spectral interrogation of the developed setup allows for the use of a simple broadband light-source, and excludes the need for a goniometer for angular interrogation.

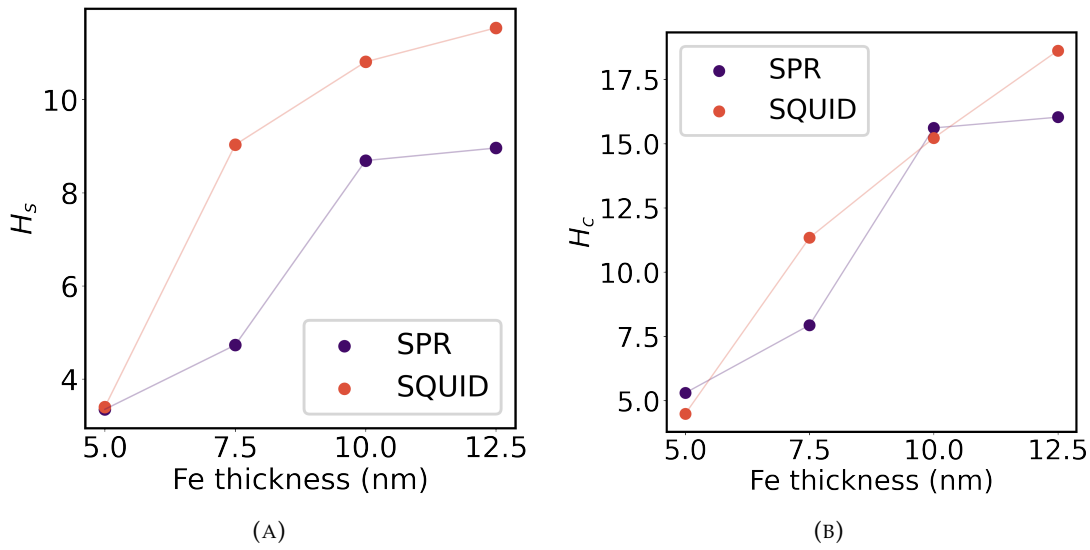


FIGURE 3.9: (A) Comparison of the measured values of saturation magnetic field, H_s , and coercive field, H_c , by the Kretschmann setup and SQUID magnetometer.

From the SQUID measurements, it is clear that for thin films of approximately the same area, the sensed net magnetic moment is proportional to film thickness, as expected. From these measurements, and estimating the volume of each sample, it is possible to estimate the magnetization of the Fe layers. The volume of the Fe layers was estimated by measuring the respective surface area of deposited material from a picture taken with a reference scale in the ImageJ software, and multiplying each area by the deposited thickness. Dividing the hysteresis curves of magnetic moment by the respective volume will return the hysteresis curves of magnetization, as plotted in Figure 3.10a.

Considering identical constituency of the Fe thin films, the magnetization absolute value should be the same. However, it is known that for Fe samples smaller than a bulk critical thickness, there is a thickness dependence on a sample's magnetic properties. As

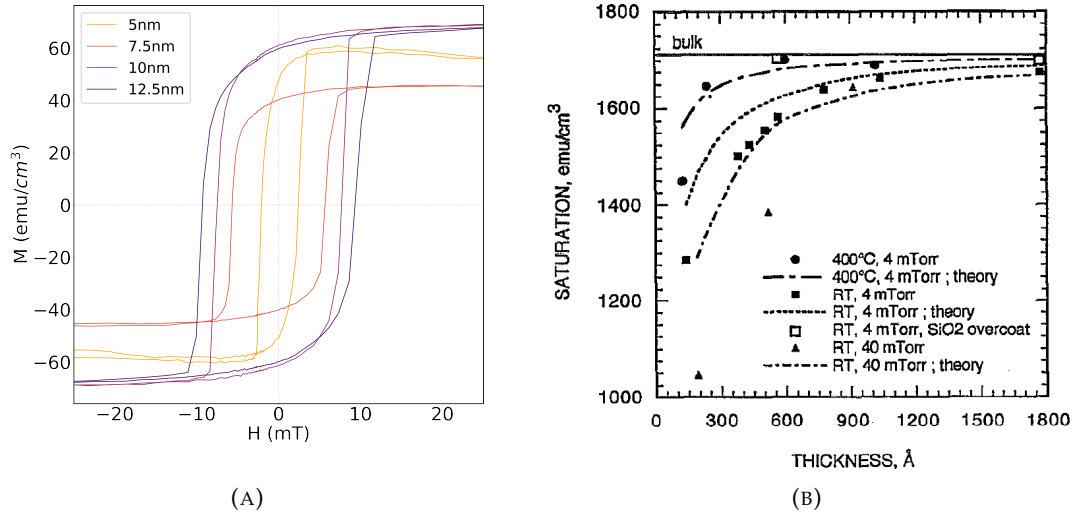


FIGURE 3.10: (A) Magnetization hysteresis cycles obtained from SQUID measurements for varying thicknesses of Fe layer. (B) Thickness dependence of saturation magnetization of Fe thin films, as reported by [33].

reported by [33] (Figure 3.10b), the saturation magnetization appears to increase exponentially for increasing thickness of Fe, converging to a bulk-like regime where it remains constant for sample scaling. This dependence originates on the higher incidence of oxidation in thinner films, where an oxidized passivation layer takes relatively higher proportions of the sample. The higher porosity shown by sputtering deposited films also contributes for such effect, although thinner evaporated films still show smaller saturation magnetization than thicker ones in the thin film regime [33]. These facts explain the much smaller magnitude of saturation magnetization for all films, and the higher saturation magnetizations obtained for the 10 and 12.5nm films. They do not, however, explain the higher magnetization registered for the 5nm than for the 7.5nm film, although errors in the thickness of the deposited films and on the measurement of film area may justify said anomaly. For the deposition of the SQUID samples, a delimited area on the substrate was masked with tape to better control the deposited area for volume estimation. In the 5nm and 7.5nm samples, the tape did not stick well to the substrate and some borders were faded, increasing the error in area determination.

It is worth to note that the limited number of points taken on the Kretschmann characterization of the hysteresis cycles impeded the precise measurement of the values of saturation magnetic field and coercive field. Additionally, the band broadening observed for the films with higher Fe thickness increases the error in the determination of the resonance wavelength, and thus of the normalized magnetization hysteresis cycles.

TABLE 3.1: Magnetic properties of Fe thin films of different thicknesses.

t_{Fe} (nm)	SPR		SQUID			
	H_s (mT)	H_c (mT)	H_s (mT)	H_c (mT)	m_s (10^{-5} emu)	M_s (emu/cm ³)
5	3.35	5.30	3.40	4.49	3.52	60.61
7.5	4.73	7.94	9.03	11.34	5.88	45.62
10	8.69	15.61	10.81	15.22	8.89	67.35
12.5	8.96	16.03	11.54	18.62	11.97	65.14

The magnitude of the magnetization in Fe thin films is, thus, very small. For reference, bulk samples of Fe at room temperature have been reported to attain magnetizations around 1700 emu/cm^3 , over $25\times$ the value for the 12.5 nm film. These values could be slightly optimized by improving film fabrication process, as to grow them in more ordered arrangements of higher crystallinity that could improve magnetic response.

These fabrication processes raise technical problems that made their implementation impossible in the time-frame of this work. For now, the optimization of magneto-optical effects, described macroscopically by off-diagonal elements in the dielectric tensor, can be made through the optimized enhancement of light-matter interaction, namely through the excitation of ESWs in the vicinity of a magnetic layer.

3.3 Simulation and Characterization of Surface Waves

In the following section, a complete, systematic simulation study is carried out in order to unveil optimized thin film structures for enhanced magneto-optical response. This study was conducted for structures capable of the excitation of either SPRs or BSWs, both for planar and multimode fiber substrate geometries. The simulations were performed for different geometries of layer disposition and for varying thicknesses of all layers within certain limits. For each defined structure, the spectra were simulated for external media of different refractive indices, and for each of these configurations, the spectra for symmetric saturation magnetizations were captured, with off-diagonal elements from [31] adjusted by the aforementioned scaling factor.

The MOSPR curves, as defined by 1.53, were simulated starting from the simulated SPR spectra for opposite polarity saturation magnetizations, $+M$ and $-M$, of each simulated refractive index.

For each set of bands, SPR and MOSPR, the spectral reflectance sensitivity to variations in refractive index was calculated, as defined in 1.54 and 1.55, respectively. These curves quantify the change in reflectance that occurs as the external refractive index is changed for each simulated wavelength of the spectrum. The absolute maximum point of this spectrum, thus, specifies the wavelength of optimized reflectance sensitivity, where the sensitivities of the SPR and MOSPR spectra will be determined. Notice that the optimal wavelength of the SPR sensitivity spectrum will likely differ from the optimal wavelength of the MOSPR's. These two sensitivities are then compared and the structures in which $\eta_{SPR} < \eta_{MOSPR}$ (for η_{SPR} and η_{spr} as defined in 1.54 and 1.55) were registered and fabricated for experimental validation.

The experimental characterization of the fabricated structures is analogous to the simulation process. Spectra for each magnetization were obtained for the same refractive indices used in the simulation. Starting from the SPR spectra, the MOSPR spectra were determined, and then the sensitivity spectra η_{SPR} and η_{MOSPR} were calculated to find the maximum point for sensitivity evaluation.

It is important to note, however, that these two sensitivities cannot be compared directly. Owing to the different natures of the SPR and MOSPR measurements, an adjustment of the signal by the respective noise figure must be made for any sensitivity comparison of experimental data. Such adjustment was made by defining a measure for the minimum detectable signal, S_{min} , as 5 times the standard deviation of the 5 samples taken for each measurement of the signal. The experimental sensitivity, η_E , adjusted for signal-to-noise ratio (SNR) is then given by

$$\eta_E = \frac{\eta}{S_{min}} \quad (3.1)$$

where η is either η_{SPR} or η_{MOSPR} .

For the simulation work, free from the constraint of noisy signals, a standard deviation figure of 3×10^{-4} was defined for both SPR and MOSPR signals, based on the magnitude of measurements of noise of the used spectrometers. This assumption will result in optimistic simulation studies, that will later be verified by experimental validation.

Optimized Magneto-optical structures in which $\eta_{E_{MOSPR}} > \eta_{E_{SPR}}$ can be useful in the measurement of very small refractive index changes, where the conventional SPR counterpart signal may be below the limit of detection. These structures could be implemented in sensors for large variations of refractive index through the standard SPR that would

also be capable of detecting small variations of refractive index by magnetic modulation of the ferromagnetic layer.

3.3.1 MOSPR based sensing structures

A set of magnetoplasmonic structures were simulated for reflectance sensitivity to refractive index variations. Being of plasmonic nature, these coatings face significant constraints on the range of possible thicknesses, due to the need for plasmonic and ferromagnetic metals, both with considerable optical losses.

A total of four target materials were used in the simulations and experimental fabrication: gold (Au) and silver (Ag) as plasmonic metals, iron (Fe) as ferromagnetic metal, and titanium dioxide (TiO_2) as a dielectric, apart from air and water-based solutions as external medium.

A. Planar Substrate

Several configurations of layer disposition were considered in the simulations of planar substrates. Bilayer structures, with Ag/Fe geometry, and trilayer structures of Au/Fe/Au, Ag/Fe/ TiO_2 , Ag/Fe/Ag and Fe/Ag/ TiO_2 geometry were simulated for their magneto-optical response. For all these simulated geometries, only Au/Fe/Au and Ag/Fe registered configurations where the sensitivity for the magneto-optical modulated structure was higher than the convention SPR counterpart.

Au/Fe/Au trilayers

The thickness range for each layer was estimated by simulation to get a reasonable equilibrium of SPR excitation and magneto-optical behavior. A very thin layer of Au will result in wide plasmonic bands of poor resonant behavior, although more light gets transmitted and is thus capable of interacting with the Fe layer. In the other hand, a thick layer of Fe could potentiate increased magneto-optical behavior, as a greater share of the optical path is through ferromagnetic material, however the optical damping of the plasmonic band due to absorption in the Fe layer would make its interrogation less precise.

