Universidad Autónoma de Madrid
Facultad de Ciencias
Departamento de Física de Materiales

MagnetoPlasmónica.
MagnetoÓptica en Sistemas Plasmónicos

Memoria de tesis para optar al grado de Doctor en Ciencias Físicas presentada por:

Juan B. González Díaz

Y dirigida por:

el Dr. Antonio García Martín y el Prof. Gaspar Armelles

Instituto de Microelectrónica de Madrid
Centro Nacional de Microelectrónica
Consejo Superior de Investigaciones Científicas

Tres Cantos (2010)
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Tres Cantos (2010)
A mi familia, lo mejor de este mundo.
- Dios, Buckman!! Esto lleva aquí desde la guerra de Corea!
- Y qué, señor? Sigue sabiendo a puré de patata...
- Pues que es... jamón en lata!!

(Uno que yo me sé...)

“Partió de Jamaica, rumbo a Nueva York, un barco velero, un barco velero, cargado de Ron (cargado de Ron)”

(Maki a vela)

“Los cacahuetes con cáscara están más que buenos”

(Clavada?)
Resumen

El objetivo principal de esta tesis es el estudio de la fenomenología asociada a nanoestructuras que presentan plasmones superficiales y actividad magnetoóptica al mismo tiempo. Estas estructuras tienen un gran potencial de aplicación en una amplia gama de áreas científicas e industriales. En particular, pueden devenir como elementos muy importantes en la sintonización de dispositivos nanoópticos basados en la resonancia de plasmon. Un ejemplo de esto ya es una realidad, donde una nueva rama de biosensores, están desarrollando dispositivos basados en la modulación magnetoóptica y cuya sensibilidad mejora.

Los materiales magnetoplasmónicos estudiados en esta tesis ofrecen la propiedad única de controlar sus propiedades de formas diferentes, ya que su respuesta óptica se ve dramáticamente afectada por la excitación de un plasmon, pero asimismo, la respuesta óptica puede ser sintonizada de forma activa por medio de una agente externo como es el campo magnético.

En esta tesis, nos centraremos principalmente en el primer aspecto, donde los diferentes materiales y estructuras considerados darán lugar a un aumento de la respuesta magnetoóptica debido a las modificaciones del campo electromagnético por al excitación de plasmones superficiales.

La tesis se estructura en 5 capítulos:

Capítulo 1: Donde se introduce la motivación de dicha tesis y una breve introducción a los plasmones superficiales.

Capítulo 2: En el cual se introducen los conceptos fundamentales del efecto magnetoóptico. Está dividido en dos secciones. La primera está dedicada a la descripción del efecto Kerr, mientras que la segunda detalla los montajes experimentales utilizados a lo largo de esta tesis.
Capítulos 3 y 4: En estos capítulos se estudia el efecto de los plasmones superficiales en la respuesta magnetooptica de un sistema nanoestructurado. Concretamente, el capítulo 3 estudia estos efectos para nanoestructuras compuestas exclusivamente por materiales ferromagnéticos. dos tipos de estructuras serán consideradas: hilos de níquel embebidos en una matriz de alúmina, y membranas de níquel perforadas. Ambas estructuras serán caracterizadas óptica y magnetoopticamente; además se encontrará un aumento de la actividad magnetooptica en la región espectral donde la excitación de plasmones superficiales tiene lugar. El capítulo 4 estudia este mismo efecto pero en este caso, materiales del tipo metales nobles son añadidos a las nanoestructuras. Esto es debido a la menor absorción que presentan estos materiales y que mejoran las propiedades plasmónicas.

Capítulo 5: Al contrario que en los capítulos 3 y 4, en este capítulo se estudian cómo los campos magnéticos afectan las propiedades de los plasmones superficiales. En particular, veremos la modificación del vector de onda del plasmon cuando un campo magnético es aplicado a sistemas compuestos por multicapas.

Por último, las conclusiones más relevantes extraídas de cada parte de la tesis serán expuestas al final de este manuscrito.
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Chapter 1

Preface

1.1 Aim of this Thesis

The main objective of the thesis is to study the phenomenology associated to nanostructures possessing surface plasmons and magneto-optical activity, the so-called magnetoplasmonic structures. These structures will be applicable in a broad spectrum of research and industrial areas. In particular, they are important elements in addressing tunability capabilities in plasmon-based nano-optical device. Also a new branch of biosensors with enhanced sensitivity are based on magneto-optical modulation.

The magneto-plasmonic materials studied along this thesis offer the unique ability to control their properties in more than one way, since the magneto-optical response will be dramatically affected by the excitation of a plasmon, but also, their optical response can be actively tuned by means of an external agent: a magnetic field.

We will mainly concentrate on the first aspect, where the different materials and structures considered will lead to the enhancement of the magneto-optical response due to the modifications of the electromagnetic field induced by plasmon resonances.
In what follows, we will briefly describe the surface plasmon polaritons, pointing out their main characteristics. Since we are going to mainly analyze the magneto-optical response, we will devote Chapter 2 to describe in detail the different magneto-optical configurations and experimental setups.

1.2 Surface Plasmon Polaritons

Surface plasmons are essentially light waves that are trapped on the interface between a metal and a dielectric because of their interaction with the free electrons of the conductor (strictly speaking, they should be called surface plasmon polaritons to reflect this hybrid nature). In this interaction, the free electrons respond collectively by oscillating in resonance with the light wave. The resonant interaction between the surface charge oscillation and the electromagnetic field of the light constitutes the SP and gives rise to its unique properties.

Nevertheless, the interest in plasmons is not that new. Well before scientists set out to study the unique properties of surface plasmons, they were employed by artists to generate vibrant colors in glass artifacts and in the staining of church windows. One of the most famous examples is the Lycurgus cup dating back to the Byzantine Empire (4th century AD). Some of the first scientific studies in which surface plasmons were observed date back to the beginning of the twentieth century. In the year 1902 Prof. Robert W. Wood observes unexplained features in optical reflection measurements on metallic gratings [1]. Around that same time, in 1904, Maxwell Garnett describes the bright colors observed in metal doped glasses [2] using the then newly developed Drude theory of metals, and the electromagnetic properties of small spheres as derived by Lord Rayleigh. In an effort to develop further understanding, in 1908 Gustav Mie develops his now widely used theory of light scattering by spherical particles [3].

![Figure 1.1 - Lycurgus cup illuminated from (a) the outside and (b) the inside. This is due to the excitation of surface plasmons in nanoscale gold particles that absorb light at 520 nm.](image)
Some fifty years later, in 1956, David Pines theoretically describes the characteristic energy losses experienced by fast electrons travelling through metals [4], and attributes these losses to collective oscillations of free electrons in the metal. In analogy to earlier work on plasma oscillations in gas discharges, he calls these oscillations “plasmons”. Coincidentally, in that same year Robert Fano introduces the term “polariton” for the coupled oscillation of bound electrons and light inside transparent media [5]. In 1957 a study is published by Rufus Ritchie on electron energy losses in thin films [6], in which it is shown that plasmon modes can exist near the surface of metals. This study represents the first theoretical description of surface plasmons. In 1968, nearly seventy years after Wood’s original observations, Ritchie and co-workers describe the anomalous behavior of metal gratings in terms of surface plasmon resonances excited on the gratings [7]. A major advance in the study of surface plasmons is made in 1968 when Andreas Otto as well as Erich Kretschmann and Heinz Raether present methods for the optical excitation of surface plasmons on metal films [8,9], making experiments on surface plasmons easily accessible to many researchers.

Since then, there has been a significant advance in both theoretical and experimental investigations of surface plasmons, which for researches in the field of condensed matter and surface physics have played a key role in the interpretation of a great variety of experiments and the understanding of various fundamental properties of solids. These include the nature of Van der Waals forces [10–12], the classical image potential acting between a point classical charge and a metal surface [13–16], the energy transfer in gas–surface interactions [17], surface energies [18–22], the damping of surface vibrational modes [23,24], the energy loss of charged particles moving outside a metal surface [25,26] and the de-excitation of adsorbed molecules [27].

1.3 State of the Art

Renewed interest in plasmonics comes from recent advances that allow metals to be structured and characterized on the nanometer scale. This in turn has enabled us to control surface plasmons properties to reveal new aspects of their underlying science and to tailor them for specific applications. For instance, plasmons are being explored for their potential in optics, magneto-optic data storage microscopy and solar cells, as well as being used to construct sensors for detecting biologically interesting molecules.

There are two types of surface plasmons, propagating and localized plasmons. The first ones take place when considering flat surfaces like those of continuous films. They are characterized as non-radiative propagating waves along the
metal/dielectric interface, whose propagation length is controlled by the properties of the metal and the dielectric.

![Diagram showing charge fluctuations at a metal-dielectric interface](image)

**Figure 1.2** - Sketch showing charge fluctuations at a metal-dielectric interface giving rise to (a) propagating surface plasmons in a continuous layer interface and (b) localized surface plasmons at a particle.

The second ones, occurring in isolated nanostructures like particles, do not propagate but are localized at the nanoparticle region (see figure 1b). This localized surface plasmon (LSP) excitation is evidenced in the optical absorption spectra by the presence of well-defined peaks. The spectral location of the LSP is sensitive to the shape, size, and composition of the nanostructure, as well as on the optical properties of the surrounding dielectric [28-30].

![Evolution of optical properties for Au-Ag alloy colloidal nanoparticles](image)

**Figure 1.3** - (a) Evolution of the optical properties for Au-Ag alloy colloidal nanoparticles with different Au content. It is worth noticing that this evolution is different from that observed in core-shell Au/Ag nanoparticles. (b) Idem but for Au-SiO core-shell nanoparticles.

Both kind of surface plasmons have been employed over the years in a wide spectrum of studies ranging from condensed matter and surface physics [10-27] to electrochemistry [31], wetting [32], biosensing [33-35], scanning tunneling microscopy [36], the ejection of ions from surfaces [37], nanoparticle growth [38, 39], surface-plasmon microscopy [40, 41] and surface plasmon resonance...
technology [42–49]. Renewed interest in surface plasmons has come from recent advances in the investigation of the optical properties of nanostructured materials [50, 51], one of the most attractive aspects of these collective excitations now being their use to concentrate light in sub-wavelength structures and to enhance transmission through periodic arrays of sub-wavelength holes in optically thick metallic films [52,53], as well as the possible fabrication of nanoscale photonic circuits operating at optical frequencies [54,55] and their use as mediators in the transfer of energy from donor to acceptor molecules on opposite sides of metal films [56].

However, to date most of the plasmon-based devices are passive, i.e. their optical properties cannot be tuned once they are fabricated. In order to find new applications it is necessary to add active functionalities to these plasmon-based devices, an area known as active plasmonics. The development of active plasmonics requires the use of materials or strategies in which the surface plasmon properties can be controlled by an external agent. So far, the feasibility of the realization of active plasmonic devices has been demonstrated by considering different control agents: temperature [57,58], electric field [59] or electromagnetic waves [60,61]. Another interesting candidate for being used as an external agent is the magnetic field.

1.4 Thesis Structure

This thesis consists in five chapters:

Chapter 1: This one in which the aim of the thesis and a very brief introduction to surface plasmons is given.

Chapter 2: A general introduction addressing the fundamental concepts of MagnetoOptics. It is divided into two major sections. The first section is devoted to the description of the MO Kerr effect. The second one details a description of the experimental characterization setups used in this thesis.

Chapters 3 and 4: In this chapters the excitation of plasmonic resonances affects the MO response of a nanostructured system will be studied. Chapter 3 is devoted to the study of nanostructures composed exclusively of ferromagnetic materials. Two types of nanostructures will be analyzed; nickel nanowires embedded in an alumina matrix, and perforated nickel membranes. Both structures will be optically and MO characterized, finding an enhancement of the MO activity at the spectral region of the plasmonic excitation. Chapter 4 involves the addition of noble metals into the nanostructures, since their plasmonic excitations are better defined due to the smaller absorption when compared with ferromagnetic metals. We will analyze the MO response in
nanostructures involving pure noble metals materials and combined noble metals and ferromagnet materials.

Chapter 5: A study on how magnetic fields affect the properties of surface plasmons will be performed. In particular, the modification of the plasmon wave vector when a magnetic field is applied to continuous layered systems will be studied. We will demonstrate that the modifications of the optical activity are due to the change in the plasmon wave vector when a magnetic field is applied.