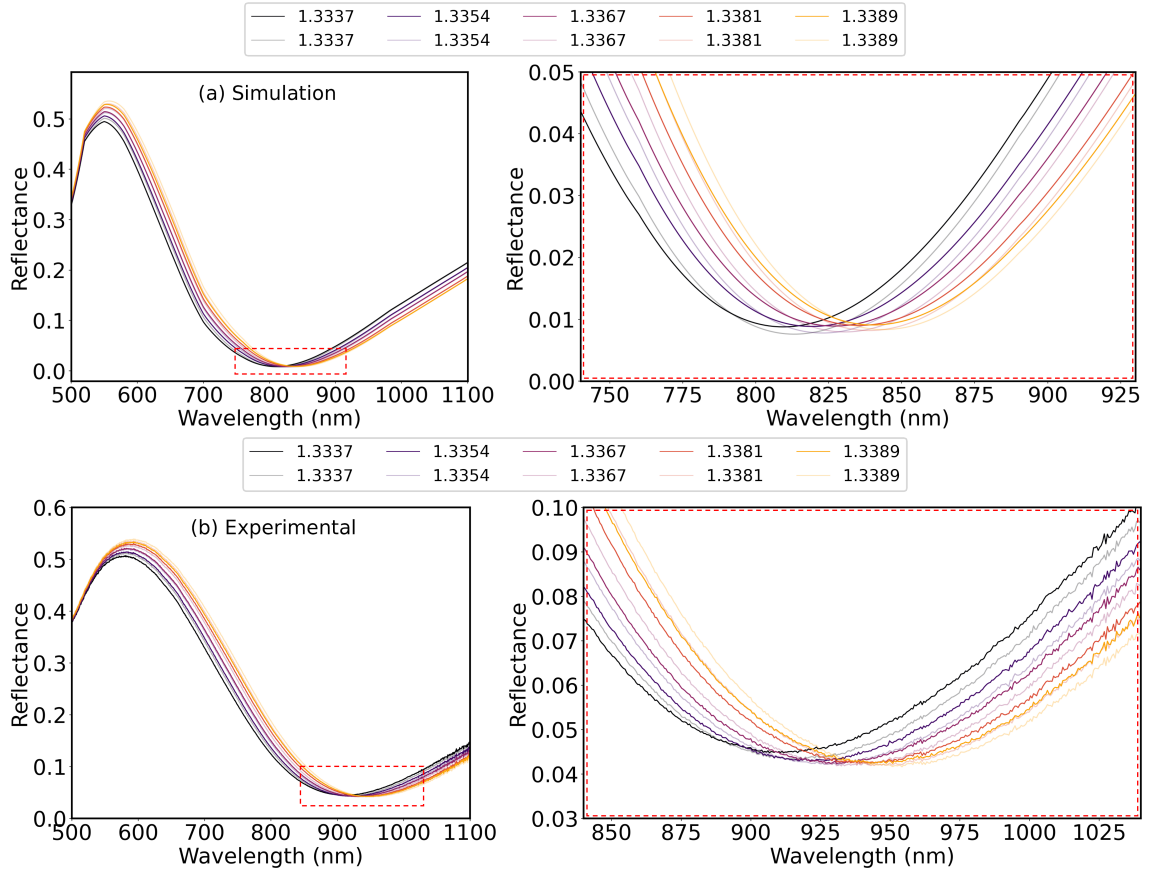


FIGURE 3.11: Reflectance spectra of (a) simulated and (b) experimental structure of 10nm Au/15nm Fe/10nm Au.

For nomenclature simplification, the first Au layer can be regarded as Au_1 and the second Au layer as Au_2 . The simulated thicknesses for layer Au_1 were $t_{Au_1} = [10, 15, 20, 25, 30] \text{ nm}$ and for layer Au_2 were $t_{Au_2} = [10, 15, 20, 25] \text{ nm}$. The middle Fe layer was simulated for thickness in $t_{Fe} = [5, 7.5, 10, 12.5, 15] \text{ nm}$. Each possible thickness combination between these values was simulated, and two structures were returned with an optimized magneto-optical sensitivity: 10nm Au/15nm Fe/10nm Au and 15nm Au/12.5nm Fe/10nm Au. These two structures were fabricated and characterized for the same refractive indices, and for both saturation magnetizations. The simulation and experimental spectra of the 10nm Au/15nm Fe/10nm film are plotted in Figure 3.11

The simulated and experimental spectra have a very similar profile, apart from the expected red shift of the experimental film. A higher refractometric sensitivity is also noticeable, as the experimental bands seem to shift slightly more for refractive index change. The magneto-optical behavior is also similar, with a small deviation being noticeable for opposite magnetization polarities. The MOSPR spectra were then both simulated and determined from the plasmonic bands in Figure 3.11, and the resultant bands are displayed

in Figure 3.12.

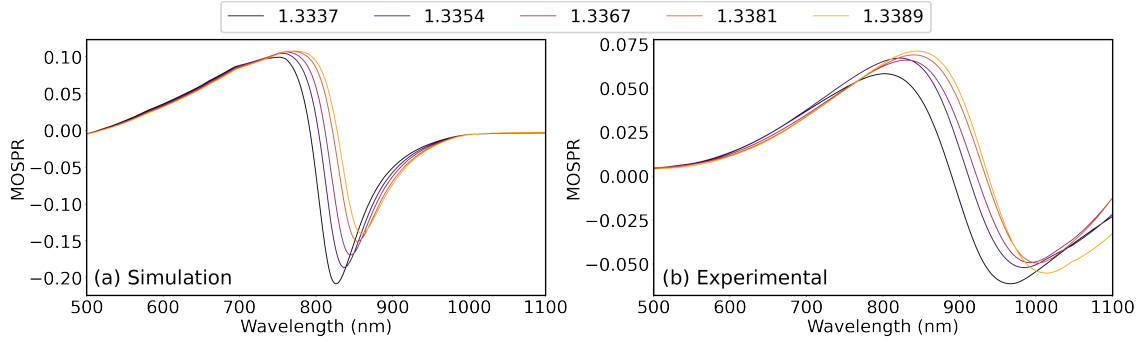


FIGURE 3.12: MOSPR spectra of (a) simulated and (b) experimental structure of 10nm Au/15nm Fe/10nm Au.

The correspondence between simulated and experimental MOSPR spectra is not as clear here. Not only is the amplitude of the signal for the experimental spectrum smaller, but it is also less well defined for refractive index variation. In the near infra-red limit of the experimental spectrum, the signal loses its profile of a Fano-like resonance, and gets noisy beyond possible refractive index detection. The spectral sensitivity curves, as defined by 1.54 and 1.55, were determined and are available in the appendix C. Despite the noticed deviant behavior noticed in 3.12, the variation of reflectance with refractive indices follows similar profiles for both SPR and MOSPR structures, as evidenced by C.1 and C.2, apart from aforementioned red shift of the experimental data.

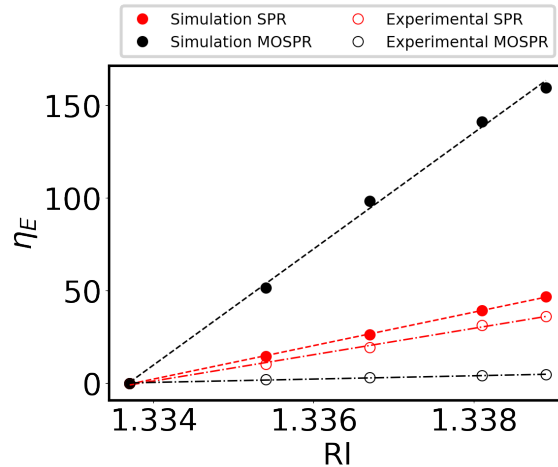


FIGURE 3.13: Simulated and experimental sensitivities adjusted for noise figure of 10nm Au/15nm Fe/10nm Au.

Setting the wavelength of highest sensitivity for the SPR and MOSPR signals, the sensitivity of reflectance changes for external variations of refractive index were measured, and as demonstrated in Figure 3.13, no enhancement of such sensitivity was attained by

the magneto-optical modulation, contradicting simulation results. This outcome comes as no surprise, given the disparity in the magnitude of the η_{MOSPR} between simulated and the actual experimentally obtained signal.

For the other structure configuration, 15nm Au/12.5nm Fe/10nm Au, the same procedure was carried out, obtaining the SPR simulated and experimental bands plotted in Figure 3.14.

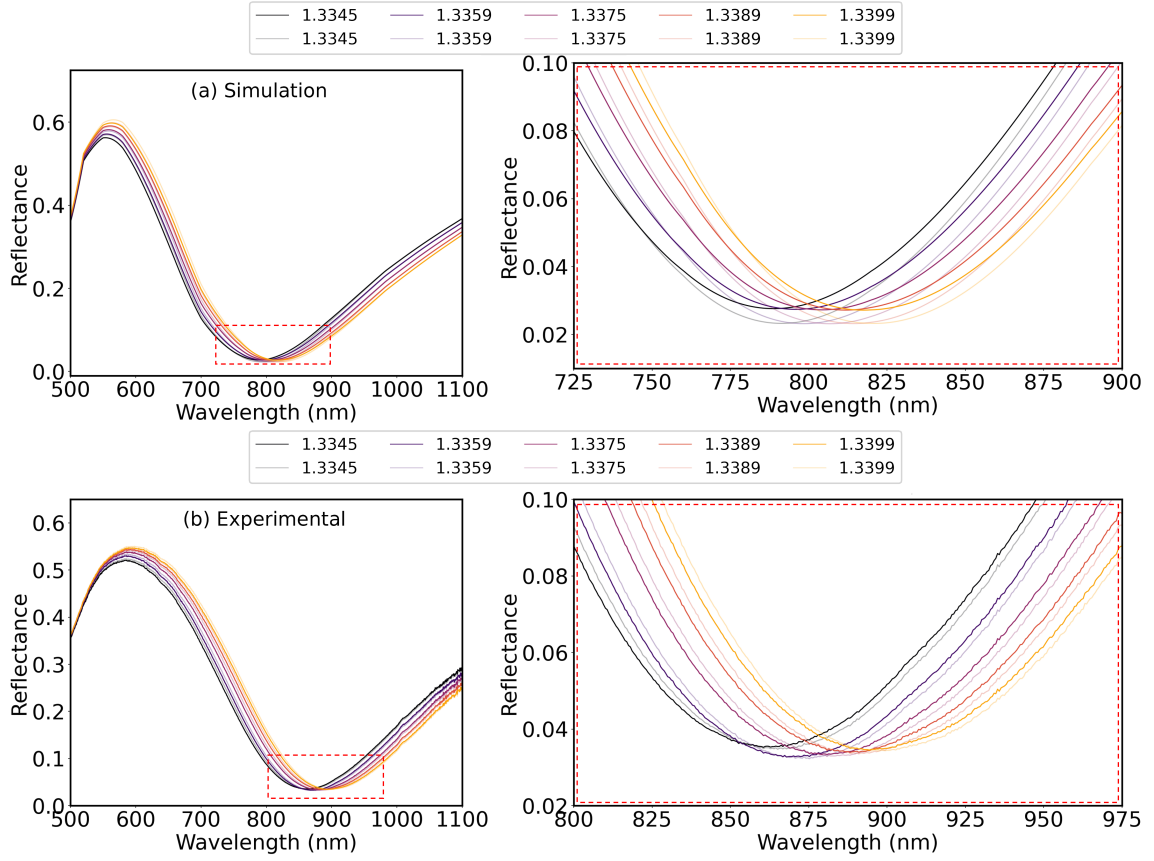


FIGURE 3.14: Reflectance spectra of (a) simulated and (b) experimental structure of 15nm Au/12.5nm Fe/10nm Au.

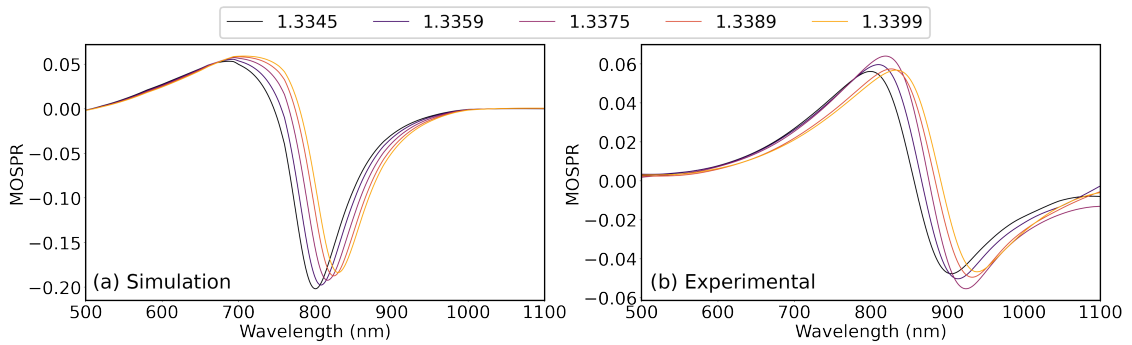


FIGURE 3.15: MOSPR spectra of (a) simulated and (b) experimental structure of 15nm Au/12.5nm Fe/10nm Au.

The simulated and experimental SPR spectra in Figure 3.14 match well, and an improvement in the MOSPR match is arguably noticeable (Figure 3.15), although a significant difference in magnitude of the signal is still present. This better match in magneto-optical optical signal can in part be explained by the thicker layers of Au and Fe in the film when compared to the previous structure. For films this thin, the optical constants of materials are heavily dependent on thickness, and since the optical constants applied in the simulations were extracted from a source referencing thicker films of 20 to 50nm [29],[30], it is only natural that a proximity to simulation results is successively obtained. Nonetheless, and despite a very good match of simulated and experimental spectral sensitivities (Figures C.3 and C.4 in C), no enhancement of the sensitivity was attained for this configuration as well, as demonstrated in Figure 3.16.

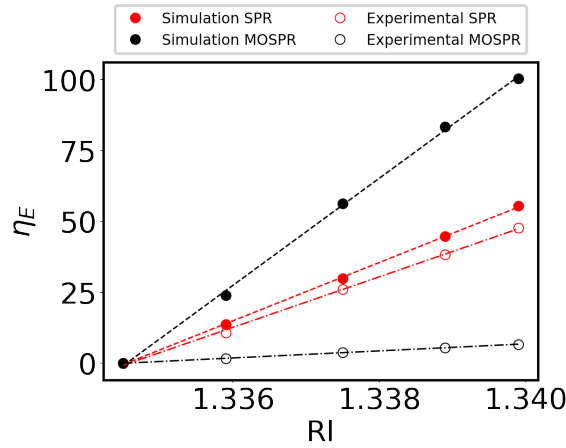


FIGURE 3.16: Simulated and experimental sensitivities adjusted for noise figure of 10nm Au/15nm Fe/10nm Au.