Finally, the most relevant results derived from each part of the thesis are summarized in the conclusions. Moreover, the thesis is complemented with some appendixes detailing some technical aspects used along the thesis.
Chapter 2

Fundamentals of MagnetoOptics

2.1 The MagnetoOptical Kerr Effect

The Magneto-Optical (MO) effect consists in a change of the polarization state and/or intensity when light propagates or reflects from a magnetized material. It was first discovered by Michael Faraday in 1845. He observed a rotation of the polarization plane when linear polarized light went through a glass subjected to a magnetic field (parallel to the direction of the applied field). Some years later, John Kerr (1888) observed a similar change in the polarization when light reflected from an electromagnet [1]. This is the so-called MagnetoOptical (MO) Kerr effect.

Macroscopically, the MO (and optical) activity of all the materials can be described by means of the dielectric tensor. In absence of magnetic fields, this tensor is symmetric ($\varepsilon_{ij} = \varepsilon_{ji}$), but the application of a magnetic field destroys the symmetry [2] and makes this tensor to become asymmetric and dependent on the magnetic field applied ($\varepsilon_{ij}(H) = \varepsilon_{ji}(-H)$). It is expressed as:
\[ \varepsilon = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ -\varepsilon_{yx} & \varepsilon_{yy} & -\varepsilon_{yz} \\ -\varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{pmatrix} \]  

[2.1]

where \( \varepsilon_{ii} \) represent the optical properties of the material and \( \varepsilon_{ij}, \varepsilon_{ji} \) the MO properties. The dependence of the different elements of the dielectric tensor on the magnetic field can be described, up to second-order terms, as follows [3]:

\[ \varepsilon_{ij} = \varepsilon_{ij}^0 + \sum_k a_{ijk} H_k + \sum_k \sum_l b_{ijkl} H_k H_l + ... \]

[2.2]

being \( \varepsilon_{ij}^0 \) the dielectric constants for zero magnetic field, and \( a_{ijk}, b_{ijkl} \) the coefficients of a third and fourth rank order tensor responsible for the linear and quadratic dependence with the magnetic field \( H_i \). It must be noted that, for ferromagnetic metals, the different elements of the dielectric tensor depend on the material magnetization \( M_i \) instead of the magnetic field applied. Since the quadratic dependence is rather small, we will consider only the linear dependence with the magnetic field. Thus, if the material is isotropic the dielectric tensor adopts the form:

\[ \varepsilon = \begin{pmatrix} \varepsilon & aH_z & aH_y \\ -aH_z & \varepsilon & -aH_x \\ -aH_y & aH_x & \varepsilon \end{pmatrix} \]  

[2.3]

As it has been previously quoted the MO Kerr effect (MOKE) manifests as a change in the polarization of the reflected light. All the information related to this effect is summarized in the reflectivity matrix of the system under consideration. This matrix contains the Fresnel reflectivity coefficients and is defined as:

\[ S = \begin{pmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{pmatrix} \]  

[2.4]

Here \( r_{pp} \), and \( r_{ss} \) are the reflectivity coefficients of the p and s-components of light, whereas \( r_{ps} \) (and \( r_{sp} \)) represents the polarization conversion of p-polarized light into s-polarized light (and vice versa). For simple systems, the Fresnel coefficients can be easily calculated. Let us consider two semi-infinite non-magnetic and magnetic mediums. Introducing [2.1] for the magnetic medium, these coefficients are expressed as [4,5]:
\[ r_{pp} = \frac{n_1 \cos \theta_0 - n_0 \cos \theta_1 + 2n_0 n_1^{-1} \cos \theta_0 \sin \theta_1 \varepsilon_{xy}}{n_1 \cos \theta_0 + n_0 \cos \theta_1} \]  

\[ r_{sp} = \frac{-n_0 n_1^{-1} \cos \theta_0 \left( \varepsilon_{xy} \cos \theta_1 + \varepsilon_{yz} \sin \theta_1 \right)}{(n_1 \cos \theta_0 + n_0 \cos \theta_1)(n_0 \cos \theta_0 + n_1 \cos \theta_1) \cos \theta_1} \]  

\[ r_{ps} = \frac{-n_0 n_1^{-1} \cos \theta_0 \left( \varepsilon_{xy} \cos \theta_1 - \varepsilon_{xz} \sin \theta_1 \right)}{(n_1 \cos \theta_0 + n_0 \cos \theta_1)(n_0 \cos \theta_0 + n_1 \cos \theta_1) \cos \theta_1} \]  

\[ r_{ss} = \frac{n_0 \cos \theta_0 - n_1 \cos \theta_1}{n_0 \cos \theta_0 + n_1 \cos \theta_1} \]  

, where \( n_i = \sqrt{\varepsilon_i} \) represents the refractive index of the ith medium, and \( \theta_i \) accounts for the angle of light within the ith medium. As it can be observed, these coefficients are different depending on the direction of the magnetic field with respect to both the incident light and sample planes. In particular, there are three main configurations (shown in Figure 2.1a and b) in which the MOKE effect may be classified:

**Figure 2.1** - Different magnetic configurations of the Kerr effect (Left) Transverse configuration: if the magnetic field is applied parallel to the sample plane but perpendicular to the light incidence plane, the Kerr effect manifests as a change in the p-component of light. (Right) Polar and Longitudinal configurations: if the magnetic field is applied perpendicular to the sample plane (Polar) or parallel to both the sample and the incidence light plane (Longitudinal), the Kerr effect manifests as a change in the polarization plane and in the ellipticity state.
2.1.1 Transverse configuration \((\varepsilon_{xz} \neq 0, \varepsilon_{xy}, \varepsilon_{yz} = 0)\)

In this configuration, the magnetic field is applied parallel to the sample plane but perpendicular to the plane of incident light. The dielectric tensor becomes:

\[
\varepsilon = \begin{pmatrix}
\varepsilon & 0 & \varepsilon_{xz} \\
0 & \varepsilon & 0 \\
-\varepsilon_{xz} & 0 & \varepsilon
\end{pmatrix}
\] \hfill [2.6]

The transverse Kerr (TMOKE) effect manifests as a variation of the p-component of the reflected light. The s-component does not experiment any variation (since we do not consider quadratic contributions) and there is no conversion polarization from p to s or vice versa. As a result the reflectivity coefficients \(r_{ps}\) and \(r_{sp}\) are zero, and therefore the transverse configuration involves no change in polarization but a change in the light intensity of the p-polarized light.

\[
\begin{align*}
    r_{pp}(H) &= r_{pp}(0) + AH \\
    r_{ss}(H) &= r_{ss}(0) \\
    r_{sp}(H) &= r_{ps}(H) = 0
\end{align*}
\] \hfill [2.7a - 2.7c]

The quantification of the Transverse Kerr effect is performed analyzing the relative variation of the p-component of reflectivity, defined as:

\[
\text{TMOKE} = \frac{R_{pp}(H) - R_{pp}(-H)}{R_{pp}(H) + R_{pp}(-H)} \approx \frac{R_{pp}(H) - R_{pp}(-H)}{2R_{pp}(0)}
\] \hfill [2.8a]

\[
R_{pp}(0) = |r_{pp}(0)|^2
\] \hfill [2.8b]

\[
R_{pp}(\pm H) = |r_{pp}(\pm H)|^2 = |r_{pp}(0) \pm AH|^2
\] \hfill [2.8c]

where \(R_{pp}(0)\) represents the p-component of reflectivity for the demagnetized state and \(R_{pp}(\pm H)\) that for the magnetized at saturation states at the two directions of the magnetic field. Typical values of this magnitude range from \(10^{-4}\) to \(10^{-2}\).

If we consider a simple system of two semi-infinite non-magnetic and magnetic mediums, the TMOKE signal presents the following form [6]:
\[
\text{TMOKE} = 2n_i^2 \sin(2\theta) \Re \left[ \frac{\varepsilon_{xz}}{(n_2^4 - n_1^4)\cos^2 \theta - n_2^2 n_1^2 + n_1^4} \right]
\]  

where the Snell’s law has been used in order to show just the dependence on the light incident angle, and only linear terms of $\varepsilon_{xz}$ have been considered. This expression shows that the relative variation of reflectivity is zero at normal incidence. The maximum value of the TMOKE signal depends on both the incident angle and on the optical and MO properties of the materials involved. For example, Figure 2.2 shows this signal for a cobalt/air system as a function of the incident angle. As it can be observed the maximum MO activity is reached at $70^\circ$ approximately.

![Graph showing TMOKE signal as a function of incidence angle.](image)

**Figure 2.2** - TMOKE signal for a Co/Air system as a function of the incident angle.

2.1.2 Polar configuration ($\varepsilon_{xy} \neq 0, \varepsilon_{xz}, \varepsilon_{yz} = 0$)

The magnetic field applied in this case is perpendicular to the sample plane (see Figure 2.1b - $M_z$). The dielectric tensor presents the following form:

\[
\varepsilon = \begin{pmatrix}
\varepsilon & \varepsilon_{xy} & 0 \\
-\varepsilon_{xy} & \varepsilon & 0 \\
0 & 0 & \varepsilon
\end{pmatrix}
\]  

[2.10]

When linear p-polarized (s-polarized) light reflects from a sample under such situation, the reflected beam presents a small component of light s-polarized (p-polarized). The Polar MOKE (PMOKE) manifests then in that light becomes elliptically polarized with its major axis rotated from its initial incident polarization plane. The $r_{pp}$ and $r_{ss}$ Fresnel coefficients are not modified. However, the coefficients related to the polarization conversion present non-zero
and equal values \((r_{ps} = r_{sp} \neq 0)\) \([7]\). The rotation of the polarization plane \((\theta_k)\) and the ellipticity state \((\varepsilon_k)\) are related to these coefficients as follows:

\[
\theta_k + i\varepsilon_k = \frac{r_{sp}}{r_{pp}}
\]

\([2.11]\)

If we consider the same system of two semi-infinite non-magnetic and magnetic mediums the rotation and ellipticity are expressed as:

\[
\theta_k + i\varepsilon_k = \frac{\cos \theta_0}{\cos(\theta_0 + \theta_1)} \frac{-n_0 n_1^{-1} \varepsilon_{xy}}{\left(n_1^2 - n_0^2\right)}
\]

\([2.12]\)

expression that for the special case of incident light from vacuum and normal incidence reduces to:

\[
\theta_k + i\varepsilon_k = \frac{\varepsilon_{xy}}{n_1 \left(1 - n_1^2\right)}
\]

\([2.13]\)

An example of these magnitudes can be observed in Figure 2.3. It shows the respective Kerr rotation (Figure 2.3a) and ellipticity (b) for a Co/Air system as a function of the incident angle (blue lines).

At normal incidence the PMOKE can be explained quite easily from a phenomenological point of view. If we diagonalized the dielectric tensor in the basis of the circular polarizations, the refractive indexes \(n_+\) and \(n_-\) for the respective right and left-hand circular light polarizations become different when reflecting from the magnetized material:

\[
\varepsilon = \begin{pmatrix}
\varepsilon & \varepsilon_{xy} & 0 \\
-\varepsilon_{xy} & \varepsilon & 0 \\
0 & 0 & \varepsilon
\end{pmatrix}
\rightarrow \begin{pmatrix}
\varepsilon_+ & 0 & 0 \\
0 & \varepsilon_- & 0 \\
0 & 0 & \varepsilon
\end{pmatrix}
\]

and \(\varepsilon_+ = \varepsilon + i\varepsilon_{xy}\) \([2.14]\)

Linear polarized light with a given plane of polarization can be represented as a superposition of the right and left-handed circular polarizations with a definite phase difference. Each polarization finds different refractive indexes \(n_\pm = \sqrt{\varepsilon_\pm} = \varepsilon \pm i\varepsilon_{xy}\), giving rise to circular dichroism. Thus, the difference in the real part of the refractive index results in a change of the polarization plane, whereas a change in the imaginary part (absorption) results in a change of ellipticity.
2.1.3 Longitudinal configuration 
\( (\varepsilon_{yz} \neq 0, \varepsilon_{xy}, \varepsilon_{xz} = 0) \)

This configuration is characterized by an applied magnetic field parallel to both the sample and incident light planes. The dielectric tensor is described as:

\[
\varepsilon = \begin{pmatrix}
\varepsilon & 0 & 0 \\
0 & \varepsilon & -\varepsilon_{yz} \\
0 & \varepsilon_{yz} & \varepsilon
\end{pmatrix}
\]  \[2.15\]

This configuration presents a similar behavior when p-polarized (s-polarized) light reflects from the magnetized material. Light experiments a change in the rotation plane and ellipticity. Despite this similarity, the Longitudinal MOKE differs from the Polar case in that the Fresnel coefficients corresponding to the polarization conversion present different signs \( (r_{ps} = -r_{sp} \neq 0) \).