Ag/Fe bilayers

For the thin films of Ag/Fe geometry, thicknesses of $t_{Ag} = [15, 20, 25, 30, 35, 40]nm$ and $t_{Fe} = [5, 7.5, 10, 12.5, 15]nm$ were simulated for all possible bilayer combinations. From all possible configurations, two structures stood out as prone to refractive index sensitivity enhancement by magneto-optical modulation: 15nm Ag/10nm Fe and 15nm Ag/12.5nm Fe.

Regarding the former structure, 15nm Ag/10nm Fe, a direct comparison between simulation and experimental spectra SPR spectra are plotted in Figure 3.17. The resemblance between spectra is poorer than for previous ones, with a significant red shift of experimental bands. Such occurrence was already expected, as it was seen in Figure 3.4 where

both the Ag and Fe appeared to contribute to the red shifting of plasmonic bands. The plasmonic bands are also extremely broad due to low Ag thickness, that does not grant optimal plasmonic properties, and to the relatively high thickness of Fe, that induces great optical losses in the signal. The fact that only a plasmonic metal layer is present, contrary to the Au/Fe/Au trilayers, reduces the reflectivity of the films away from the resonance wavelength. The zero reflectance noticeable in the simulated spectra is also, again, related to the optical constants of the referenced data, that assumes the optical properties of a metallic silver sample in a thin film of limited conductivity.

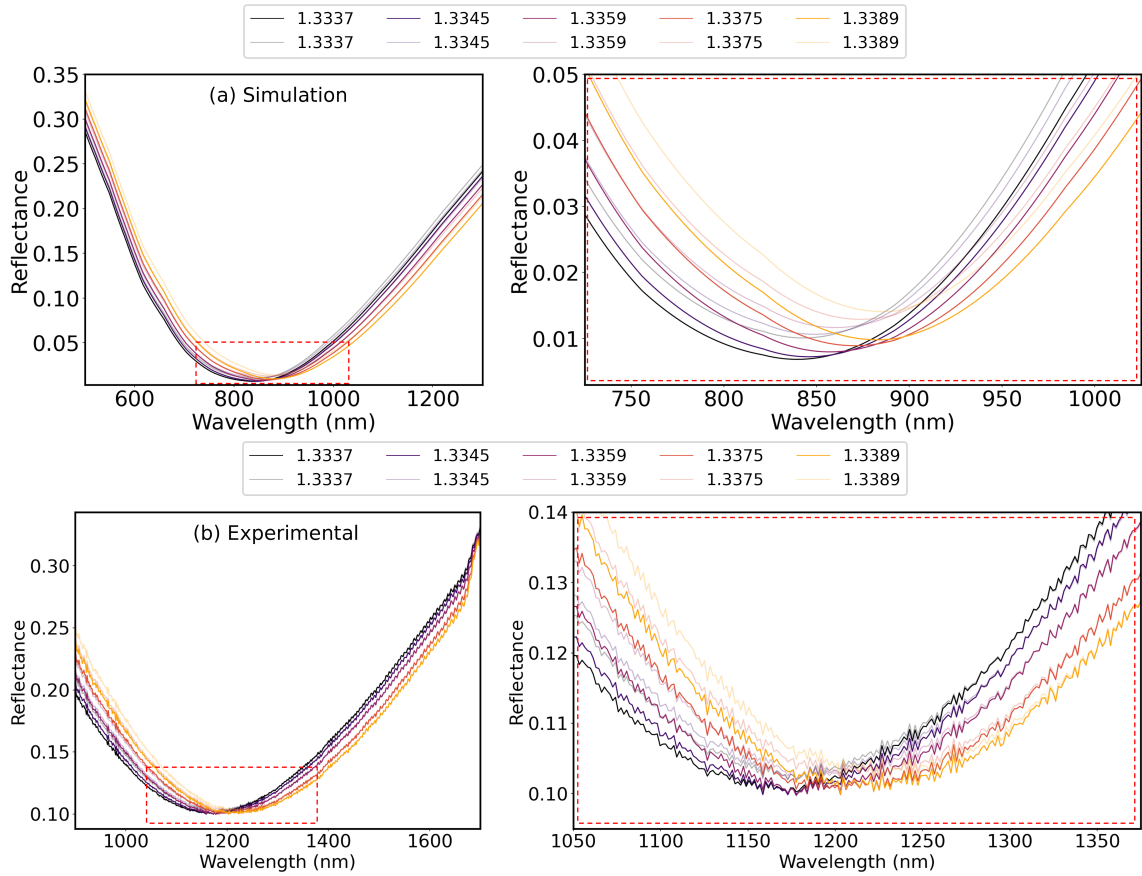


FIGURE 3.17: Reflectance spectra of (a) simulated and (b) experimental structure of 15nm Ag/10nm Fe.

This way, the experimental MOSPR band, depicted in Figure 3.18, also shows a wider profile than the simulated one, although the profile follows a similar trend, indicating coherent magneto-optical response. The η_{SPR} curves, shown in Figure C.5 in appendix C are concordant. That is not the case of η_{MOSPR} , whose difference in magnitude did not provide an enhanced sensitivity for the magnetically modulated approach, compared to the conventional SPR reflectance interrogation (Figure 3.19).

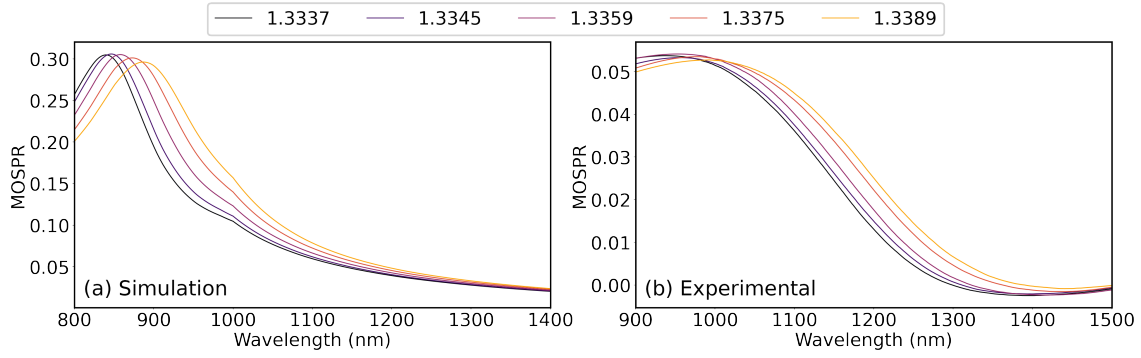


FIGURE 3.18: MOSPR spectra of (a) simulated and (b) experimental structure of 15nm Ag/10nm Fe.

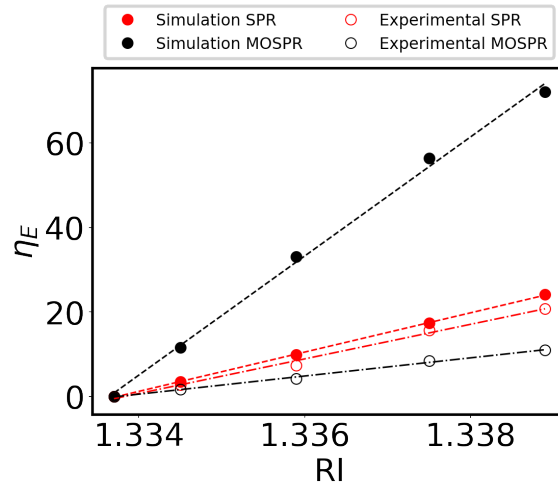


FIGURE 3.19: Simulated and experimental sensitivities adjusted for noise figure of 15nm Ag/10nm Fe.

The other Ag/Fe geometry structure, 15nm Ag/12.5nm Fe, was lastly fabricated and characterized for varying refractive indices. The reflectance and the magneto-optical modulated spectra are shown in Figure 3.20 and Figure 3.21, respectively.

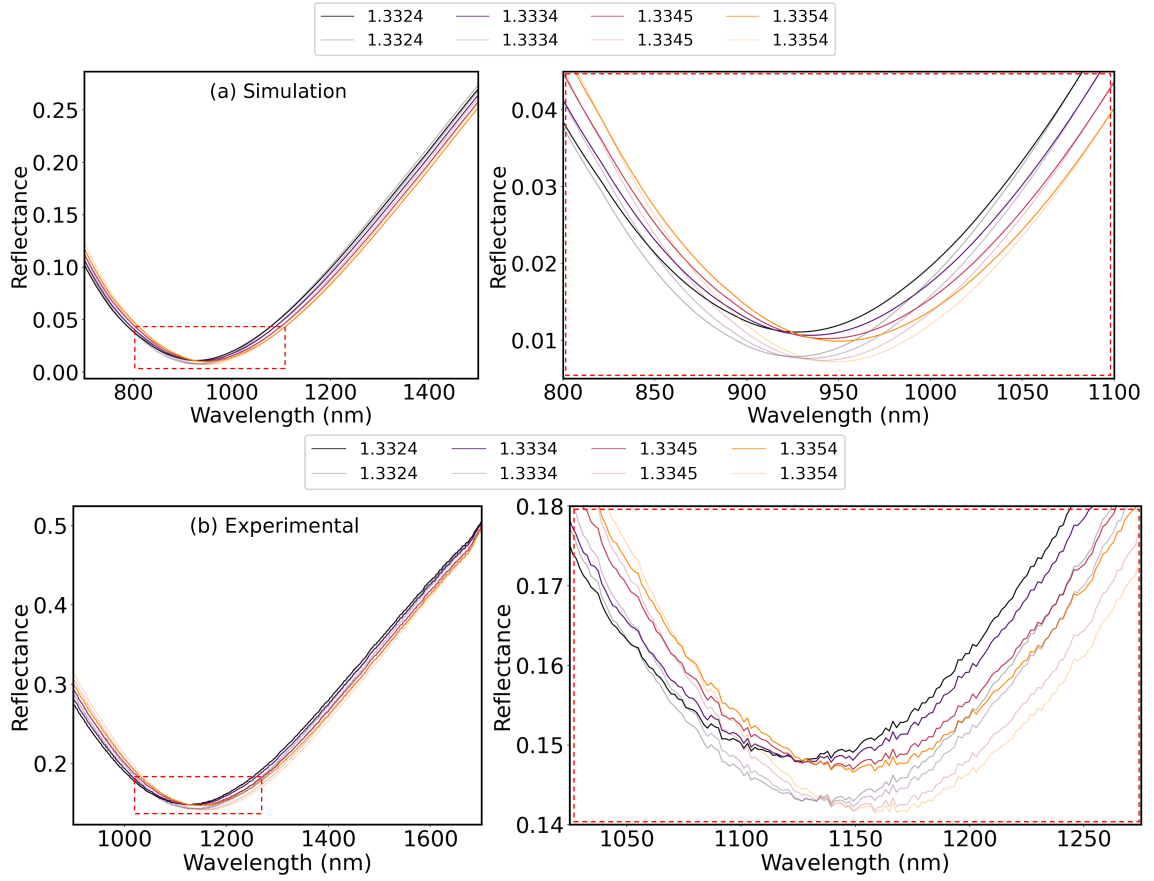


FIGURE 3.20: Reflectance spectra of (a) simulated and (b) experimental structure of 15nm Ag/12.5nm Fe.

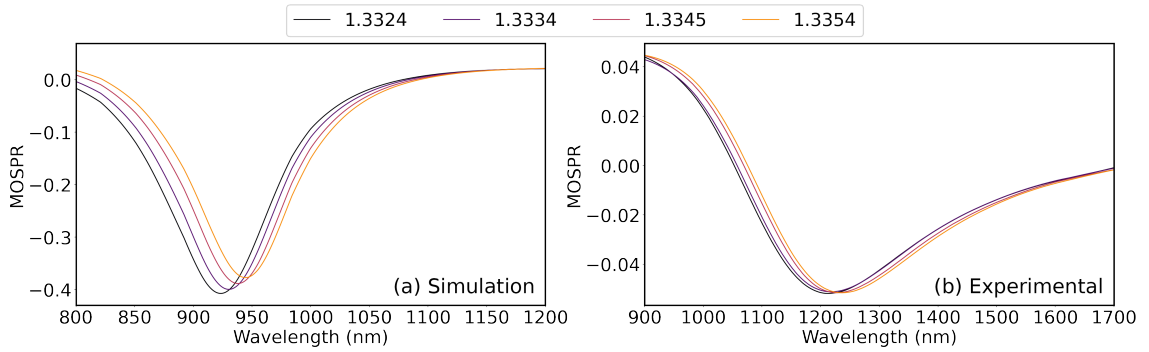


FIGURE 3.21: MOSPR spectra of (a) simulated and (b) experimental structure of 15nm Ag/12.5nm Fe.

The plasmonic bands are similar, and display a similar behavior noticed for the 15nm Ag/10nm Fe structure, with a wide plasmonic band and an offset in reflectance at resonance frequency. This time, however, there seems to be more prominent magneto-optical modulation of the plasmonic bands, with a more expressive shift of resonance wavelength. The MOSPR bands demonstrate a also similar profile, although with a wider

dip, and inferior signal magnitude. This difference in signal intensity is reflected on the η_{MOSPR} curves of Figure C.8 in appendix C. Despite this inconsistency with the simulated curves, it was still possible to obtain an enhanced experimental sensitivity of the magneto-optical MOSPR band in comparison to the conventional reflectance spectrum. The sensitivity is shown in Figure 3.22.