Figure 2.3a and b shows the LMOKE effect (Kerr rotation and ellipticity in cyan lines) for a Co/Air system as a function of the incident angle. As it can be observed there is no MO activity at normal incidence. Because of this, and also because the intensity of this effect is much smaller than that of PMOKE, the longitudinal configuration is not usually employed in MO characterizations.
2.2 MagnetoOptical Characterization

In this section we describe the MO characterization techniques used in this thesis. Two different experimental setups were employed to measure the MO response depending on the orientation of the magnetic field: the Transverse Kerr goniometer and the Polar Kerr spectrograph.

2.2.1 The Polar Kerr spectrograph

This setup is designed to obtain the rotation of the polarization plane and ellipticity state of the reflected light coming from a magnetized material under the Polar configuration (see previous section) at normal incidence. It is configured to allow measuring MO hysteresis loops at a particular wavelength or alternatively the measurement of MO spectra at saturation.

![Diagram of the Polar Kerr spectrograph]

Figure 2.4 - Scheme of the Polar Kerr spectrograph.

The Polar Kerr setup employs the widely used modulation of polarization technique because of its high sensitivity (~10³ degrees) [8,9]. A scheme of the setup for measuring the Ker rotation and ellipticity quantities is depicted in Figure 2.4. The light source consists of a Xenon lamp that cover a range of wavelengths from 240nm to 2000nm approximately. The light coming from this lamp is reflected in a series of concave mirrors (not shown in the scheme) and focused onto the access aperture of a monochromator. This monochromator (a SpectraPro150 model) has a 150mm focal length, dual indexable gratings of 600 and 1200g/mm and provides a spectral resolution of 0.4nm with 10µm wide slit. The light wavelength is then chosen, and the beam reaches a first polarizer which has its principal axis oriented at 45° degrees with respect to the setup plane.
Thereafter, the linear polarized beam is periodically modulated between linear and circular polarized light by a PEM-90 photo-elastic modulator. Modulation takes place with a frequency of 50 kHz and arbitrary phase amplitude $\varphi = \varphi_0 \sin \omega t$. The light is then focused onto the sample that is placed in between two magnetic coils. These are able to generate strong magnetic fields (up to 1.6T) perpendicular to the sample plane, i.e., polar configuration. After the reflection, the light goes through a second polarizer whose principal axis can be adjusted arbitrarily (for calibration measurements) and reaches a photo-detector. Finally, the signals from the photo-detector and from the photo-elastic modulator (reference signal) are sent to a lock-in amplifier.

- Jones matrices analysis.

In the following, we describe the principle of MOKE with polarization modulation technique in terms of the Jones matrix method [10]. Each optical component in Figure 2.4 can be expressed by a Jones matrix. All angles are relative to the plane of incidence. The electric field amplitude of the reflected beam $E^r$ at the photo-detector can be represented by the equation:

$$
\begin{pmatrix}
E_p^r \\
E_s^r
\end{pmatrix} = P_2 \cdot S \cdot M \cdot P_1 \cdot \begin{pmatrix}
E_p^i \\
E_s^i
\end{pmatrix}, \quad E_p^i = E_s^i = \frac{I^i}{\sqrt{2}}
$$

where $E^i$ is the amplitude of incident light. $E_p$ and $E_s$ are the amplitudes in the direction parallel and perpendicular to the plane of incidence. The $P_2$, $S$, $M$, and $P_1$ matrices represent the Jones matrices of the final polarizer, the sample, the photo-elastic modulator and the initial polarizer respectively. These are given by:

$$
P_1 = \frac{1}{2} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad [2.17a] \quad P_2 = \begin{pmatrix} \cos^2 \beta & \sin \beta \cos \beta \\ \sin \beta \cos \beta & \sin^2 \beta \end{pmatrix} \quad [2.17b]
$$

$$
M = \begin{pmatrix} e^{i\varphi/2} & 0 \\ 0 & e^{-i\varphi/2} \end{pmatrix} \quad [2.17c] \quad S = \begin{pmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{pmatrix} \quad [2.17d]
$$

where the axis of the initial polarizer matrix has been fixed to 45°, the axis of the final polarizer can rotate an arbitrary angle $\alpha$, and the sample matrix accounts for all the reflectivity coefficients. Introducing these expressions into [2.16] and operating, the intensity that reaches the photo-detector can be calculated, yielding:

$$
I^r = \left| E_p^i \right|^2 + \left| E_s^i \right|^2 = \frac{I^i}{2} (A + B \cos \varphi + C \sin \varphi)
$$

[2.18]
where the coefficients $A$, $B$, and $C$ are given by:

$$A = r_{pp}^2 + r_{sp}^2$$  \hspace{1cm} [2.19a]

$$B = \sin 2\alpha \left[ r_{pp}^2 - r_{sp}^2 \right] + 2r_{pp}r_{sp} \left[ (\sin^2 \beta - \cos^2 \beta) \cos (\delta_{pp} - \delta_{sp}) \right]$$  \hspace{1cm} [2.19b]

$$C = 4r_{sp}r_{pp} \sin (\delta_{pp} - \delta_{sp})$$  \hspace{1cm} [2.19c]

and the reflectivity coefficients have been defined in the polar form as [Note that for the particular case of normal incidence of light and polar magnetic configuration, $r_{ps} = r_{sp}$ and $r_{ss} = r_{pp}$ simplifying the overall calculation]:

$$r_{ss} = r_{pp} = r_{pp} e^{i\delta_{pp}}$$  \hspace{1cm} [2.20a]

$$r_{ps} = r_{sp} = r_{sp} e^{i\delta_{sp}}$$  \hspace{1cm} [2.20b]

Therefore, the intensity collected by the photo-detector (Expression [2.18]) depends on the sine and cosine of the phase-shift introduced by the photo-elastic modulator. As previously seen, this phase-shift presents a sinusoidal variation, so that the sine and cosine can be developed in function of Fourier series:

$$\sin \varphi = \sin \left( \varphi_0 \sin \omega_M t \right) = 2J_1 \left( \varphi_0 \right) \sin \omega_M t + ...$$  \hspace{1cm} [2.21a]

$$\cos \varphi = \cos \left( \varphi_0 \sin \omega_M t \right) = J_0 \left( \varphi_0 \right) + 2J_2 \left( \varphi_0 \right) \cos 2\omega_M t + ...$$  \hspace{1cm} [2.21b]

where $J_n(\varphi_0)$ are the Bessel functions of the nth kind evaluated at $\varphi_0$. The reflected intensity normalized to the incident intensity finally reads:

$$\tilde{I} = \tilde{I}_0 + \tilde{I}_1 \sin \omega_M t + \tilde{I}_2 \cos 2\omega_M t + ...$$  \hspace{1cm} [2.22]

where $\tilde{I}_0$, $\tilde{I}_1$, and $\tilde{I}_2$, are the relative intensities for the DC, first and second harmonic terms:

$$\tilde{I}_0 = A + BJ_0 \left( \varphi_0 \right)$$  \hspace{1cm} [2.23a]

$$\tilde{I}_1 = 2CJ_1 \left( \varphi_0 \right)$$  \hspace{1cm} [2.23b]

$$\tilde{I}_2 = 2BJ_2 \left( \varphi_0 \right)$$  \hspace{1cm} [2.23c]

and $A$, $B$, and $C$ are the expressions [2.19] previously deduced. Finally, in order to obtain the Kerr rotation and ellipticity, we attend to Equation [2.11]. These are defined as:
\[ \theta_p + i\varepsilon_p = \frac{r_{sp}}{r_{pp}} \frac{r_{sp}}{r_{pp}} e^{i(\delta_{sp} - \delta_{pp})} \]  

where we have introduced the polar form of the reflectivity coefficients. Taking this into consideration, normalizing the relative intensities of the first and second harmonics to the DC term, two different expressions are obtained proportional to the rotation and ellipticity of the reflected light:

\[ \frac{\tilde{I}_2}{I_0} = -4 J_2(\varphi_0) \left( \frac{r_{sp}}{r_{pp}} \right) \cos(\delta_{sp} - \delta_{pp}) = -4 J_2(\varphi_0) \theta_p \]  
\[ \frac{\tilde{I}_1}{I_0} = +4 J_1(\varphi_0) \left( \frac{r_{sp}}{r_{pp}} \right) \sin(\delta_{sp} - \delta_{pp}) = +4 J_1(\varphi_0) \varepsilon_p \]

where it was assumed that \( r_{sp} \ll r_{pp} \) and that \( \alpha = 0 \). Hence, apart from numerical factors, the normalized lock-in signals at \( \omega \) and \( 2\omega \) directly yield the Kerr ellipticity and rotation respectively. In addition, normalizing with respect to the DC signal, fluctuations due to instabilities of the light source and of the optical set-up are then very efficiently eliminated.

However, the values of \( J_2(\varphi_0) \) and \( J_1(\varphi_0) \) cannot be calculated since the real phase-shift introduced by the photoelastic modulator presents modifications. These are mainly due to the wavelength dependence of the piezo-optical coefficients of the modulator so \( \varphi_0 = \varphi_0(\lambda) \) \[11\], and to deviations from the paraxial geometry that makes the phase-shift to be different in function of the angle of the incident ray.

- **Calibration of the Polar Kerr Spectrograph**

Due to this, the rotation and ellipticity measurements need of a calibration process at each wavelength to obtain their values. To do so, let us consider again the expression for the intensity of the second harmonic \[2.23c\]. For very small angles of the polarizer, this expression reduces to:

\[ \tilde{I}_2 \approx \left[ 4\alpha \left( r_{pp}^2 - r_{sp}^2 \right) + 4r_{pp} r_{sp} \cos(\delta_{pp} - \delta_{sp}) \right] J_2(\varphi_0) = C_\theta(\beta - \theta_p) \]

Therefore, the intensity of the second harmonic is directly proportional to the Kerr rotation and to the rotated angle of the polarizer, being \( C_\theta \) the slope defined as \( C_\theta = 4\left( r_{pp}^2 - r_{sp}^2 \right) J_2(\varphi_0) \). This slope depends on the experimental setup and on the sample considered and therefore it is not known.
In order to obtain it, the angle of the final polarizer is rotated a series of small steps when the sample is demagnetized \((\theta_p = 0)\). The values obtained for \(I_2\) are then linearly fitted providing the value of the slope \(C_{\phi}\). Once this constant is calculated, the experimental Kerr rotation is obtained by fixing the polarizer angle to \(\alpha = 0\), measuring the intensity of the second harmonic \(I_2\) when the sample is magnetized at saturation, and then calculating the ratio between this quantity and that of the slope \(C_{\phi}\). An example of the calibration process can be observed in Figure 2.5 for a 300nm thick Ni film at \(\lambda=700\)nm. The blue and cyan dots represent the calibration measurements for \(\varphi_0 = 2.206\) and \(\varphi_0 = 3.029\) phase-shifts of the photoelastic modulator.

**Figure 2.5** - Calibration process of a Ni bulk film at a given wavelength (\(\lambda=700\)nm) and different phase shifts of the photo-elastic modulator. The linear fit provides the value of \(C_{\phi}\).

As it can be observed, the slope depends on the phase-shift introduced by the photo-elastic modulator. A larger slope entails a larger sensitivity in the measurement, so the phase-shift must be chosen wisely to maximize the value of \(J_2(\varphi_0)\) and therefore \(C_{\phi}\). This maximum is reached at \(\varphi_0 = 3.029\). However, the real phase-shift introduced by our photo-elastic modulator depends on the wavelength of light. For that reason, this is chosen so that the value of \(J_2(\varphi_0)\) is large enough along the spectral region where the measurements are performed (see Figure 2.6).

**Figure 2.6** - Spectral dependence of \(J_2(\varphi_0)\) as a function of the phase-shift introduced for a particular wavelength.
Finally, the experimental Kerr ellipticity cannot be obtained through a similar process. Attending to Expressions [2.19c and 2.23b], there is no dependence on the polarizer angle and therefore no calibration can be performed. To overcome this problem, a compensator can be introduced in the system, as it is pointed out in reference [8]. In our case, we used a Ni bulk sample (whose ellipticity is known) to calibrate the setup.