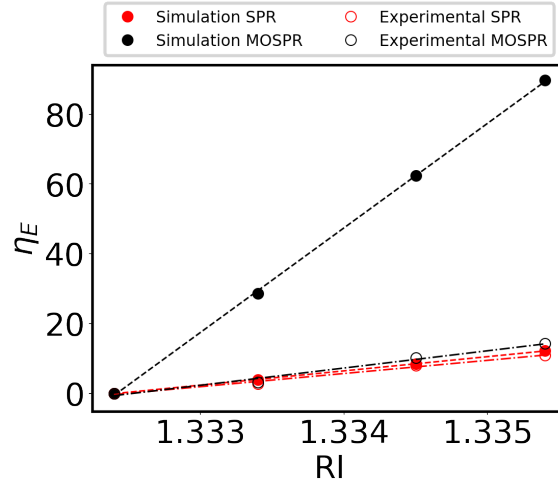


FIGURE 3.22: Simulated and experimental sensitivities adjusted for noise figure of 15nm Ag/12.5nm Fe.

An increase in refractive index sensitivity of the 15nm Ag/12.5nm Fe structure was obtained by magneto-optical modulation of the SPR signal. The increase in the thickness of the Fe layer appears to have intensified the magneto-optical effects just enough for the overtake of MOSPR sensitivity to conventional SPR one.

B. Multimode Fiber

A similar simulation routine was followed for the same geometries of plasmonic films in multimode optical fiber matrix. For the same simulations parameters, namely the magnitude of off-diagonal coefficients applied in planar substrates, no configuration of any geometry was predicted to grant increased sensitivity by interrogation of MOSPR signal, instead of by the conventional SPR bands. Nonetheless, a multimode fiber segment was fabricated with the configuration that yield positive results for planar substrates, the bilayer 15nm Ag/12.5nm Fe.

The simulated and characterized reflectance spectra of the plasmonic bilayer structure in fiber is sketched in Figure 3.23. The match between both these spectra is poorer than in the case of planar substrate films. It is also interesting to notice that the simulated

resonance wavelength for optical fibers appears blue shifted in comparison to the planar counterparts. Such feature is due to the bigger incidence of higher angles in multimode fiber interrogation, where angle integration ranges from the critical angle up to 90° .

The blue shift of the experimental data in comparison with the simulated data is likely related to the calibration of the sputtering system for multimode optical fiber depositions, or due to errors in the input parameters fed to the simulation related to geometry properties of the fiber, namely fiber core diameter and length of the exposed section where light interacts with the plasmonic film. The reflectance at resonance wavelength is also higher than the simulated value, and a higher sensitivity to refractive index variation is also apparent.

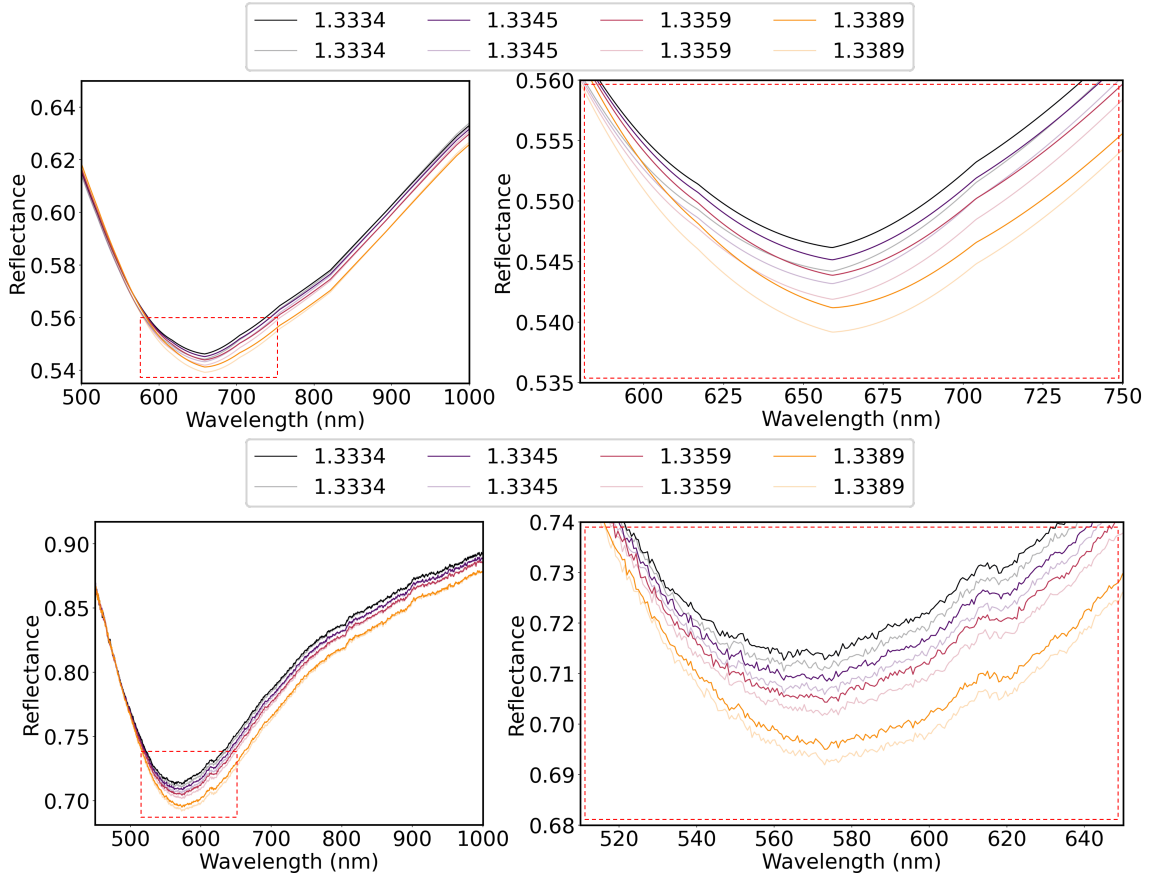


FIGURE 3.23: Reflectance spectra of (a) simulated and (b) experimental structure of 15nm Ag/12.5nm Fe in multimode fiber.

The simulated magneto-optical modulation in the experimental spectrum is far more modest than in the simulated counterpart, and such difference in behavior is clear in the MOSPR curves, depicted in Figure 3.24. The curves exhibit a similar trend, despite the

disparity in magnitude and the noisy signal. All these conditions led to an inferior sensitivity to refractive index, shown in Figure 3.25, that render the magnetic modulation of this structure in fiber as not fit, at this point, for actual applications in the sensing of refractive index variations.

The reflectance sensitivity to refractive index variation values are determined by a Python based routine that determines the change in reflectance for all interrogated experimental and simulated wavelengths, for each of the characterized refractive indexes. This data is fitted to a linear regression, whose parameters determine experimental and simulation sensitivities. From these values, the final, noise figure adjusted sensitivities ($\eta_{E_{exp}}$ and $\eta_{E_{min}}$) are calculated by simply dividing the sensitivities by the respective noise figure, $S_{min_{exp}}$ or $S_{min_{sim}}$. All these values are present in Table 3.2 and Table 3.3, for the characterization of the SPR and MOSPR spectra, respectively.

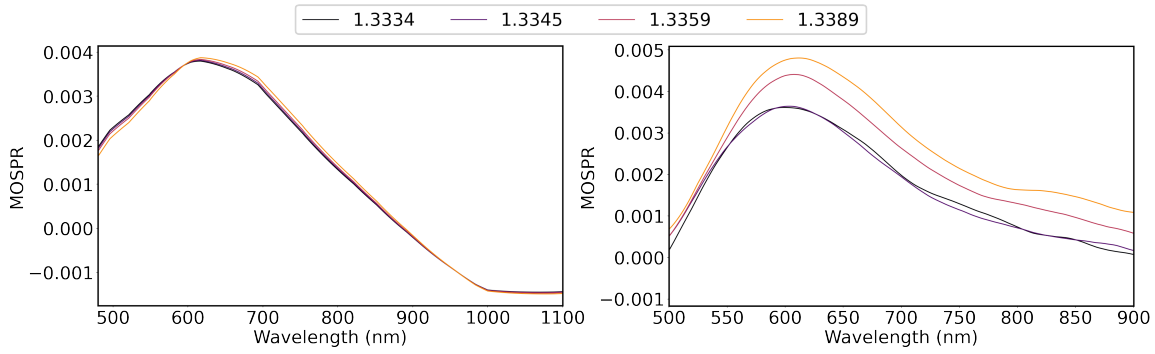


FIGURE 3.24: MOSPR spectra of (a) simulated and (b) experimental structure of 15nm Ag/12.5nm Fe in multimode fiber.

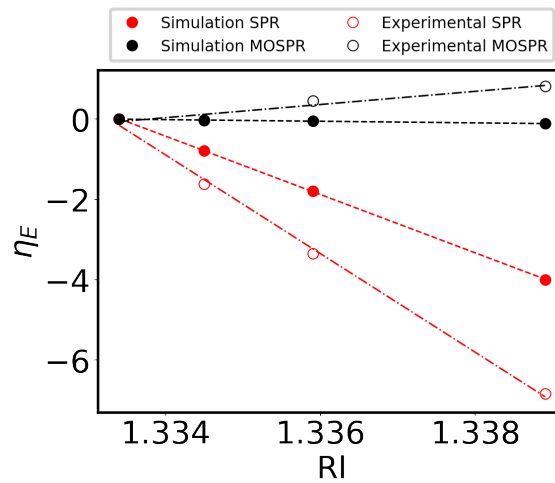


FIGURE 3.25: Simulated and experimental sensitivities adjusted for noise figure of 15nm Ag/12.5nm Fe in MM fiber substrate.

TABLE 3.2: Simulation and experimental parameters for reflectance sensitivity determination for conventional SPR spectra of all fabricated structures.

SPR						
Substrate	Structure	η_{sim}	η_{exp}	$S_{min_{exp}}$	$\eta_{E_{sim}}$	$\eta_{E_{exp}}$
Planar	10Au/15Fe/10Au	13.5	13.1	0.00185	9000	7081
	15Au/12.5Fe/10Au	15.4	16.1	0.00180	10276	8911
	15Ag/10Fe	7.0	7.7	0.00188	4643	4089
	15Ag/12.5Fe	6.1	8.7	0.00230	4077	3777
MM Fiber	15Ag/12.5Fe	-1.4	-4.0	0.00327	-935	-1230

TABLE 3.3: Simulation and experimental parameters for reflectance sensitivity determination for MOSPR spectra of all fabricated structures.

MOSPR						
Substrate	Structure	η_{sim}	η_{exp}	$S_{min_{exp}}$	$\eta_{E_{sim}}$	$\eta_{E_{exp}}$
Planar	10Au/15Fe/10Au	47.0	12.6	0.01410	31323	893
	15Au/12.5Fe/10Au	28.3	12.1	0.00973	18881	1245
	15Ag/10Fe	21.1	2.4	0.00113	14081	2147
	15Ag/12.5Fe	45.0	4.0	0.00081	29977	4946
MM Fiber	15Ag/12.5Fe	-0.04	0.25	0.00157	-26	163

3.3.2 MOBSW based sensing structures

The sensitivity of BSWs to refractive index variations of the external medium has also been employed in sensors for refractive index measurement. Although these electromagnetic modes are generally less sensitive to these changes in external medium properties, the all-dielectric nature of PhCs has the advantage of being capable of operating under harsh and corrosive environments, where the metal films of SPR-based sensors would fail or underperform. The magneto-optical modulation of PhCs has also been addressed, although the inferior confinement of light in BSWs carries limitations in the magnitude of such magneto-optical effects.

For this work, the study of magneto-optical effects on BSWs propagating in PhCs was carried out by the insertion of a planar film of varying thicknesses of Fe. In fact, the dielectric composition of PhCs enables the fabrication of structures for a wider range of Fe thicknesses, as the geometric parameters of the dielectric layers can be optimized to achieve well defined Bloch mode bands that can compensate the optical damping effect of a thicker layer of Fe. All the dielectric layers of simulated PhCs were of TiO_2 and SiO_2 . The considered thicknesses were $t_{TiO_2} = [90, 110, 130, 150, 170, 190, 210, 230, 250]nm$ for the TiO_2 layers, and $t_{SiO_2} = [60, 75, 90, 105, 120, 135, 150, 165, 180, 195, 210, 225, 240]nm$ for SiO_2 layers. The thicknesses of the TiO_2 capping layer of the truncated PhC were simulated for $t_{TiO_2_{cap}} = [30, 40, 50, 60, 70, 80, 90]nm$. The simulation for all possible combinations is time consuming, so that exclusively the configurations where $t_{TiO_2} > t_{SiO_2}$ and $t_{SiO_2} > t_{TiO_2_{cap}}$ were considered. The magneto-optical effects originated from the presence of the Fe layer, simulated for thicknesses $t_{Fe} = [5, 10, 20, 30]nm$.

All simulated and fabricated PhCs contained two unit cells, as a brief preliminary simulation study suggested that the increase in number of unit cells did not appear to improve magneto-optical behavior, even at larger thicknesses. It did, however, excite added guided modes that, together with the optical damping caused by the Fe layer, resulted in poorly defined spectra with sometimes unintelligible and degenerate electromagnetic modes. Besides, the sputtering deposition of PhCs with several unit cells of hundreds of nanometers of dielectric material would take a copious amount of time.

A. Planar substrate

All simulated structures consisted of two unit cells of TiO_2/SiO_2 , followed by the Fe layer, and finally the TiO_2 cap. From the simulated results, many configurations returned an

improved reflectance sensitivity to refractive index variations for the spectra of relative change in reflectance – the magneto-optical Bloch Surface Wave spectrum – MOBSW. Of all these, two structures were fabricated and characterized: $2 \times 170\text{nm TiO}_2/90\text{nm SiO}_2 + 20\text{nm Fe}/80\text{nm TiO}_2$ and $2 \times 170\text{nm TiO}_2/150\text{nm SiO}_2 + 30\text{nm Fe}/80\text{nm TiO}_2$. The dispersion relation of these fabricated structures were numerically calculated and are depicted in Figure 3.27.