2.2.2 The Transverse Kerr Goniometer

This setup is designed to obtain the MO response of samples under the magnetic Transverse configuration. The measurements are performed at a single wavelength but as a function of the incident angle of light. The setup also uses a modulation technique in order to obtain the largest accuracy in the measurement. However, in this case instead of the light polarization, the modulation is performed over the magnetic field.

The scheme of the setup for measuring the Transverse Kerr MO quantities is depicted in Figure 2.7. The light source consists of a He-Ne laser (λ=632nm). Light coming from this source reaches a polarizer with its principal axis oriented at 0° allowing only the p-component of light to go through. Then, the p-polarized beam reflects at the sample, which is subject to an oscillating magnetic field with a frequency of ω=100Hz approximately. This magnetic field is generated by a magnetic coil able to produce fields of 400Oe, strong enough to saturate the samples. After the reflection, the beam reaches the photo-detector. Its signal and that of the magnetic coils (reference signal) are finally sent to a lock-in amplifier.

Figure 2.7 - Scheme of the Transverse Kerr Plasmonic Goniometer.
As it can be observed, this setup presents a much simpler configuration than the Polar Kerr setup. The Jones matrices analysis is indeed very simple. The electric field amplitude of the reflected beam $E'$ at the photodetector can be represented by the equation:

$$\begin{pmatrix} E'_p \\ E'_s \end{pmatrix} = S \cdot P_1 \cdot \begin{pmatrix} E'_p \\ E'_s \end{pmatrix}, \quad E'_p = E'_s = \frac{I^i}{\sqrt{2}}$$  \hspace{1cm} [2.27]

, and the S and P₁ matrices represent the sample and polarizer:

$$P_1 = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \hspace{1cm} [2.28a] \quad S = \begin{pmatrix} r_{pp} & 0 \\ 0 & r_{ss} \end{pmatrix} \hspace{1cm} [2.28b]$$

Operating these matrices, the total relative intensity that reaches the photodetector is proportional to the p-component of reflectivity.

$$\tilde{I}' = \tilde{I}_0 + \tilde{I}_1 \sin \omega_M t \hspace{1cm} [2.29a]$$

$$\tilde{I}_0 = r_{pp} \hspace{1cm} [2.29b]$$

$$\tilde{I}_1 = \Delta r_{pp} \hspace{1cm} [2.29c]$$

From this expression, the TMOKE signal can be calculated. The DC term accounts for the reflectivity, whereas the first harmonic accounts for the variation of reflectivity with the magnetic field.
Chapter 3

MagnetoOptical Activity in Pure Ferromagnetic Nanostructures

3.1 Motivation

The present chapter constitutes the first experimental and theoretical studies about the interrelation between plasmonic excitations and magneto-optical (MO) activity. Due to the particular conditions that must be fulfilled to show both MO and plasmonic activity, the choice of the material involved in such study is not straightforward. It must present a metallic character (to allow surface plasmon resonances) and at the same time it must exhibit MO activity at low magnetic fields.

These two conditions highly restrict the candidates to be employed, where ferromagnetic metals stand out as they present both characteristics spontaneously. The most usual ferromagnetic metals are: iron, cobalt and nickel. The first two ones may be very suitable as they present large values of the magnetization at saturation, which give a large MO activity. In contrast, these elements require high saturation magnetic fields (2.1T and 1.7T for iron and cobalt respectively), which may pose a drawback for future applications. Therefore, nickel becomes the best starting option as it is easier to saturate (0.8T). Another advantage is that it is more resistant to oxidation during and after the fabrication process of the nanostructures.
The chapter is organized as follows. The first part is devoted to the study of the MO response of nanostructures consisting in Ni nanowires embedded in an alumina (Al₂O₃) matrix. An enhancement of the MO activity will be observed in a particular spectral region, which coincides with that of the plasmonic excitation [1,2]. In order to verify that such enhancement is due to surface plasmons, a series of numerical simulations will be carried out to analyze the dependence of the MO response on the size and global dielectric variations of the refractive index of the host matrix. The second part of the chapter studies the MO response of the inverse nanostructure, that is, perforated nickel membranes. An enhancement of the MO activity due to plasmon-like excitations will be shown as well, its spectral position depending on the size and global variations of the infilling medium.

### 3.2 Ordered arrays of Ni nanowires.

In this section, we analyze the MO response of a structure consisting in Ni nanowires embedded in an alumina matrix. The choice for this structure comes from the great interest magnetic nanowires arrays have attracted for their particular magnetic properties with possible applications as perpendicular magnetic storage media [3]. The section is organized as follows. First, a brief description of the fabrication process and morphological characterization is addressed. Afterwards, the MO response of such system is obtained when the magnetic field is applied perpendicular to the sample plane, finding an enhancement with respect to a Ni bulk film. We will analyze the origin of such enhancement through extensive numerical simulations.

#### 3.2.1 Structure design and sample fabrication

The samples consist in arrays of nickel nanowires embedded in alumina membranes obtained by anodization of aluminum foils. The fabrication of these ordered alumina membranes was obtained using two different approaches. Samples with small pore diameter were obtained via a two-step anodization process [4]. It consists in a first long-duration anodization that causes the formation of an alumina membrane with a high pore diameter distribution and a non-ordered pore array. Immersing the sample in a CrO₃, H₃PO₄ and H₂O solution, the oxide structure is completely removed. The aluminum surface then presents a regular hexagonal texture which act as a self-assembled seed for a second anodization process. An ordered alumina nanopore array is obtained with straight pores from top to bottom, pore diameter of ~40 nm, interpore distance of ~105 nm, and a thickness of 5μm [5,6].
Figure 3.1 - Schematic representation of the fabrication process. A first anodization is achieved in order to obtain a hexagonal pattern on the aluminum that acts as a seed for a second anodization. Afterwards, the Ni nanowires are grown inside the perforated alumina membrane. In order to obtain larger pore diameters, the first anodization process is replaced by a nanoimprint lithography process.

The preparation of samples with larger pore diameter was accomplished by replacing the first anodization step with nanoimprint lithography for pre-patternning the aluminum surface. This pre-patterned aluminum foil was subsequently anodized and, controlling the anodization conditions, a perfectly ordered hexagonal array of pores with a pore diameter of approximately 180nm and interpore distance of about 500nm was obtained [7].

Figure 3.2 - SEM micrographs of hexagonal arrays of Ni nanowires embedded in an alumina matrix (Al₂O₃). In (a) the nanowire diameter is d=40nm and the interwire distance a=90-105nm and in (b) d=180nm and the interwire distance a=500nm approximately. The resulting Ni concentration oscillates between a 13% and a 17%.

The final step in the fabrication process of the nickel nanowires is common to both kinds of alumina membranes. It consisted in an electrodeposition process of nickel inside the nanopores. As a result, two different samples were obtained: sample A with 40nm diameter Ni pores and 90-105nm lattice constant; and sample B with 180nm diameter Ni nanowires and 500nm lattice constant. In Figure 3.2a and b it is depicted a SEM micrograph of the samples A and B. It can be observed that the nanowires present an almost perfect hexagonal arrangement, with a small diameter and lattice parameter dispersion. The amount of Ni within the nanostructures is approximately 17%.
3.2.2 MO response: Enhancement of the MO activity.

The magnetic behavior of the samples was analyzed performing a MO hysteresis loop on a polar Kerr spectrograph (see Chapter 2) at an arbitrary wavelength ($\lambda=530\text{nm}$, in this case). This technique was applied to both samples A and B. In Figure 3.3 it is depicted the magnetic behavior of sample A. Saturation of magnetization is reached at 3000 Oe approximately. Additional information about the magnetic anisotropy of the samples can be obtained by attending to the presence or absence of hysteresis in the loop. In this case, the presence indicates that the easy axis of magnetization is parallel to the cylinder axis.

![Figure 3.3 - Polar Kerr loop at $\lambda=530\text{nm}$ for the 40nm diameter Ni nanowires. The hysteresis in the loop reveals that the easy axis of magnetization coincides with that of the cylinder.](image)

The MO response of the nanostructures was analyzed by means of the same polar Kerr spectrograph at saturation conditions (magnetic field $\sim16\text{ kOe}$). Figure 3.4a show the Kerr rotation spectra obtained corresponding to the 40nm Ni nanowires diameter (sample A). The triangles represent the experimental Kerr rotation whereas the black line represents the Kerr rotation for a Ni bulk film. The Kerr Rotation may present positive or negative values, which only states for the direction of the rotation, clockwise (positive) or counter clockwise (negative). From these results it is possible to distinguish two different spectral regions, below and above 2.5eV. In the low energy region the Kerr rotation is smaller than that of Ni bulk. This is consistent with the amount of nickel in the nanostructure, 17%, which is much smaller with respect to the Ni bulk. However, above 2.5eV the Kerr rotation for the nanostructured sample is very similar to that of Ni bulk, meaning that for this energy range it is possible to obtain an enhancement effect despite having a smaller amount of magnetooptically active material.

The results for sample B (same Ni concentration but larger nanowire diameter with respect to sample A) are depicted in Figure 3.4b, where it can be seen how the signal is significantly smaller than for Ni bulk, being in this case the
nanostructure-to-bulk ratio in close relation with the nickel percentage. The
differences arising from the two samples indicate that the Kerr spectrum
strongly depends on the diameter and separation of the motives. Apparently, the
structure at 3.1eV in Figure 3.4a red-shifts as the diameter of the nanowires
increases. If it is so, a possible origin to this behavior could be the excitation of
a localized plasmonic (LSP) resonance, since it is extremely sensitive to
modifications of the nanoparticle size or surrounding environment. It could also
explain the enhancement of the Kerr spectra observed in the first sample.

![Figure 3.4](image)

**Figure 3.4** - (a) Kerr rotation spectra of an array of nanowires with d=40nm and
a=105nm. The continuous line represents the Kerr rotation of nickel bulk. The
enhancement of the rotation around 3eV is clearly visible. The blue line corresponds to
the SMM calculation. (b) The same as in (a) but the nanowire diameter is d=180 and the
lattice constant a=500nm (the nickel concentration remains the same).

### 3.2.3 Origin of the enhanced MO activity.

To corroborate this hypothesis and to analyze the enhancement and its
characteristics we employed the numerical SMM method described in Appendix
2. The optical and MO constants for nickel were taken from Refs. [8] and [9]
respectively. In Figure 3.4a and b (blue lines) we can see the results of applying
the SMM to the two different structures studied experimentally. The blue line in
Figure 3.4a is the result of considering the nickel wires to be embedded in an
$n=1.7$ dielectric environment. As it can be seen the agreement with the
experiment is qualitatively very good for both cases, although a better
quantitative agreement is reached in the case of sample A.

These calculations show the capability of the SMM method to reproduce the
experimental Kerr rotation, and therefore it can be used to explain the origin of
the differences observed for samples A and B. In this section we perform
extensive numerical calculations to show that the observed behavior for the MO
response of the samples is precisely due to the excitation of localized surface
plasmons (LSPs).
• 3.2.3.1 Size effects on the MO response.

Figure 3.5 presents the result of numerical simulations calculated for the Kerr spectra of nickel wires embedded on alumina and for different diameters, namely 40, 80 and 125nm. The lattice parameter was adjusted so that the nickel concentration remained constant, what means 90, 180, and 281nm inter-wire distance, respectively. These simulations reveal that the structure actually experiments a red-shift as the size of the wires increases. The feature is located in the near ultraviolet energy range, shifting from 3.1eV to 2.25eV approximately as we increase the diameter from 40nm to 125nm. All these facts reinforce the suggestion of a plasmon-like behavior [10]. It is worth noticing that despite the fact that the amount of Ni in the nanowire layers is only about an 17%, the Kerr rotation intensity of the system is similar than that of a continuous Ni film at the location of the resonance.

![Figure 3.5](image)

**Figure 3.5** - Theoretical Kerr rotation spectra for arrays of nickel nanowires with different diameters. The lattice parameter has been adjusted to keep the nickel concentration constant. The wires are embedded in an alumina matrix. The spectra appear red-shifted as the diameter of the wires increases. Despite that the nickel concentration is always around 18%, the rotation is of the order than that of the bulk material in all the cases.