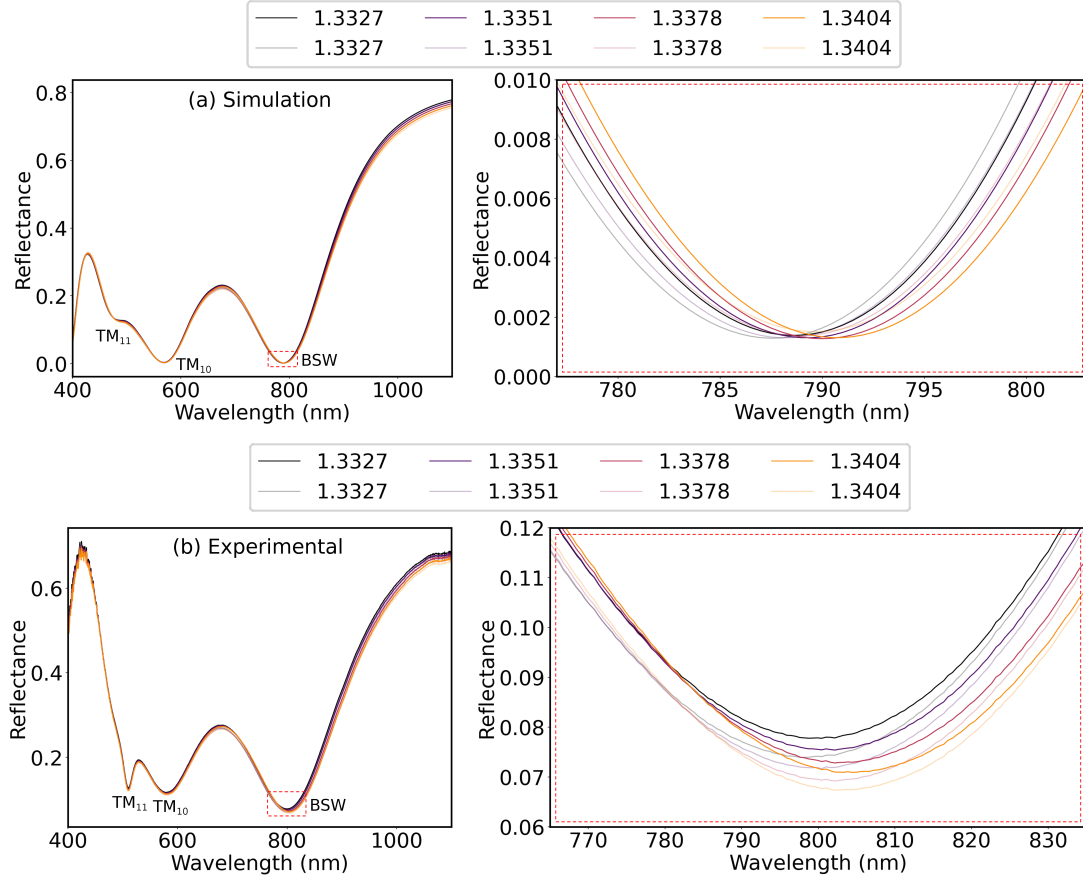


FIGURE 3.26: Reflectance spectra of (a) simulated and (b) experimental structure of $2 \times 170\text{nm TiO}_2/90\text{nm SiO}_2 + 20\text{nm Fe}/80\text{nm TiO}_2$.

The reflectance spectra of the former is plotted in Figure 3.26 in a direct comparison of simulation and experimental results. The match between simulation and experimental spectra is near perfect, both for the guided modes visible for wavelengths below 650nm, and the Bloch mode visible at just under 800nm. The BSW mode can be distinguished from the guided modes by inspection of the dispersion relation of Figure 3.27(a). The two guided modes intercepting the prism dispersion light line at around $K_x = 0.0135\text{nm}^{-1}$ and $K_x = 0.0108\text{nm}^{-1}$, corresponding to wavelengths $\lambda = 2\pi/K_x$ of 465nm and 580nm, accurately matching the reflectance spectra of both experimental and simulation data.

The BSW intersects the prism dispersion line at $K_x = 0.0078 \text{ nm}^{-1}$, corresponding to a wavelength of 805nm, also matching both the reflectance spectra of Figure 3.26. The other guided modes shown in Figure 3.27(a) are not experimentally accessible due range limitation of the used spectrometers.

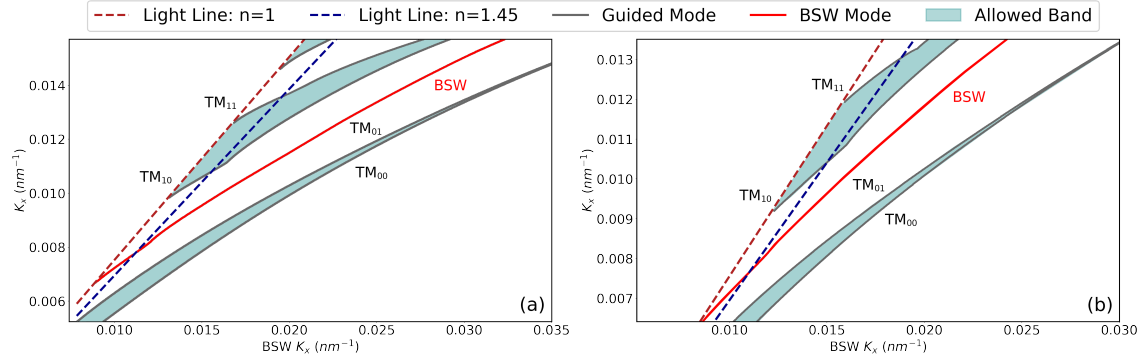


FIGURE 3.27: Dispersion relation of fabricated PhCs: (a) $2 \times 170 \text{ nm TiO}_2/90 \text{ nm SiO}_2 + 20 \text{ nm Fe}/80 \text{ nm TiO}_2$ and (b) $2 \times 170 \text{ nm TiO}_2/150 \text{ nm SiO}_2 + 30 \text{ nm Fe}/50 \text{ nm TiO}_2$.

It is noticeable that the red shift verified in all SPR experimental spectra is drastically reduced. This is likely related to the bigger thicknesses of the deposited structure, that makes errors in the deposited thickness less significant, and mainly to the lower sensitivity of resonance frequency of BSWs to deviations in the effective index of the structure, in which layer thickness takes part in. The magneto-optical behavior of the simulated structure slightly differs from the experimental one. The obtained MOBSW curves are plotted in Figure 3.28. For the first characterized refractive index, the experimental results match that of the simulated, both in magnitude of MOBSW signal. For the following refractive indices, the experimental bands follow a similar profile, while the simulated bands appear to undergo an inversion of sign, leading to a huge variation of MOBSW signal for varying refractive indices, that results in high sensitivities to refractive index when compared to experimental ones, as shown in Figure 3.29.

This diverging behavior between simulation and experimental spectra is a direct result of the different spectral dependencies of the off-diagonal elements of the simulated structure to those of the fabricated ones.

The same effect occurred for the other fabricated sample, $2 \times 170 \text{ nm TiO}_2/150 \text{ nm SiO}_2 + 30 \text{ nm Fe}/80 \text{ nm TiO}_2$, whose reflectance and magneto-optical response spectra are plotted in Figure 3.30 and Figure 3.31, respectively. The reflectance spectra matches very well the simulated one and in fair accordance to the dispersion relation of Figure 3.27(b),

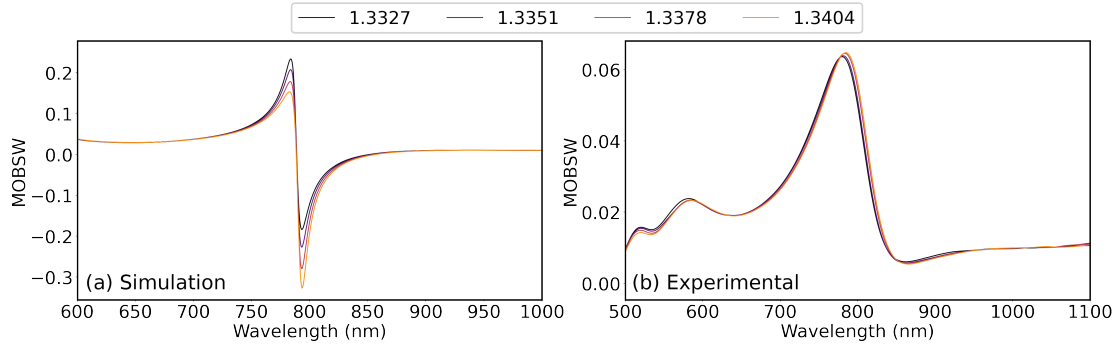


FIGURE 3.28: MOBSW spectra of (a) simulated and (b) experimental structure of $2 \times 170\text{nm TiO}_2/90\text{nm SiO}_2 + 20\text{nm Fe}/80\text{nm TiO}_2$.

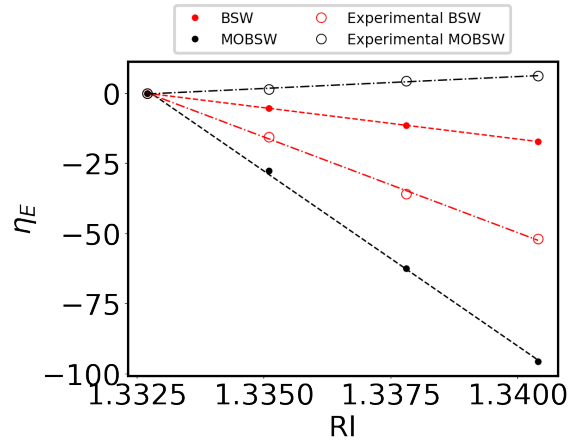


FIGURE 3.29: Simulated and experimental sensitivities adjusted for noise figure of a $2 \times 170\text{nm TiO}_2/90\text{nm SiO}_2 + 20\text{nm Fe}/80\text{nm TiO}_2$ PhC.

with guided modes at $K_x = 0.01025\text{nm}^{-1}$ and $K_x = 0.013\text{nm}^{-1}$, corresponding to wavelengths of 615nm and 480nm, and the BSW mode at $K_x = 0.0075\text{nm}^{-1}$, corresponding to wavelength of 840nm. Although the BSW mode appears to be slightly wider, leading to wider MOBSW bands, the amplitude of the MOBSW signal is very similar to the simulated bands and with increased sensitivity from the former structure, although not enough to achieve a higher sensitivity than the achieved for conventional BSW spectrum (Figure 3.32). All the parameters involved in the determination of the noise figure adjusted reflectance sensitivities to refractive index are shown in Table 3.4 and Table 3.5, for BSW and MOBSW spectra, respectively.

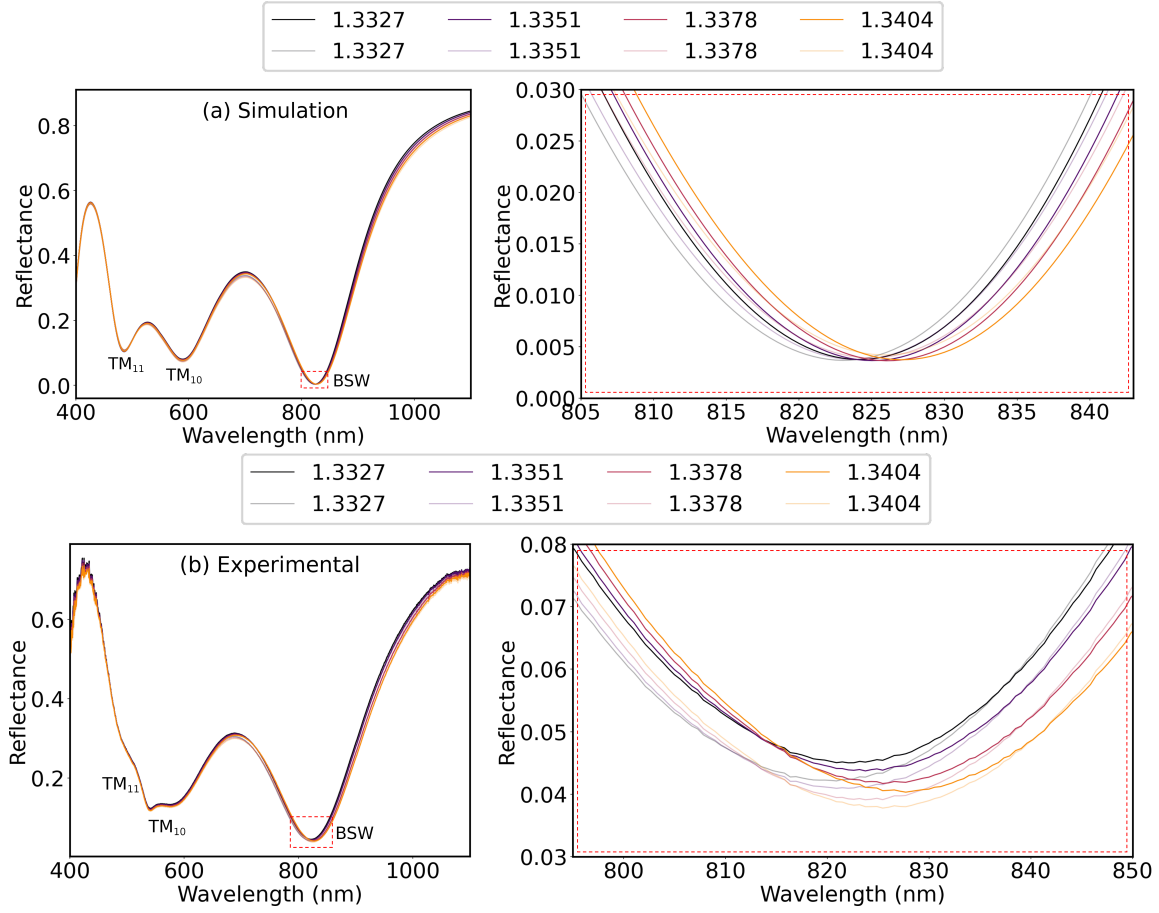


FIGURE 3.30: Reflectance spectra of (a) simulated and (b) experimental structure of $2 \times 170\text{nm TiO}_2/150\text{nm SiO}_2 + 30\text{nm Fe}/50\text{nm TiO}_2$.