On the other hand, the nanowires diameter and lattice constant are much smaller than the wavelength of light, thus allowing to describe the nanowires-alumina system in terms of an effective homogeneous medium (see Figure 3.6) with effective optical constants, $n$ and $k$. Figure 3.7a shows the effective absorption coefficients $k$ and its evolution as a function of the diameter for the same systems as in Figure 3.5. These coefficients are calculated from the theoretical values of transmission and reflection. Plasmonic structures manifest themselves as a peak in the spectrum of $k$. As we can see, all the spectra show a peak in the same region where the MO enhancement is found and, moreover, the peak position red-shifts when the diameter of the wires increases.
Moreover, from the SMM calculations the total MO activity, defined as the modulus of the complex Kerr rotation $|\theta + i\varphi| = |r_{ps}/r_{pp}|$, can be obtained. Figure 3.7b shows this magnitude for the same systems as those of Figure 3.5. Each spectrum shows a peak that shifts to lower energies as the diameter of the nanowires increases. Their spectral are very similar to those of $k$, indicating that the ultimate responsible of the Kerr rotation enhancement is the plasmon-like resonance.

From these results we could conclude that the plasmon resonance acts just as a pure optical effect. However, the Kerr rotation has two components (see Chapter 2): $r_{ps}$ and $r_{pp}$ which accounts for the pure MO and pure optical effects respectively. To point out that the MO enhancement is not only optical, Figure 3.7c presents the $|r_{ps}|$ spectra for the same systems as before and for a Ni bulk film (black line). Their shapes are again very close to that of $k$ since the origin is the same, i.e. the presence of the plasmon. Attending to the value of $|r_{ps}|$, they are of the order of 50% of that corresponding to Ni bulk. Since the Ni concentration is 17%, it represents an enhancement of the polarization conversion which may be ascribed to the localization of the electromagnetic field (EM) within the MO active material, consequence of the plasmon excitation.
Figure 3.7 - (a) Effective absorption constant for the same systems as in Fig. 3. The evolution of the peak nicely reflects that observed in the Kerr rotation spectra. (b) Total MO activity for the same nanowire system. The peak structures reproduce that of the effective $k$, indicating the origin of the red-shift and of the enhancement. (c) Purely magneto-optical part of the MO activity ($|r|_p$ polarization conversion) for the same systems. The black line corresponds to the $|r|_p$ of a Ni bulk film.

- 3.2.3.2 Environment effects on the MO response.

One way to confirm the connection between the EM field and the polarization conversion $|r|_p$ is to compare these two magnitudes as the refractive index of the embedding medium changes. To perform such comparison, we will use the SMM method to obtain theoretical $|r|_p$ spectra for different values of the refractive index of the host material. These spectra are complemented with 3D-FDTD simulations to obtain the EM field distribution at selected wavelengths [11].

The structure considered for the theoretical analysis consists in an ordered hexagonal array of nickel nanowires embedded in a dielectric matrix. We have
analyzed wires of 80nm diameter, a lattice parameter of 180nm, and a height of 2μm. The diagonal and off-diagonal dielectric constants of nickel are the same used in the preceding section. The refractive index of the surrounding medium remains energy independent (n=1.7) and similar to that of alumina. A schematic view of the model system is shown in Figure 3.8, the rectangle superimposed corresponds to the unit cell employed in the FDTD calculations, and it also shows the distribution of the field intensity at the top of the nanostructure and energy of a plasmonic resonance.

**Figure 3.8** - Schematic view of the model employed in the calculations, consisting on a hexagonal array of nickel nanowires embedded in a dielectric medium. The rectangle superimposed represents the unit cell for the FDTD calculations, and shows the EM field distribution at 3eV.

The calculations corresponding to the |r_{ps}| are presented in Figure 3.9a for different surrounding mediums (from n=1.7 to n=1.4). A peak can be observed in all of them, blue-shifting as the refractive index decreases. This is in agreement with the observed behaviour in non-magnetic plasmonic nanoparticles [12]. In addition, the intensity of |r_{ps}| also increases with the refractive index. The results of the average distribution of the EM field within the nanowires are shown in Figure 3.9b. As it can be observed the spectra reproduce the same trend observed for the polarization conversion.
Figure 3.9 - Theoretical spectra of the (a) polarization conversion $|r_{ps}|$ and (b) average EM field intensity within the Ni nanowires for the same system described in Figure 3.8. The black, blue, cyan and purple lines correspond to a decreasing refractive index of the embedding medium (from $n=1.7$ to $n=1.4$ respectively).

### 3.3 Perforated Ni membranes.

So far, we have shown an enhancement of the MO activity in nanowires due to the excitation of plasmon-like modes. However, there are a great number of nanostructures showing plasmonic excitations that may be suitable to enhance the MO activity as well. For example, noble metal membranes are under extensive study since they exhibit anomalous optical responses resulting from the excitation of plasmons, such as the extraordinary optical transmission (EOT) bands observed in metallic patterned thin films with arrays of sub-wavelength holes [13]. In this way, some preliminary works [14] have analyzed these EOT in ferromagnetic materials like cobalt perforated membranes. Therefore, these nanostructures are appropriate candidates to study the influence of plasmons on the MO response. Very recently, a study has observed the enhancement of the MO response in cobalt perforated membranes, finding a correlation between this and the excitation of surface plasmons propagating along the pores [15,16].

In particular for this section, we will study the MO response and possible enhancements in perforated membranes out of the EOT regime. The structure proposed, perforated Ni membranes, consists in the complementary of that analyzed in the previous section. The aim of this choice is to compare both MO activities and their origins, since the perforated membrane also shows an enhancement of the MO response. The section is organized in a similar way to the previous: fabrication and morphological description, observation of an enhanced MO response, and explanation of this through numerical simulations.
3.3.1 Structure design and sample fabrication

Figure 3.10 - Scanning electron micrographs of both sides of a representative membrane obtained by the replica/antireplica method.

The samples prepared for the analysis consists in Ni membranes prepared by replica/antireplica methods using nanoporous anodic alumina membranes as templates [17]. The pores are filled in with melted polymethyl methacrylate (PMMA). An Au thin layer, 15 nm thick, is sputtered on top of the alumina membrane. Removing both Al substrate and alumina membrane by chemical etchings, a PMMA antireplica of the alumina membrane (i.e. an array of pillars) is obtained. Due to the Au film previously deposited, a Ni membrane can be fabricated by electrodeposition. Finally, the PMMA pillars are chemically removed. An example of the final process can be appreciated in Figure 3.10, which shows SEM images of the Ni (top) and Au (bottom) sides of a 200 nm thick membrane.

Figure 3.11 - Schematic representation of the fabrication process. After obtaining an alumina membrane through the methods described for the fabrication of the nickel nanowires (see Figure 3.), a PMMA solution fills the pores and subsequently UV radiated in order to create PMMA nanopillars. An Au layer is deposited on top of this membrane. Then, the Al substrate and alumina are chemically removed. The nanopillars are then used as a mask in order to grow the Ni membrane by an electrodeposition process.
3.3.2 MO response: Enhancement of the MO activity.

The analysis following is carried out in the same way as for the nanowires. Figure 3.12 shows a MO loop at $\lambda=530\text{nm}$ showing that the saturation of the magnetization is reached at the same value as for the Ni nanowires ($\sim 3000\text{Oe}$). Again, the presence or absence of hysteresis gives information about the magnetic anisotropy of the sample. In this case, there is no noticeable hysteresis in the loop, which pinpoints that the easy axis of magnetization is this time perpendicular to the axis of the holes. This is due to the small thickness of the perforated membrane ($\sim 200\text{nm}$).

![Kerr Rotation vs H (Oe)](image)

**Figure 3.12** - Polar Kerr loop at $\lambda=530\text{nm}$ for the 40nm diameter Ni membrane. The absence of hysteresis in the loop reveals that the easy axis of magnetization is perpendicular to the axis of the cylindrical hole.

The spectral MO response was analyzed for a Ni membrane with pore diameter $d=45\text{nm}$ and lattice parameter $a=90\text{nm}$. These results are compared with the Ni nanowires array from the last section ($d=40\text{nm}$ and $a=105\text{nm}$) and Ni bulk, since the diameter and the lattice constant are similar to those of the Ni nanowires previously studied [2]. The polar Kerr rotation spectra are shown in Figure 3.13, where the black line corresponds to a Ni bulk film, the blue triangles correspond to the ordered array of Ni wires and the cyan triangles to the perforated Ni membrane. As observed, the spectra show two different regions: the high energy region where the spectra seem to be similar to each other and the rotation reaches its maximum, and the low energy region where the three spectra present a monotonous evolution and the rotation is different for each sample.
Figure 3.13 - Polar Kerr rotation spectra of the membrane observed in Figure 3.10 (cyan triangles), a nanowire array (blue triangles) and Ni bulk (black line).

In the low energy region, the different values for the three samples may be explained by the absence of any plasmon resonance or other anomalous effect, and thus the Kerr rotation is expected to be more or less proportional to the amount of ferromagnetic material. For instance, from the nominal parameters, the Ni concentration is a 77% in the hexagonal array of holes whereas it is a 13% in the nanowires array. From the values shown in Figure 3.13 at long wavelengths, we can see that the Kerr rotation for the nanowire array and for the membrane is roughly proportional to the Ni concentration.

However, for higher energies the Kerr rotation signal does not follow this simple rule of thumb. In fact, a minimum around 3.1eV with similar Kerr rotation can be observed for the three samples. In the case of Ni nanowires, it has been demonstrated [2] that the excitation of plasmon-like modes provides an intensification of the EM field compensating the smaller amount of MO active material when compared to the bulk, therefore leading to similar Kerr rotation values. The similar behavior observed in the Ni membranes suggests an analogous mechanism for the enhancement. In particular, the enhancement should be related to the excitation of plasmon-like modes in the pore. When this plasmon mode is excited we expect an enhancement of the EM field in the Ni material around the pore and therefore an increase of the MO activity. If plasmon excitation may be then regarded as responsible of such enhancement in the MO response of the membrane, the spectral location of the maximum Kerr rotation must vary as the refractive index n of the material inside the pore is modified, exactly as it has been observed in the nanowires case [18]. The fabrication method used to prepare the membranes provided the means to obtain an experimental verification of the red shift originated on the increase of the infilling n: if we leave out the last step, a Ni membrane with PMMA inside the pores can be obtained.
Figure 3.14 - (a) Experimental polar Kerr rotation spectra of the same membrane as in Figure 3.13, before (blue triangles) and after (cyan triangles) the PMMA is removed; (b) calculated spectra of the membrane under the same conditions (Parameters employed diameter $d=45\text{nm}$ and lattice constant $a=90\text{nm}$).

Figure 3.14a shows the Kerr rotation of the same Ni membrane as in Figure 3.14, but before (blue triangles) and after (cyan triangles) the PMMA was chemically removed. As it can be observed, there are clear differences, more evident in the high energy region, where a clear red-shift of the structure is observed as the refractive index of the material filling the pores increases. This behavior is the expected one provided the enhancement is due to surface plasmons resonances of the pores: any increase of $n$ in the pore (from $n=1$ to $n=1.5$ in this case) makes the plasmon resonance shift towards lower energies in a similar way it has been observed for the Ni nanowires. In order to confirm such behavior, theoretical calculations were performed for these membranes. The results are displayed in Figure 3.14b. As can be observed the simulation reproduces the enhancement of the MO activity in the high energy region as well as the ref shift observed in the experimental spectra due to the increase of $n$ in the infilling material. The minor differences observed between the experimental and theoretical spectra may be ascribed to the different refractive index of the material filling the pores and that employed in the calculations.

3.3.3 Origin of the enhanced MO activity.

In the same way it has been observed in the nanowires analysis, these results pointed out to a plasmonic origin. This time the plasmonic resonance seems to be localized at the membrane holes. These are localized plasmonic resonances, so they must be affected by changes in the size of the hole or in the material filling them. Therefore, if the Kerr rotation behavior and enhancement has a plasmonic origin, the MO response may be controlled by changing these parameters. To prove this, we performed numerical calculations by means of the SMM method.
and FDTD solutions as the hole diameter changes and for global changes of the refractive index within the holes.

- 3.3.3.1 Size effects on the MO response.

Figure 3.15 presents the calculated Kerr rotation spectra for structures that present the same amount of Ni (60%) but different pore diameter and interpore distances. The blue line corresponds to d=70nm hole diameter and a=105nm lattice constant, whereas the cyan line corresponds to d=120 nm and a=180 nm. The material filling the hole has a refractive index of \( n=1.5 \). In the low energy region, where no plasmon exists, all the simulated membranes exhibit similar Kerr rotation value, proportional to the amount of nickel. However, in the high energy region a large enhancement and a red shift of the Kerr spectra are observed, which is ascribed to the increase of the diameter. This red shift is precisely what is expected if LSP are excited in the pores. Moreover, similar to the behavior found in the experimental results, an enhancement of the MO rotation can be appreciated with respect to the nickel bulk in all the cases, pointing out the effect of the plasmon excitation on the MO activity [19].

![Kerr Rotation vs Energy](image)

**Figure 3.15** - Theoretical polar Kerr rotation spectra of four different membranes (pore-magnetic material ratio constant). Blue lines: \( n=1.0 \) (continuous line) and \( n=1.5 \) (dotted line) filled pore of a membrane with 70nm diameter. Cyan lines: same as blue lines but for a 120nm diameter membrane.

- 3.3.3.2 Environment effects on the MO response.

So far, it has been observed that the perforated Ni membranes behaves in a equivalent manner to what is has been observed in the Ni nanowires, in terms of the MO response dependence to the pore size and refractive index. Consequently, it may be expected to find the same connection between the localization of the EM field within the Ni membrane due to the plasmonic excitation and the enhancement of the polarization conversion \(|r_{ps}|\). Thus, here we analyze the relation between these magnitudes as a function of the refractive index filling the pores. The structure considered for the theoretical analysis
consists in a perforated membrane, whose holes are hexagonally arranged. For the sake of comparison, the geometrical parameters are identical to that of the nanowires structures (80nm and 180nm for the wire diameter and lattice parameter respectively), as it can be observed in the schematic view shown in Figure 3.16. Once more, the rectangle superimposed corresponds to the unit cell employed in the FDTD calculations and shows the distribution of the field intensity in the nanostructure at an arbitrary energy.

**Figure 3.16** - Schematic view of the model employed in the calculation, consisting in a hexagonally perforated nickel membrane, inverse structure of the nickel nanowires system previously analyzed. The holes are filled with a dielectric medium. The rectangle superimposed represents the unit cell employed in the FDTD calculations and shows the EM field distribution in the system.

Figure 3.17a represents the $|r_{ps}|$ spectra of the perforated membrane as the refractive index filling the pore changes from $n=1.7$ to $n=1.4$. The maximum of the $|r_{ps}|$ peaks lie in the same energy region of those corresponding to the nanowires nanostructure, pointing out to a plasmonic origin. Moreover, the peak shifts to higher energies as the refractive index decreases, which is again compatible with a plasmonic behaviour. Comparing these results with those obtained for the average EM field intensity within the Ni membrane (see Figure 3.17b), it can be observed the same trend with the refractive index.
2.5 3.0 3.5 4.0
0.6
0.7
\(|E|\)
2
wires
\(|\mathbf{E}|^2 \cdot \text{m}^{-2}\)
Energy [eV]
Wavelength [nm]

(a) (b)

Figure 3.18 - Theoretical spectra of the (a) polarization conversion \(|r_{ps}|\) and (b) average EM field intensity within the nickel for the latter system. The black, blue, cyan and purple lines correspond to a decreasing refractive index of the infilling pore medium (from \(n=1.7\) to \(n=1.4\) respectively).

3.4 Conclusions and Outlook

Along this chapter, two nanostructures made of ferromagnetic metals, exhibiting MO activity and plasmonic properties simultaneously, have been studied: the first one, an array of Ni nanowires embedded in an alumina matrix and the second one a perforated Ni membrane, inverse of the first one. We have shown that the excitation of plasmon-like modes leads to strong modifications in the MO response.

In the plasmon excitation region, the nanowire as well as the perforated membrane arrays presented similar Kerr rotation intensities despite the different Ni amount in the system. In the other spectral region, where no plasmon is excited, the intensity of the rotation turns out to be basically proportional to the amount of ferromagnetic material. This effect has found to be not only due to a reduction of the optical reflectivity of the system, but also to an increase of the polarization conversion due to the increase of the EM field within the MO active material. The spectral position of the enhancement could be tuned by changing the refractive index of the material surrounding (filling) the wires (pores) and/or changing the geometrical parameters.

In order to complete the study of the ferromagnetic nanostructures, it would be interesting to analyze other magnetic configurations as well as alternative designs for the nanostructures. In that sense, measurements both in polar and transversal configurations are currently being carried out on a structure of Ni inverse opals. However, there are some open questions that could considerably extend our understanding about the connection between plasmon excitations and MO response (some of them will be addressed in the following chapters): for
example, the effect of nanoparticle concentration on the MO response. This may be of great importance since concentration affects especially the interactions of plasmons, as it has been shown in non-magnetic materials [20]. Moreover, these nanostructures have shown that the plasmonic resonances are highly extended along the energy range, making much less attractive their use in the search of a practical sensing application. Consequently, the main question that stays behind is: is there any way to enhance the MO activity by means of plasmon excitations so that the resonance could be narrower?
Chapter 4

MagnetoOptical Effects in Nanostructures involving Noble-Metals and Ferromagnets

4.1 Motivation

In the previous chapter a strong correlation between the enhancement of the MO activity and the excitation of LSP resonances has been demonstrated. The analysis was presented for a series of nanostructures in which the surface plasmon lies in a wire/hole. In that study, the LSP resonance is very broad. This is due to the high optical absorption of the ferromagnetic material that results in a strong damping of any intrinsic LSP resonance. In order to obtain MO activity with narrower plasmon resonances, which would be desirable for the development of high sensitivity devices, other metals presenting smaller absorption must be employed. These are for example, noble-metals like gold or silver, although they present a weak MO activity respect to ferromagnets.

However, combining both kinds of materials we might achieve MO activity together with narrow plasmonic features.

In this chapter, we analyze how the excitation of LSPs affects the MO activity in systems containing noble and MO active materials. Theoretical studies performed by M. Abe and T. Suwa [1,2], have shown a large enhancement of the MO activity in core-shell (ferromagnets-noble metals) nanoparticles due to LSP excitations. Prior to our work, several attempts have been carried out using different chemical synthesis methods to fabricate complex onion-like
nanoparticles made of noble metals and ferromagnetic materials [3-6], but the effect of LSP excitations on the MO activity was not reported.

The chapter is organized as follows. The first section will be devoted to the study of the MO response of pure Au nanodisks. This system should present a negligible MO activity, since gold is not a ferromagnetic material. However, due to the excitation of LSPs, a measurable MO signal larger than that of a continuous gold film will be observed despite the smaller amount of material. The MO activity of such a system will be studied as a function of the nanodisk diameter to explain the physical origin of the observed enhancement. The second section of the chapter will be devoted to the enhancement of the MO signal in a nanostructure composed of Au/Co/Au nanodisks. The dependence of the MO activity will be studied as a function of the nanodisk diameter as well as on the amount of cobalt in the nanodisks. For both systems, a large enhancement of the MO activity assisted by LSPs will be shown and, moreover, that this enhancement is much more localized in a spectral region due to the presence of noble metals. The final section will be devoted to compare the results obtained for the previous structures and to analyze the differences in the shape of their MO spectra as a function of the MO dielectric constants of gold and cobalt.

4.2 MO activity in pure Au nanodisks

The physical mechanism that gives rise to magnetism in ferromagnets and noble metals is completely different. Ferromagnets spontaneously present a large magnetic moment (and subsequently MO activity) at small magnetic fields (~1 Tesla). In contrast, the magnetization in noble metals depends linearly on the magnetic field applied. These differences turn into different values of the MO dielectric constants. For example, Figure 4.1 shows the real ($\varepsilon_{xy}'$) and imaginary ($\varepsilon_{xy}''$) parts of the MO constants for cobalt (a) and gold (b). As it can be seen, those of the ferromagnetic metal are more than 500 times larger than that of gold. This stress the huge magnetic fields noble metals need in order to present a MO activity equivalent to that of ferromagnets.
Figure 4.1 - Real and imaginary parts of the MO dielectric constants of (a) cobalt at saturation and (b) gold for a magnetic field of 16000 Oe. Note that gold is a diamagnetic material.

However, despite the extremely small value of $\varepsilon_{xy}$ for gold, we may expect an enhancement of the MO response when surface plasmons are excited, as it has been pointed out using continuous films [7,8]. This section is therefore devoted to the study of the MO response of nanostructures composed exclusively of noble metals [9]. In particular we will analyze nanodisks composed of pure gold. The choice for gold is not arbitrary. Silver certainly presents better plasmonic properties than gold, but instead, it easily oxidizes increasing complexity in the fabrication process.

4.2.1 Fabrication process and Optical characterization

The fabrication of the samples was performed via a hole-mask colloidal lithography process [10]. First, a PMMA layer is spin coated over a glass substrate. Then, the sample is exposed to a colloidal solution of latex beads. After that, an Au layer (10nm thick) is sputtered over the sample. The following step is a tape stripping removal of the latex beads leaving the area beneath the latex unprotected. Subsequently, the sample is exposed to a reactive oxygen plasma etching. The holes are finally filled with gold and the photoresist is eliminated to obtain the nanodisks.
Using this process, different samples were obtained with different nominal aspect ratios: a) gold nanodisks with 60nm diameter and 32nm height; and b) gold and silver nanodisks with 100nm diameter and 20nm height. In Figure 4.2 we show SEM and AFM images of the final outcome for a sample of gold nanodisks. From these images we can obtain the nanodisks concentration (10%), being the actual diameter of the nanodisks 120nm and with small dispersion. Similar results were obtained for the 60nm diameter nanodisks, being the real diameter 70nm.

The optical characterization of the samples was performed through extinction measurements. In Figure 4.3 the extinction spectra of the Au nanodisks samples with 70 (cyan line) and 120nm (blue line) diameters respectively are depicted. These spectra show the expected peak [11-13] due to the excitation of the LSP,
whose spectral position shifts to lower energies (from 2.1eV to 1.8eV approximately) as the nanodisks diameter increases.

![Graph showing extinction spectra of Au nanodisks](image)

**Figure 4.3** - Experimental extinction spectra of Au nanodisks with diameters \(d=70\text{nm}\) (cyan line) and \(d=120\text{nm}\) (blue line). As the diameter of the disk increases the plasmon resonance shifts to lower energies, as expected.

### 4.2.2 MO characterization: Enhancement and size effects on the MO response.

The MO characterization of the samples was carried out using the Polar Kerr spectrograph described in Chapter 2. Considering the nature of the samples involved, the process to obtain the MO response of the Au nanodisks presents some difficulties. The MO signal obtained through the Polar Kerr setup is extremely weak and therefore very close to the noise signal since the MO activity of gold is several orders of magnitude smaller than that of ferromagnetic metals. Added to this, one has to consider the low concentration of nanodisks in the sample. Furthermore, the nanodisks were grown over a glass substrate that presents a large Faraday rotation activity. For that reason, the methodology applied on the measurement process was adapted to take account of these problems. Specifically, several spectra were obtained and subsequently averaged to remove the system noise signal; the Faraday rotation was removed by attaching a prism to the glass substrate by means of an index matching liquid, to avoid the reflected light coming from the last glass-air interface reaching the photo detector (see **Figure 4.4**). This method presents a drawback: the maximum magnetic field that can be applied is significantly smaller due to a larger separation of the magnetic coils when inserting the prism. As a result, all the measurements were performed at 0.8T.
Figure 4.4: From A) to B): Modification of the Polar Kerr experimental setup in order to remove any possible Faraday signal coming from the glass substrate of the samples. A prism with identical refractive index to the substrate is attached by means of a matching liquid to the bottom side of the sample, and as a result the Faraday is reduced.

An example of the MO response after the elimination of the Faraday contribution is presented in Figure 4.5, showing a polar Kerr loop at $\lambda=530\text{nm}$ for the 70nm diameter Au nanodisks. As gold is a diamagnetic material, we observe the typical linear behaviour as a function of the magnetic field.

![Kerr Rotation vs Magnetic Field](image)

Figure 4.5 - Polar Kerr loop at $\lambda=530\text{nm}$ for the 70nm diameter Au nanodisks.