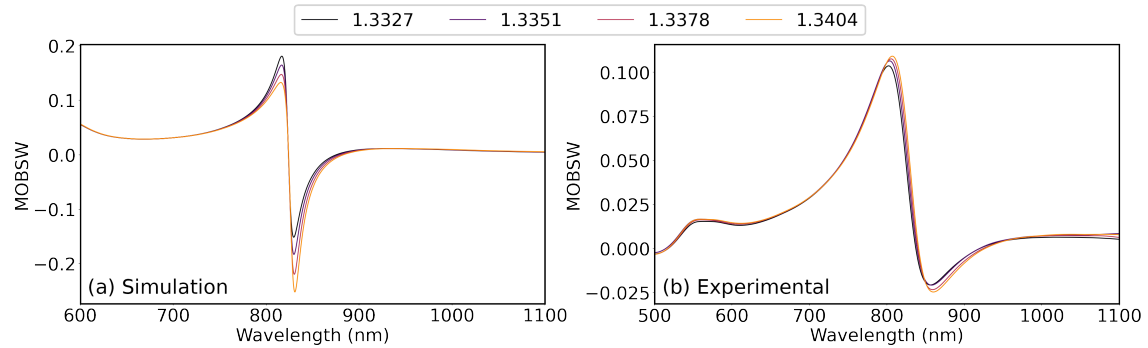


FIGURE 3.31: MOBSW spectra of (a) simulated and (b) experimental structure of $2 \times 170\text{nm TiO}_2/150\text{nm SiO}_2 + 30\text{nm Fe}/50\text{nm TiO}_2$.

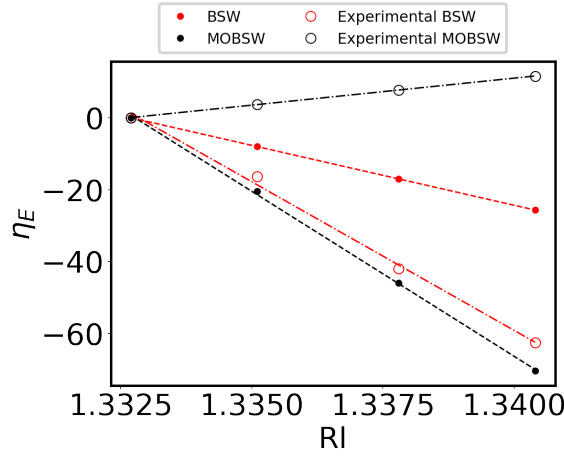


FIGURE 3.32: Simulated and experimental sensitivities adjusted for noise figure of a $2 \times 170\text{nm TiO}_2/150\text{nm SiO}_2 + 30\text{nm Fe}/50\text{nm TiO}_2$ PhC.

TABLE 3.4: Simulation and experimental parameters for reflectance sensitivity determination for BSW spectra of all fabricated structures.

BSW						
Matrix	Structure	η_{sim}	η_{exp}	$S_{min_{exp}}$	$\eta_{E_{sim}}$	$\eta_{E_{exp}}$
Planar	$2 \times 170\text{TiO}_2/90\text{SiO}_2+20\text{Fe}/80\text{TiO}_2$	-3.4	-4.2	0.00062	-2236	-6822
	$2 \times 170\text{TiO}_2/150\text{SiO}_2+30\text{Fe}/50\text{TiO}_2$	-5.0	-5.2	0.00063	-3333	-8272

TABLE 3.5: Simulation and experimental parameters for reflectance sensitivity determination for MOBSW spectra of all fabricated structures.

MOBSW						
Matrix	Structure	η_{sim}	η_{exp}	$S_{min_{exp}}$	$\eta_{E_{sim}}$	$\eta_{E_{exp}}$
Planar	$2 \times 170\text{TiO}_2/90\text{SiO}_2+20\text{Fe}/80\text{TiO}_2$	-18.7	1.0	0.00119	-12482	848
	$2 \times 170\text{TiO}_2/150\text{SiO}_2+30\text{Fe}/50\text{TiO}_2$	-13.8	3.1	0.00204	-9181	1502

An overall relative success was achieved for the simulation of magneto-optical surface waves in this work, although some struggles persist, limiting the enhancement of RI sensing by relative change in reflectance measurements for a magneto-optical modulation. Since magneto-optical effects are of such small magnitudes, any slight deviation of experimental reflectance spectra to simulated ones is enough to result in imprecise magneto-optical behavior of the experimental structures. Part of the difficulty lies in the

own nature of the MOSPR signal: being defined as in 1.53, this signal can be interpreted as a first derivative of the original SPR spectrum, since the magneto-optical modulation merely results in an infinitesimal change in SPR spectra. This way, minor variations in the original SPR signal get amplified, resulting in a more noisy MOSPR signal, that halts the experimental sensitivity of fabricated structures. The use of spectral interrogation in this work, with broadband light sources and spectrometers ranging from the near ultraviolet to the near infrared introduces spectral noise dependencies, as the noise figure of any broadband instrument is not constant throughout its entire range, and is usually more noisy near the edges of available interrogation. This causes that, depending on the fabricated structure, the resonance wavelength of enhanced magneto-optical activity will change, and the noise figure for a given measurement will differ, making it harder to accurately simulate experimental sensitivities as defined by 3.1. Some questions regarding the reproducibility of results can also be raised, since the same structure can attain different sensitivities for interrogation with different equipment.

Despite this, the spectral interrogation of ESW allows a better visualization of spectral dependencies in optical and magneto-optical effects.

A precise characterization of the optical constants of resourced materials is also of extreme importance. Only with precise data on the spectral permittivities of non-magnetic media – and particularly the off-diagonal permittivities of the dielectric tensor of magnetic media – is it possible to accurately simulate the magneto-optical response of any structure. In that regard, the characterization of the optical constants of materials by ellipsometry would be extremely helpful. This problem is particularly prevalent in this work in the characterization of magnetoplasmonic films of very thin layers of both Ag, Au and Fe. Even the Au thin films that appeared to show a good match between simulation and experimental spectra in the 50nm film in 3.2, began to diverge for the magnetoplasmonic films in 3.3.1. Although the dimension of the impact of the Fe layer in the observed red shift is uncertain, the dependence of the optical constants and plasmonic properties of Au thin films with thickness has been reported [34]. The case of Ag is also documented, with explicit dependence [35], [36]. As for the Fe layer, not only the optical constants but also the magnetic properties are dependent on the thickness and structural composition of thin films [33],[37]. The deposited Fe films by RF magnetron sputtering were mostly amorphous, while it has been reported on the effects of a more ordered, crystalline structure on the enhancement of the magnetic properties of Fe thin films [38], [39]. However,

thermal annealing of the magnetoplasmonic structures was not viable due to the coexistence with Ag or Au thin films that undergo dewetting at lower temperatures than those required for Fe thin film annealing [40].

Ultimately, the developed simulation routine is capable of accurately simulating reflectance bands of SPRs and BSWs, particularly for structures of higher layer thicknesses. The simulation of BSWs spectra was consistently more precise, something that can be justified by the fact that PhCs are composed of stable dielectric layers, whose optical constants do not vary significantly due to oxidation, structural composition or thickness. The fact that PhCs are overall less sensitive to variations of external conditions also helps contain experimental deviations to the simulation predicted results.

The simulation of magneto-optical effects in ESWs, however, is much more sensitive to erratic optical constants and off-diagonal elements, in the case of magnetic materials. Nonetheless, relative success was achieved for some configurations, validating the previewed enhanced of reflectance sensitivity to refractive index variation in the 15nm Ag/12.5nm Fe structure.

Chapter 4

Conclusions

A theoretical introduction on the fundamentals of ESWs, magneto-optics, and properties of their conjugated behavior and applications is presented. Two kinds of ESWs are studied: SPPs, generated at metal-dielectric interfaces, and BSWs, triggered at the interface between a PhC and a dielectric. The enhancement of magneto-optical effects by surface wave excitations led to a thorough simulation study based on Berreman's 4×4 matrix formalism to simulate reflectance spectra of optimized magnetophotonic structures. The spectra of relative reflected intensity for opposite magnetizations is simulated for structures of different geometries, and configurations of enhanced reflectance sensitivity to refractive index, compared to the non-modulated signal, are unveiled.

These structures were fabricated by RF magnetron sputtering deposition and characterized for external media of different refractive indices in a Kretschmann configuration setup, with two induction coils capable of generating controlled transverse magnetic fields for magneto-optical modulation of the ESWs. The optimization and calibration of the thin film deposition system is described, enabling fabrication of uniform films of more controlled thicknesses. This calibration was successful, given the good match between simulated and experimental spectra for SPPs and BSWs. In plasmonic structures, either in planar or multimode fiber substrate, a persistent shift in resonance wavelength is verified and is a result of both contributions from thickness deposition errors, given that the calibration from SEM pictures is made for films of much larger thicknesses, and differences between the optical constants of referenced samples and those fabricated in the laboratory. These differences in optical constants are in part due to their dependence in film thickness for very thin films.

The magneto-optical modulation rises from the magnetization of a Fe layer by the incident of a transverse magnetic field. The induction coils were designed to be identical and with dimensions that maximize the uniformity of magnetic field lines in the central region between them, where the fabricated thin films sit for characterization. This setup optimization ensures that only contributions from transverse magnetic fields are present, and an accurate simulation of magneto-optical effects is attained. In this regard, the experimental reflectance spectra for opposite magnetization modulation are perfectly comparable to those obtained by simulation, although the magnitude of simulated effects is overall larger. Again, this difference is related to differences in the referenced off-diagonal permittivity data, and the actual effects induced in the fabricated samples. The thickness dependency of the films is also a factor in this deviation, given that the magnetization of Fe has been reported to drastically increase with thickness for films under 50nm . The structural properties of Fe films are also relevant for magneto-optical properties, and the dominant amorphous composition of sputtering deposited Fe thin films may be halting the expression of magneto-optical effects. The grain size and the interface uniformity are also important. A larger grain size in Ag films grants better electrical conductivity, that helps SPP propagation at the interface.

The simulation study of thin film configurations for increased magneto-optical refractive index sensitivity led to the selection of only two geometries for magnetoplasmonic structures (Au/Fe/Au and Ag/Fe) and a single one for magnetophotonic crystals ($2 \times \text{TiO}_2/\text{SiO}_2 + \text{Fe}/\text{TiO}_2$). All of the fabricated structures showed inferior MOSPR sensitivity than in the simulations, and only one (15nm Ag/ 12.5nm Fe) achieved higher sensitivity for the MOSPR signal than for the conventional SPR spectrum. Such difference of results is caused mainly by the different spectral dependencies of the off-diagonal elements applied in the simulations and those present in the fabricated structures. Since magneto-optical modulations are relatively small, any minor deviation from the simulated behavior will lead to significant differences of MOSPR signal in the experimental signal.

A multimode fiber was also deposited with a magnetoplasmonic structure, and despite not achieving the simulated result for index sensitivity, a fair match in SPR and MOSPR spectra was obtained, validating the simulation work on magneto-optical effects in magnetoplasmonic structures in fiber, that hasn't yet been explored. As for the case of magnetophotonic crystals, only two films were deposited in planar substrate, and despite neither showing increased sensitivity by characterization of the MOBSW signal, an

accurate description of the magneto-optical signal was possible.

The developed simulation work has been demonstrated to return accurate results and be a good tool for the study of magneto-optical effects in layered media. Improvements on the simulation work should be possible by the characterization of the optical and magneto-optical constants of the fabricated samples, by means of ellipsometry measurements. For the fabricated planar structures, a recurrent band broadening (relative to simulation results) was noticed. Such peak broadening may be related to a poor estimate of the angular dispersion of light incident on the film after reflection into the prism, that may be slightly higher than the 2° applied in the simulations. The average 74° angle was chosen for optimization of spectral refractometric sensitivity to refractive index variations, and is not optimized for magneto-optical dependence on angle. The implementation of an experimental setup for variable incident angles could provide a versatile solution for optimization of different parameters.

As of now, the fabricated samples are not optimized for sensing applications. The use of Fe as a magnetic material has the obvious advantage of being widely accessible and showing strong magnetic properties. Other transition ferromagnetic metals either show weaker magnetic behavior (Ni) for the same optical losses, or are less accessible and potentially toxic (Co). Fe, however, presents huge optical losses that damp and widen plasmonic and Bloch bands, which is not ideal for optical sensing applications. Fe is also prone to oxidation particularly environments with high humidity, making its use not suitable for refractive index sensing in aqueous solutions. For these reasons, the use of dielectric ferromagnets has already been studied to some extent, although these garnets are not nearly as accessible and can be challenging to produce, while their magnetic properties tend to be fainter than Fe. As such, Fe is not obsolete for magneto-optical applications, and an optimization of the magnetic properties of the deposited thin films, its application in magnetic nanoparticles for Localized Surface Plasmon Resonances (LSPRs), or other vaguely unexplored ESWs, such as Tamm polaritons, are still open routes for research.

Appendix A

Berreman's Matrix Entries

$$\begin{aligned} S_{11} &= \Delta_{21} = M_{11} + M_{13}a_{31} + M_{16}a_{61} \\ S_{12} &= \Delta_{23} = M_{12} + M_{13}a_{32} + M_{16}a_{62} \\ S_{13} &= -\Delta_{24} = M_{14} + M_{13}a_{34} + M_{16}a_{64} \\ S_{14} &= \Delta_{22} = M_{15} + M_{13}a_{35} + M_{16}a_{65} \\ S_{21} &= \Delta_{41} = M_{21} + M_{23}a_{31} + (M_{26} - c\tilde{\zeta}/\omega) a_{61} \\ S_{22} &= \Delta_{43} = M_{22} + M_{23}a_{32} + (M_{26} - c\tilde{\zeta}/\omega) a_{62} \\ S_{23} &= -\Delta_{44} = M_{24} + M_{23}a_{34} + (M_{26} - c\tilde{\zeta}/\omega) a_{64} \\ S_{24} &= \Delta_{42} = M_{25} + M_{23}a_{35} + (M_{26} - c\tilde{\zeta}/\omega) a_{65} \\ S_{31} &= -\Delta_{31} = M_{41} + M_{43}a_{31} + M_{46}a_{61} \\ S_{32} &= -\Delta_{33} = M_{42} + M_{43}a_{32} + M_{46}a_{62} \\ S_{33} &= \Delta_{34} = M_{44} + M_{43}a_{34} + M_{46}a_{64} \\ S_{34} &= -\Delta_{32} = M_{45} + M_{43}a_{35} + M_{46}a_{65} \\ S_{41} &= \Delta_{11} = M_{51} + (M_{53} + c\tilde{\zeta}/\omega) a_{31} + M_{56}a_{61} \\ S_{42} &= \Delta_{13} = M_{52} + (M_{53} + c\tilde{\zeta}/\omega) a_{32} + M_{56}a_{62} \\ S_{43} &= -\Delta_{14} = M_{54} + (M_{53} + c\tilde{\zeta}/\omega) a_{34} + M_{56}a_{64} \\ S_{44} &= \Delta_{12} = M_{55} + (M_{53} + c\tilde{\zeta}/\omega) a_{35} + M_{56}a_{65} \end{aligned} \tag{A.1}$$

$$\begin{aligned}
a_{31} &= (M_{61}M_{36} - M_{31}M_{66}) / d, \\
a_{32} &= [(M_{62} - c\tilde{\xi}/\omega) M_{36} - M_{32}M_{66}] / d, \\
a_{34} &= (M_{64}M_{36} - M_{34}M_{66}) / d, \\
a_{35} &= [M_{65}M_{36} - (M_{35} + c\tilde{\xi}/\omega) M_{66}] / d, \\
a_{61} &= (M_{63}M_{31} - M_{33}M_{61}) / d, \\
a_{62} &= [M_{63}M_{32} - M_{33} (M_{62} - c\tilde{\xi}/\omega)] / d, \\
a_{64} &= (M_{63}M_{34} - M_{33}M_{64}) / d, \\
a_{65} &= [M_{63} (M_{35} + c\tilde{\xi}/\omega) - M_{33}M_{65}] / d, \\
d &= M_{33}M_{66} - M_{36}M_{63}
\end{aligned} \tag{A.2}$$

(A.3)

Appendix B

Reflection and Transmission Coefficients at Non-Magnetic/Magnetic Interfaces

$$r_{ss} = r_{ss}^0 \quad (\text{B.1})$$

$$r_{pp} = r_{pp}^0 (1 + i\rho_p) \quad (\text{B.2})$$

$$\rho_p = \frac{n_1 r_{12}^p Q \sin \phi}{2g_2} \quad (\text{B.3})$$

$$r_{sp} = i t_{12}^s t_{21}^p (\cos \alpha + m \cos \beta) \delta_1 \quad (\text{B.4})$$

$$r_{ps} = i t_{12}^s t_{21}^p (\cos \alpha - m \cos \beta) \delta_1 \quad (\text{B.5})$$

$$t_{ss} = t_{ss}^0 \quad (\text{B.6})$$

$$t_{pp} = t_{pp}^0 (1 + i\tau_p) \quad (\text{B.7})$$

$$\tau_p = (1 + r_{21}^p) \frac{n_1 Q \sin \phi}{2g_2} \quad (\text{B.8})$$

$$t_{sp} = i t_{12}^p r_{21}^s (\delta_1 \cos \alpha + m \cos \beta) = \frac{t_{12}^p r_{21}^s}{t_{12}^s r_{21}^p} r_{sp} \quad (\text{B.9})$$

$$t_{ps} = i t_{12}^p r_{21}^s (\delta_1 \cos \alpha - m \cos \beta) = \frac{t_{12}^p r_{21}^s}{t_{12}^s r_{21}^p} r_{ps} \quad (\text{B.10})$$

where the Fresnel coefficients for s and p polarized waves at an interface between a non-magnetic (1) and magnetic of complex refractive indexes n_i and n_j are known to be equal to

$$r_{j,k}^s = \frac{g_j - g_k}{g_j + g_k} \quad r_{j,k}^p = \frac{g_j n_k^2 - g_k n_j^2}{g_j n_k^2 + g_k n_j^2} \quad (\text{B.11})$$

$$t_{j,k}^s = \frac{2g_j}{g_j + g_k} \quad t_{j,k}^p = \frac{2g_j n_j n_k}{g_j n_k^2 + g_k n_j^2} \quad (\text{B.12})$$

$$g_j = \sqrt{n_j^2 - n_1^2 \sin^2 \phi} \quad g_1 = n_1 \cos \phi \quad (\text{B.13})$$

The parameter δ_1 relates to magneto-optical properties of the interface and is proportional to Q , and m is given by

$$\delta_1 = \frac{n_2}{4g_2} Q \quad m = \frac{n_1}{g_2} \sin \phi \quad (\text{B.14})$$

Appendix C

Spectral Reflectance Sensitivities to Refractive Index Variation

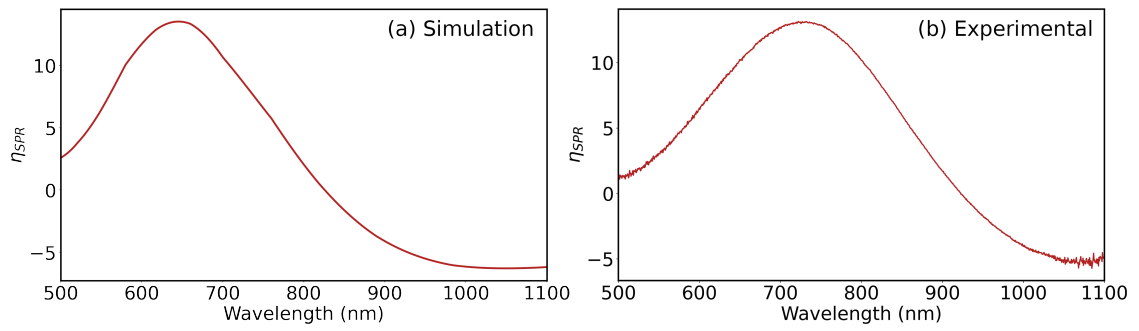


FIGURE C.1: η_{SPR} spectra of (a) simulated and (b) experimental films of 10nm Au/15nm Fe/10nm Au.

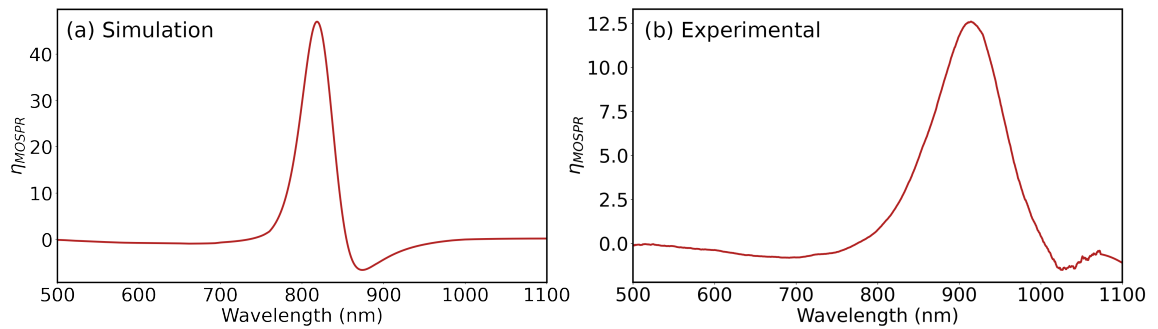


FIGURE C.2: η_{MOSPR} spectra of (a) simulated and (b) experimental films of 10nm Au/15nm Fe/10nm Au.

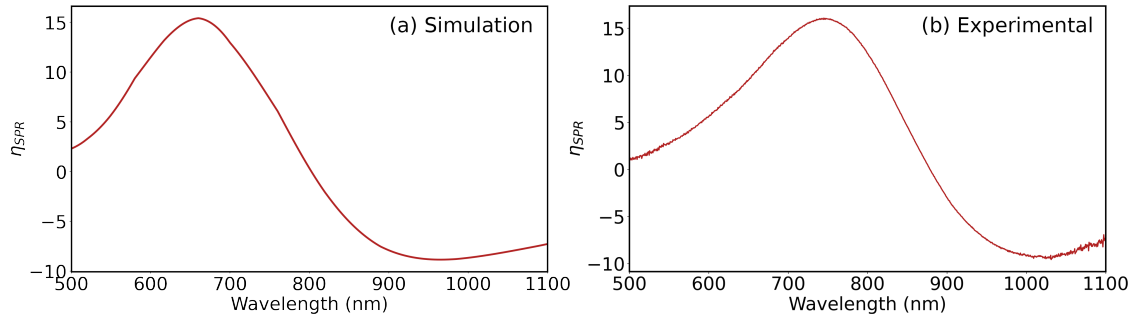


FIGURE C.3: η_{SPR} spectra of (a) simulated and (b) experimental films of 15nm Au/12.5nm Fe/10nm Au.

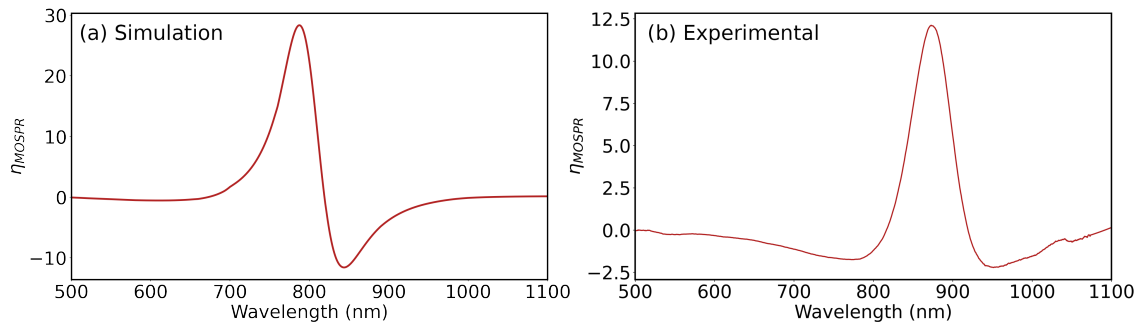


FIGURE C.4: η_{MOSPR} spectra of (a) simulated and (b) experimental films of 15nm Au/12.5nm Fe/10nm Au.

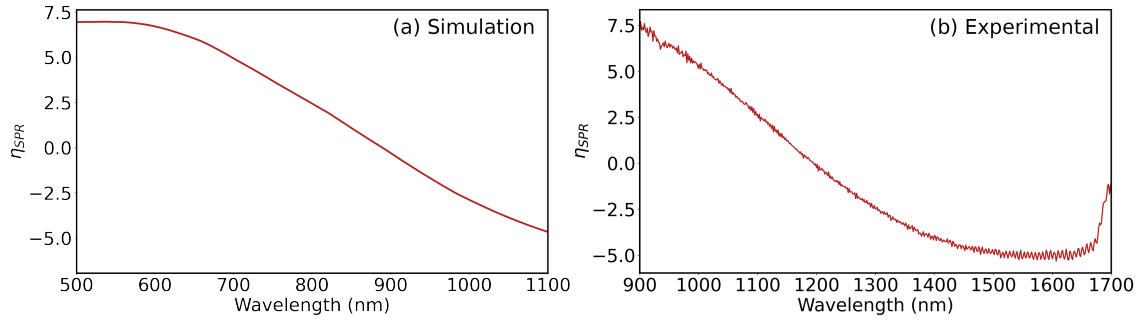


FIGURE C.5: η_{SPR} spectra of (a) simulated and (b) experimental films of 15nm Ag/10nm Fe.