The results of the spectral MO response of the Au nanodisks are depicted in Figure 4.6a (Kerr rotation) and Figure 4.6b (Kerr ellipticity). The cyan and blue triangles correspond to the 70 and 120nm diameter nanodisks respectively. These spectra show a peak for the rotation and an s-like structure for the ellipticity, related by the Kramers-Kronig relations, lying in the same spectral position to those corresponding to the extinction measurements. As expected, the structures shift to lower energies as the diameter of the nanodisks increases, denoting the excitation of LSPs on the Au nanodisks and its influence on the MO response.
Despite that these facts confirm the excitation of LSPs, the origin of the observed MO enhancement could be due to different reasons. When an LSP is excited the electromagnetic (EM) field concentrates at the nanoparticle. This can be better seen in Figure 4.7a and b, where we show the EM field distribution of Au nanodisks with 150nm diameter and 32nm height, out of and in resonance, respectively. At the LSP resonance, the field surrounds the nanodisk and at the same time it partially permeates the glass substrate. Therefore, the enhancement of the MO response may be due to the MO activity of the Au nanodisks or to the MO activity of the glass substrate.

In order to confirm that the enhancement of the MO response comes from gold and not to the Faraday signal coming from the glass substrate, complementary samples of Au nanodisks with same diameter were obtained replacing the standard $n=1.5$ glass (BK7) substrate with a MOS105 glass ($n=1.72$) that
presents a much larger Faraday activity. The Verdet constants for these glasses are shown in Figure 4.8a and b, respectively. As it can be observed the Faraday activity of the MOS105 glass is approximately 5 times larger than that of BK7 glass and opposite in sign.

\[ \frac{\text{Verdet}}{\text{rad/(T·m)}} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Energy [eV]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Wavelength [nm]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Energy [eV]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Wavelength [nm]} \]

\[ \text{Verdet [rad/(T·m)]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Energy [eV]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Wavelength [nm]} \]

\[ \text{Verdet [rad/(T·m)]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Energy [eV]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Wavelength [nm]} \]

\[ \text{Verdet [rad/(T·m)]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Energy [eV]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Wavelength [nm]} \]

\[ \text{Verdet [rad/(T·m)]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Energy [eV]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Wavelength [nm]} \]

\[ \text{Verdet [rad/(T·m)]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Energy [eV]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Wavelength [nm]} \]

\[ \text{Verdet [rad/(T·m)]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Energy [eV]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Wavelength [nm]} \]

\[ \text{Verdet [rad/(T·m)]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Energy [eV]} \]

\[ 800 \quad 700 \quad 600 \quad 500 \]

\[ 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \]

\[ \text{Wavelength [nm]} \]

Figure 4.8 - Spectral dependence of the Verdet constant for the (a) BK7 and (b) MOS105 glasses.

The experimental ellipticity spectra obtained from these samples is depicted in Figure 4.9b as green triangles. If the MO response was due to the MO signal of the glass substrate, the spectrum should present a much larger intensity and a change of sign with respect to the Au nanodisks grown over the BK7 substrate. It can be observed that the spectrum simply presents a lower intensity, and a slight shift to lower energies, confirming that the MO signal is exclusively due to the MO activity of the Au nanodisks. The differences found between the signals may be ascribed to two possible reasons. First, to the slightly different disks concentrations that can be found in the samples (~9.5% for the BK7 substrate and ~12% for the MOS105 substrate). When the disks concentration increases, the reflectivity of the effective medium increases as well, making the MO activity to decrease (in contrast, a decrease in the nanodisks concentration makes the reflectivity to decrease and the MO activity to reach that of a single nanoparticle). Second, to the different refractive index the glasses present. As the MOS105 refractive index is larger than that of BK7 glass, the reflectivity increases and consequently the MO activity decreases. Moreover, due to the extreme sensitivity of LSP to modifications of the refractive index, the plasmonic resonance will be located in a slightly different spectral position, which may be the origin of the red-shift.
Figure 4.9 - Experimental Kerr Ellipticity spectra for the gold nanodisks sample grown over a BK7 (blue triangles) or a MOS105 (green triangles) glass substrate. The refractive indexes of these glasses are $n=1.5$ and $n=1.7$, respectively.

In addition, samples of Ag nanodisks with similar parameters to that of the Au nanodisks were obtained, in which the glass substrate was replaced by a 400nm SiO2 layer on top of a Si wafer (see sketch of Figure 4.10). In principle, silver presents a narrower and stronger LSP resonance, becoming in principle the best candidate for the study of its MO response. Nevertheless, as it was mentioned at the beginning of the section, it presents a severe disadvantage, oxidation, which requires the adoption of protection measures. Thus, the Ag nanodisks were coated with 160nm PMMA as protecting layer.

Figure 4.10 - (a) Absorption coefficient $k$, (b) Kerr Rotation activity, and (c) Kerr Ellipticity activity of a sample consisting in Ag disks of 100nm diameter and 20nm height on top of a 400nm SiO2 + Si layer, and coated with a 160nm PMMA protecting layer.

Figure 4.10a and b present respectively, the optical absorption coefficient (obtained from ellipsometry measurements), and the Kerr rotation and ellipticity measurements. As it can be noticed, similar features to that observed in the optical and MO response of the Au nanodisks are found: the optical peak lies in
the same energy region where a peak and s-like structures appear for the rotation and ellipticity. In particular, when these features are compared to the 120nm diameter Au nanodisks, a blue-shift can be appreciated indicating yet again the presence of LSPs resonances in the Ag nanodisks. Therefore, the MO enhancement is not a consequence of the gold properties, but is instead a generic property of all nanoparticles made with materials with Drude-like conductivity.

### 4.2.3 Physical origin of the MO enhancement.

The experimental results have shown an enhancement of the MO response in the region were the LSP is excited. The physical mechanism that gives rise to MO activity in noble metals comes from the Lorentz force that experiment the in motion conduction electrons. If these electrons are under the influence of an oscillating electromagnetic field, then the Lorentz force can be expressed as [14]:

$$\vec{F}_{\text{Lor}} = \frac{d\vec{p}}{dt} \times \vec{B}$$  \[4.1\]

This equation suggests that every material illuminated with a light beam is susceptible to undergo a Lorentz force since:

$$\vec{F}_{\text{Lor}} = i\omega \alpha \vec{E}_0 \times \vec{B}$$  \[4.2\]

where $\alpha$ is the polarizability of the material, $E_0$ the electric field amplitude of the incident the light beam, and $\omega$ its frequency. This effect is in general very weak, unless either the external magnetic field or the polarizability is very strong. Since the intensity of the applied magnetic field on the samples is quite small, it leaves the second option as the origin of the MO enhancement. Thus, to study in detail the behaviour of polarizability, a simple model was considered consisting in a spherical metal nanoparticle of radius $a$, much smaller than the light wavelength $\lambda$. In the presence of a magnetic field, the polarizability of the nanoparticle becomes non-diagonal, and can be described by the following tensor:

$$\tilde{\alpha} = 4\pi a^3 (\tilde{\varepsilon} - \varepsilon_d \tilde{I})(\tilde{\varepsilon} + 2\varepsilon_d \tilde{I})^{-1}$$  \[4.3\]

where $\tilde{I}$ represents the identity 3x3 matrix, $\tilde{\varepsilon}$ the dielectric tensor of the metal and $\varepsilon_d$ the dielectric constant of the external dielectric medium [15,16]. If the external magnetic field is applied in the z-direction, the components of the $\tilde{\alpha}$ tensor are:

$$\alpha_{xx} \approx 4\pi a^3 \frac{\varepsilon_{xx} - \varepsilon_d}{\varepsilon_{xx} + 2\varepsilon_d}$$  \[4.4a\]
\[ \alpha_{xy} \approx 4\pi a^3 \frac{3\varepsilon_{xy}\varepsilon_d}{(\varepsilon_{xx} + 2\varepsilon_d)^2} \]  

Equations [4.4] shows that the LSP is excited when \( \varepsilon_{xx} = -2\varepsilon_d \) and predicts the generation of an electric dipole transversal to that excited by the induced light, which is proportional to the magnetic field amplitude, and exhibits the same resonant condition. Therefore, in the presence of an external magnetic field, the net induced dipole in the nanoparticle oscillates at an angle \( \theta_K \) with respect to the electric field of incident light (see Figure 4.11), given by:

\[ \theta_K = \arctan \left( \frac{\alpha_{xy}}{\alpha_{xx}} \right) \approx \frac{3\varepsilon_d\varepsilon_{xy}}{(\varepsilon_{xx} - \varepsilon_d)(\varepsilon_{xx} + 2\varepsilon_d)} \]  

The angular shift of the induced electric dipole is proportional to the polarizability of the particles and, therefore, will be maximize when the LSP condition is fulfilled. This dependence is consistent with the creation of a Lorentz force proportional to the polarizability of the nanoparticle. As a consequence of the angular shift, the polarization of the scattered light by the nanoparticle in the far field will present the same shift. Since \( \theta_K \) is a complex number, this translates into a rotation of the polarization plane and a change in the ellipticity state, being both effects amplified at the LSP wavelength.

**Figure 4.11** – Schematic of the MO effect induced by the Lorentz force in a metal nanoparticle.

In order to corroborate the origin of the MO enhancement, theoretical calculations were performed to reproduce the experimental results. Two different approaches were employed. Specifically, these are: a transfer matrix (TMM)
method together with an effective medium EM approximation (Maxwell-Garnett), where the nanodisks were modelled as ellipsoidal particles with similar diameters and heights, embedded in a dielectric medium average of the substrate glass and air; and a scattering matrix method (SMM), which allows an accurate representation of the nanodisks. Both formalisms present advantages and drawbacks. For example, the SMM method is able to reproduce the exact shape of the nanodisks and the interactions between close neighbours but at the same time it cannot take into account a random arrangement of nanoparticles. The effect is, however, small if the concentration is small as well, since the interactions play a minor role, and lattice effects due to the artificial periodicity are negligible. On the other hand, the EM approximation allows considering a random distribution of nanoparticles but it does not consider the possible interaction between them, and also their shape is restricted to ellipsoids. A detailed description of the different theoretical methods can be found in Appendixes 1, 2 and 3.

The optical constants of gold were experimentally obtained from ellipsometry measurements, whereas the MO constants were calculated by means of a Drude model approximation. The obtained spectra for the Kerr rotation and ellipticity respectively are plotted in Figure 4.12a and b. The cyan and blue lines represent the 70 and 120nm diameter Au nanodisks, whereas the continuous and dashed lines correspond to the SMM and effective medium methods, respectively.

![Figure 4.12](image)

**Figure 4.12** - (a) Theoretical Kerr Rotation spectra calculated for an array of Au nanodisks with 70nm (blue lines) and 120nm diameters (cyan lines). The continuous and dashed lines correspond to different theoretical calculations, SMM and MG respectively. The continuous black line corresponds to the MO response of an Au thin film 32nm height. (b) Theoretical Kerr Ellipticity spectra calculated for the same disk diameters as in (a).

As it can be observed, the theory reproduces qualitatively well the experimental results, although the intensity of the theoretical spectra is considerably larger. This may be ascribed to an inaccurate description of the gold optical constants, probably due to the higher damping in the experimental nanostructures caused by dephasing effects and surface roughness.
4.3 Enhanced MO activity in Au/Co/Au nanodisks

As it has been shown, noble metals exhibit a very weak MO activity that can be enhanced by exciting LSP resonances. Nevertheless, the enhancement of the MO activity is still too weak when compared to that of ferromagnetic metals, and thus presenting difficulties for developing MO active optical devices. On the other hand, ferromagnetic materials exhibit a too much broad plasmon resonance that is also not adequate for developing devices.

A way to overcome this problem is to combine both kinds of materials in a single nanostructure, keeping the amount of ferromagnetic material small enough to show a fairly good MO activity and a narrow LSP resonance. This is the purpose of the following section, presenting an improvement to the structures previously analyzed: the study of nanodisks made of gold and cobalt, i.e., Au/Co/Au nanosandwiches [17,18].

The choice of gold and cobalt as the materials to conform the nanostructures obeys to different reasons. First, as it was pointed at the beginning of section 4.2, silver has better plasmonic properties but on the other hand, it oxidizes easily. On the contrary, gold does not easily react with other substances and therefore it may be used as a protecting layer for the ferromagnetic material. Second, the best option for the ferromagnetic material should be iron, as it presents the highest saturation magnetization ($\sim 1.75 \times 10^6$ A/m at 0K) [19]. However, iron usually alloys with gold and so changing their optical and MO properties. Cobalt, on the other hand, does not alloy with gold and also presents a high saturation magnetization ($\sim 1.45 \times 10^6$ A/m at 0K). Moreover, the magnetic field needed to saturate it is lower (1.7Teslas) than that for iron (2.1Teslas), so that becoming a better candidate for our nanostructures.