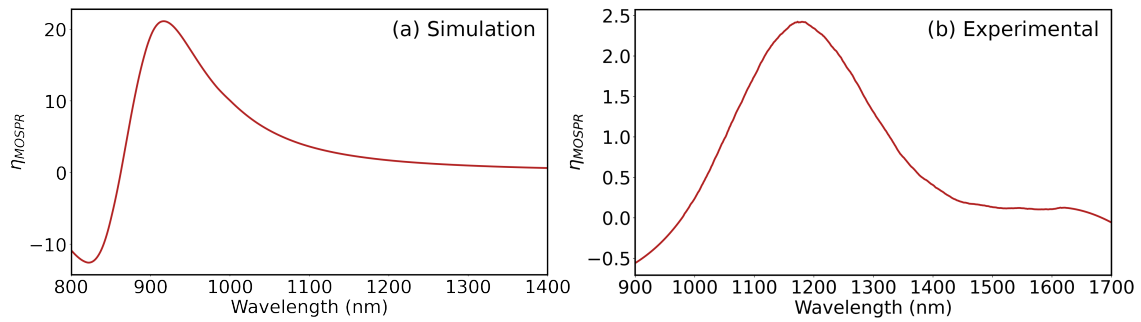


FIGURE C.6: η_{MOSPR} spectra of (a) simulated and (b) experimental films of 15nm Ag/10nm Fe.

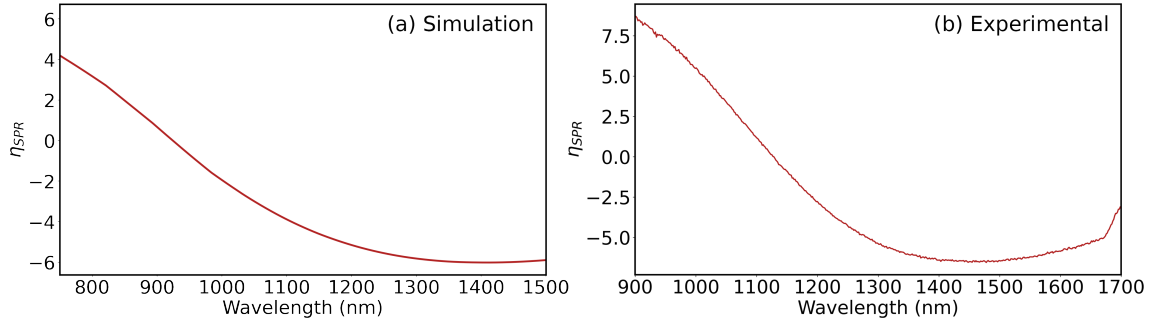


FIGURE C.7: η_{SPR} spectra of (a) simulated and (b) experimental films of 15nm Ag/12.5nm Fe.

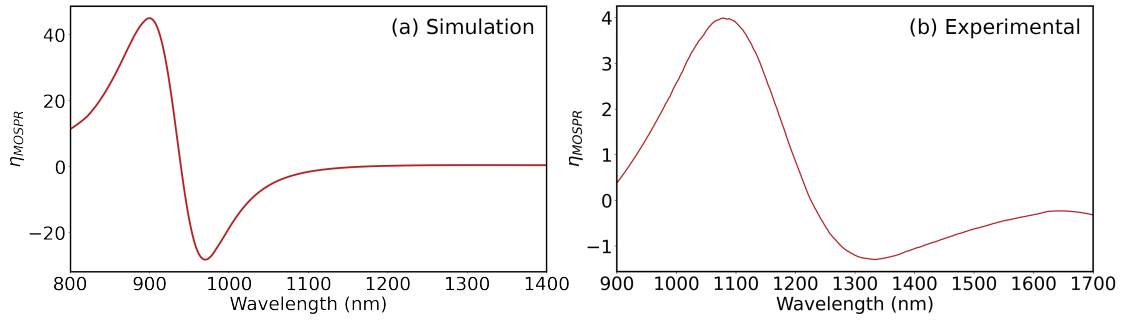


FIGURE C.8: η_{MOSPR} spectra of (a) simulated and (b) experimental films of 15nm Ag/12.5nm Fe.

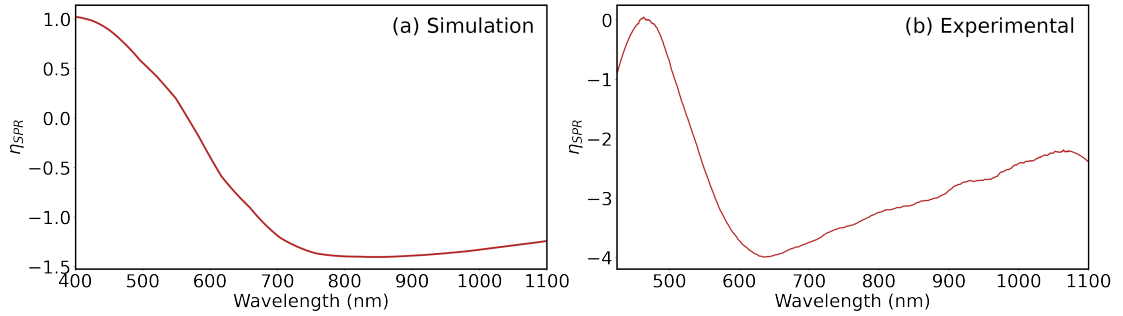


FIGURE C.9: η_{SPR} spectra of (a) simulated and (b) experimental films of 15nm Ag/12.5nm Fe in multimode fiber.

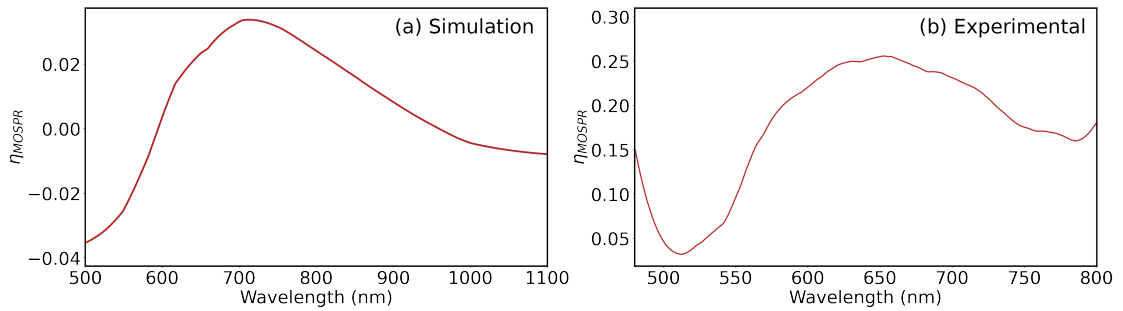


FIGURE C.10: η_{MOSPR} spectra of (a) simulated and (b) experimental films of 15nm Ag/12.5nm Fe in multimode fiber.

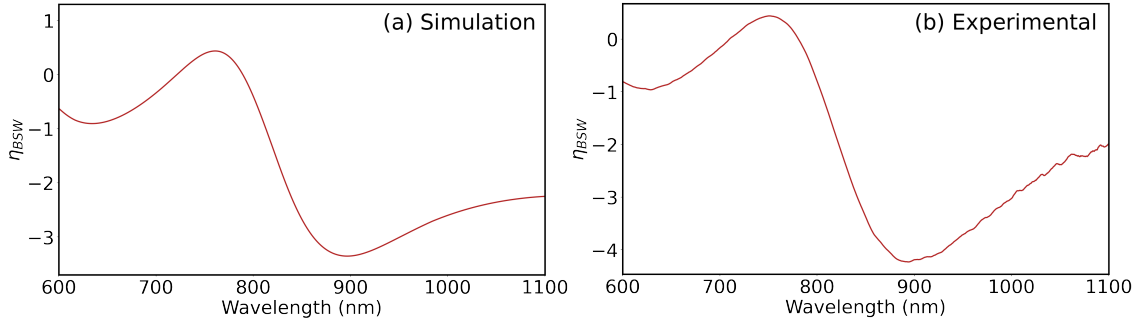


FIGURE C.11: η_{BSW} spectra of (a) simulated and (b) experimental PhC of $2 \times 170\text{nm}$ $\text{TiO}_2/90\text{nm SiO}_2 + 20\text{nm Fe}/80\text{nm TiO}_2$.

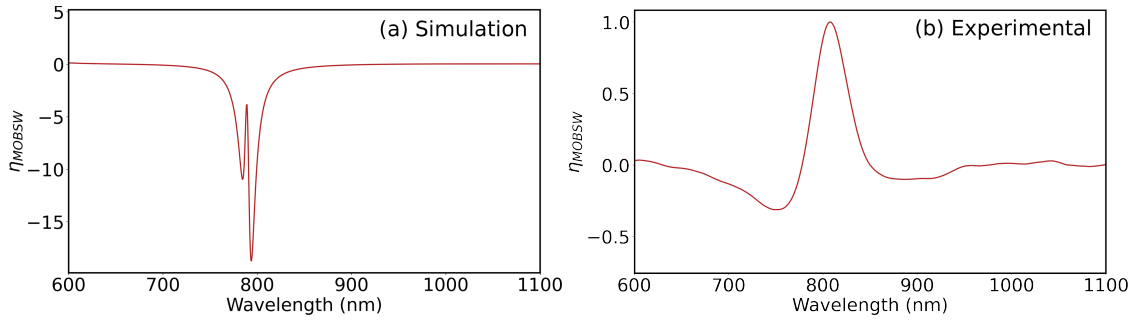


FIGURE C.12: η_{MOBSW} spectra of (a) simulated and (b) experimental PhC of $2 \times 170\text{nm}$ $\text{TiO}_2/90\text{nm SiO}_2 + 20\text{nm Fe}/80\text{nm TiO}_2$.

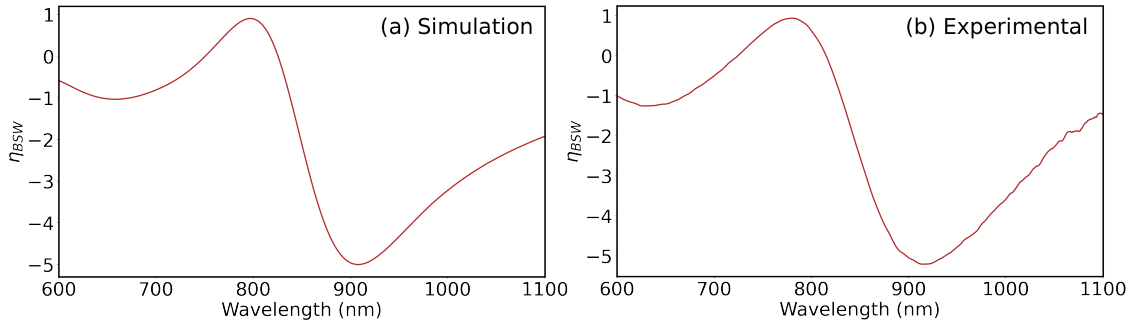


FIGURE C.13: η_{BSW} spectra of (a) simulated and (b) experimental PhC of $2 \times 170\text{nm}$ $\text{TiO}_2/150\text{nm SiO}_2 + 30\text{nm Fe}/50\text{nm TiO}_2$.

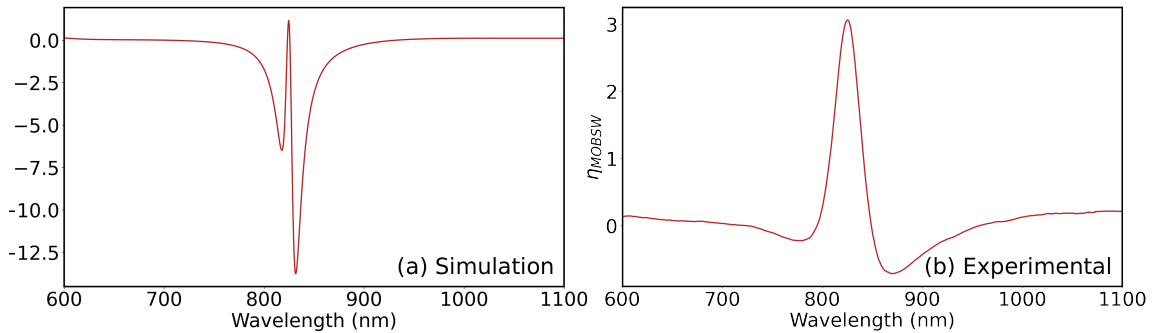


FIGURE C.14: η_{MOBSW} spectra of (a) simulated and (b) experimental PhC of $2 \times 170\text{nm}$ $\text{TiO}_2/150\text{nm SiO}_2 + 30\text{nm Fe}/50\text{nm TiO}_2$.

Appendix D

Magnetic field profile parameters

$$(D.1) \quad \begin{aligned} Q_{pp} &= \frac{w}{2} + (x + \frac{r}{2}) \\ Q_{np} &= \frac{w}{2} - (x + \frac{r}{2}) \\ Q_{pn} &= \frac{w}{2} + (x - \frac{r}{2}) \\ Q_{nn} &= \frac{w}{2} - (x - \frac{r}{2}) \end{aligned}$$

$$(D.2) \quad \begin{aligned} K_{pp} &= \frac{D + \sqrt{D^2 + 4Q_{pp}^2}}{d + \sqrt{d^2 + 4Q_{pp}^2}} \\ K_{np} &= \frac{D + \sqrt{D^2 + 4Q_{np}^2}}{d + \sqrt{d^2 + 4Q_{np}^2}} \\ K_{pn} &= \frac{D + \sqrt{D^2 + 4Q_{pn}^2}}{d + \sqrt{d^2 + 4Q_{pn}^2}} \\ K_{nn} &= \frac{D + \sqrt{D^2 + 4Q_{nn}^2}}{d + \sqrt{d^2 + 4Q_{nn}^2}} \end{aligned}$$

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