4.3.1 Fabrication process, morphological and optical characterization

The nanodisks structures were obtained using the same fabrication process described in section 4.2.1. Through this method, series of Au/Co/Au nanodisks were obtained where the nanodisks diameter and Co amount was varied. Their total height was fixed again to 32nm. Thus, in order to get different Co amounts, the thicknesses grown correspond to 11nmAu-10nmCo-11nmAu, 13.5nmAu-5nmCo-13.5nmAu and 32nmAu, for a 30%, 15% and 0% Co concentration respectively. The nanodisks diameter was varied using two bead diameters of 60nm and 110nm.
Figure 4.13 - Experimental 3D AFM images of 11nmAu-10nmCo-11nmAu nanodisks obtained through hole-mask colloidal lithography for the (a) 60 nm and (b) 110 nm diameters respectively. It can be observed a clear reduction of the leftovers with respect to the etching fabrication process. The scheme shows the nominal diameters and Co amount within the nanodisks.

In Figure 4.13 an AFM image of the final outcome from the fabrication process can be seen. Figure 4.13a shows the 3D representation of the 60nm diameter nanodisks and 11nmAu-10nmCo-11nmAu thicknesses. The nanodisks present a cylinder-like shape despite some imperfections. It can be also observed that its height is different to the nominal one. These imperfections are ascribed again to the leftovers from the fabrication process. Figure 4.13b shows the same that in Figure 4.13a but for the 110nm diameter nanodisks and same thicknesses. By contrast, the nanodisks do not present a mountain-like shape, as the leftovers appear only at particular locations over the nanodisks. The nanodisks concentration is close to a 34%, and similar in all the samples.

Figure 4.14 - (a) Experimental normalized extinction spectra of Au/Co/Au nanodisks with diameter d=60nm. The blue line represent a 0% Co concentration in the nanodisks, the cyan line represent a 15% of Co amount, and the green line represent a 30% Co. (b) same as in (a) but for the d=1100nm diameter nanodisks.
The extinction measurements for these samples are depicted in Figure 4.14. Figure 4.14a shows the experimental normalized extinction for the 60nm diameter samples whereas Figure 4.14b is for the 110 nm diameter and three different Co amounts (the corresponding to the aforementioned 0, 5 and 10 nm Co thicknesses). As it can be observed, the extinction spectra of all the samples are characterized by a peak associated to the LSP resonance. In Figure 4.14 the LSP resonance peak lies around 2.0eV. The effect of the Co absorption can be observed in the slightly shift of the spectral position of the peak to higher energies as the amount of Co in the nanodisks is increased, and in the peak broadening. On the other hand, the extinction peak of the Au/Co/Au nanodisks shifts towards lower energies as we increase the disk diameter. This behaviour is similar to the one observed in pure noble metal nanoparticles, reflecting its plasmonic origin.

4.3.2 MO activity: Enhancement of the MO response.

The magnetic behaviour of the samples was studied by performing Polar Kerr loops at same light wavelength (λ=530nm). The results are depicted in Figure 4.15, showing a low remanence and magnetization saturation at 12000Oe approximately.

![Polar Kerr loops](image)

**Figure 4.15** – (a, b) Normalized Polar Kerr loops at λ=530nm for 11nm Au/10nm Co/11nm Au nanodisks as a function of the disk diameter (60nm and 110nm, respectively). (c, d) Same as in (a, b) but for 13.5nm Au/5nm Co/13.5nm Au nanodisks. These thicknesses correspond to a 30% and 15% of Co amount within the nanodisks.
The MO response of the samples was analyzed by measuring the Polar Kerr spectra. We will first focus on nanodisks of 60nm diameter. Figures 4.16a and b show the rotation and ellipticity spectra obtained for two different Co concentrations: 15% (blue triangles) and 30% (cyan triangles). Figure 4.16c and d shows the same spectra for the 110nm diameter nanodisks. It can be observed that the consequence of the excitation of the LSP is that the Kerr ellipticity presents a peak whereas the Kerr rotation an s-like shape. For comparison, we present in the Figure 4.16e and f the Kerr rotation and ellipticity spectra for the continuous Au/Co/Au trilayer system with identical Au and Co thicknesses (blue and cyan circles for the 15 and 30% Co concentrations).

**Figure 4.16** - (a,b) Experimental Kerr rotation and ellipticity spectra obtained for the 60nm diameter nanodisks as a function of the Co concentration. The blue and cyan triangles correspond to a 15% and a 30% of Co amount. (c,d) Same spectra as in (a,b) but obtained for the 110nm diameter nanodisks. (e,f) Same spectra as in (a,b) but obtained for the continuous trilayers [Thicknesses for a 15% Co: 13.5nmAu/5nmCo/13.5nmAu; Thicknesses for a 30% Co: 11nmAu/10nmCo/11nmAu].
As it can be observed, these spectra are completely different, manifesting the effect of the LSP in the MO response of the Au/Co/Au nanodisks. Furthermore, although the amount of Co in the continuous layer is almost 3 times higher than that of the nanoparticle layer, the magnitude of the MO effects are similar, indicating that the LSP induces a large enhancement of the MO activity.

Increasing the Co concentration leads not only to an enhancement of the MO activity but also to a broadening of these features, similarly to what it has been observed for the absorption spectra (see Figures 4.14a and b). The spectral behaviour upon increasing the Co concentration is a red-shift of the peak (s-structure) in the ellipticity (rotation). This is in contrast to the observed behaviour in the optical analysis, the difference coming from the spectral dependence of the MO constants.

To analyze this fact we used the effective medium (EM) together with the transfer matrix (TMM) method. The nanodisks were designed as an ellipsoidal Co core surrounded by an ellipsoidal Au shell with identical height and diameter of the nanodisks. The optical properties of Au and Co used in the calculations were obtained from continuous Au/Co/Au trilayers with identical thicknesses to that of the nanodisks: 11nmAu/10nmCo/11nmAu, and 13.5nmAu/5nmCo/13.5nmAu, respectively. On the other hand, the MO constants of Co were extracted using the process described at the end of Appendix 1, which requires the experimental results of the Polar Kerr rotation and ellipticity spectra measured from the continuous Au/Co/Au films. The real and imaginary parts of these MO constants are plotted in Figure 4.17a and b, showing a good agreement with the MO constants of Co bulk [20].

![Figure 4.17](image_url) - (a) Real and (b) imaginary parts of the MO dielectric constants ($\varepsilon_{xy}$) obtained for several Au/Co/Au trilayers. The thicknesses for trilayers B and C are 11nmAu/10nmCo/11nmAu, and 13.5nmAu/5nmCo/13.5nmAu. The line corresponds to the MO dielectric constants of Co bulk [20].
The Kerr rotation and ellipticity calculations are plotted in Figures 4.18a and b for the 60nm nanodisks diameters. Figure 4.18c and d shows the same results for the 110nm diameter nanodisks. For both nanodisks diameters, it can be observed that the theoretical curves reproduce the red-shift as the amount of Co increases and as the diameter of the nanodisks reduce and manifesting that the MO constants of Co have a large influence on the MO response of the nanodisks. On the other hand, the intensity of all the theoretical spectra is higher than the experimental ones. This effect may be attributed to an overestimation of the amount of Co inside the simulated nanoparticles, since Co is laterally exposed to air and, therefore, could be slightly oxidized. This effect has been demonstrated via exchange bias measurements [21]. In addition, the MO constants of the nanodisks and that used in the calculations could be slightly different, since they were obtained from continuous Au/Co/Au trilayers.

Figure 4.18 - (a,b) Theoretical (EM approximation + TMM) Kerr rotation and ellipticity spectra obtained for the 60nm diameter nanodisks as a function of the Co concentration. The blue and cyan triangles correspond to a 15% and a 30% of Co amount. (c,d) Same spectra as in (a,b) but obtained for the 110nm diameter nanodisks. [Thicknesses for a 15% Co: 13.5nmAu/5nmCo/13.5nmAu; Thicknesses for a 30% Co: 11nmAu/10nmCo/11nmAu].
4.4 Comparison between the MO activities of Au and Au/Co/Au nanodisks.

From the results analyzed in the previous sections for the pure Au and Au/Co/Au nanodisks, it is possible to establish a direct comparison of their MO responses. At first glance, two major differences can be appreciated. First, attending to the intensity of the rotation and ellipticity, it can be observed that the MO signal related to the Au nanodisks is two orders of magnitude smaller than that of the Au/Co/Au nanodisks. This is consistent with the fact that cobalt presents a much higher MO activity than gold and consequently the MO response is much larger. A more intriguing aspect is that the peak and s-like structures observed in the rotation and ellipticity spectra of the Au nanodisks, change to an s-like and a peak structures when considering the Au/Co/Au nanodisks. The origin of this change in the shapes of the spectra may be a consequence of the different physical mechanisms that give rise to the MO activity in gold and cobalt. Macroscopically, it has been shown that these mechanisms manifest themselves in the non-diagonal dielectric constants (\(\varepsilon_{xy}\)). We will thus now study how these constants affect the shape of the MO spectra.

![Diagram of a three layer system for which an analytical expression of the reflectivity coefficients can be achieved.](image)

**Figure 4.19** - Scheme of a three layer system for which an analytical expression of the reflectivity coefficients can be achieved.

To do so, we need to obtain information of the MO response of the system through the refractive index of the nanostructure. First, let us express the complex Kerr rotation in terms of the right and left-handed reflectivity coefficients (corresponding to the right and left-handed polarizations of circular light):

\[
\begin{align*}
\theta + i\varphi &= \frac{r_{ps}}{r_{pp}} = \frac{r^+ - r^-}{r^+ + r^-}, \\
&= \left\{ 
\begin{array}{l}
r^+ = r_{pp} + r_{ps} \\
r^- = r_{pp} - r_{ps}
\end{array}
\right.
\end{align*}
\]

[4.7]

Changing to this base provides the advantage of considering different refractive indexes for each polarization (as it was pointed out in Chapter 2) and thus incorporating the MO dielectric constants (\(\varepsilon^\pm = \varepsilon_{xx} \pm i\varepsilon_{xy}\)). The system is composed of three layers (see Figure 4.19), each one characterized by a refractive index. Note that the refractive index of the central layer is a function
of the optical and MO properties of the nanodisks $\varepsilon^\pm_{Au} = \varepsilon_{xx} \pm \varepsilon_{xy}$ and the surrounding medium $\varepsilon_d$. For very thin layers, the reflectivity coefficients for such a system can be analytically expressed in terms of the dielectric constants for each medium. Attending to expression [20] from reference [22], a simple manipulation to consider normal incidence gives:

$$r^\pm = \frac{n_s - n_0}{n_s + n_0} + \frac{4\pi i d n_0}{\lambda (n_s + n_0)^2} (\varepsilon_s - \varepsilon^\pm_1)$$  \[4.8\]

being $n_0 = \sqrt{\varepsilon_0}$ and $n_s = \sqrt{\varepsilon_s}$ the refractive indexes of each layer, $d$ the thickness of the central layer, $\lambda$ the light wavelength, and $\varepsilon_s$ and $\varepsilon^\pm_1$ are the dielectric constants for the substrate and central layers. Finally, replacing this expression into Equation 4.7, it is easy to observe that the complex Kerr rotation is proportional to the difference of the right and left-handed dielectric constants:

$$\theta + i \varphi = C(n_0, n_s, d, \lambda) (\varepsilon^+_1 - \varepsilon^-_1)$$  \[4.9\]

This expression provides a way to observe the relation between the MO dielectric constants and the shape of the MO spectra. The experimental difference of $(\varepsilon^+_1 - \varepsilon^-_1)$ was obtained by means of ellipsometry measurements performed over the Au and Au/Co/Au nanodisks (to obtain the optical constants) and from the Kerr rotation and ellipticity responses (to obtain $\varepsilon_{xy}$). As the real and imaginary parts of the dielectric constants are related by the Kramers-Kronig relations, the analysis can be restricted to one of them. For simplicity, we have chosen the imaginary part, since it is related to the absorption coefficient. Figures 4.20a and b show the results as a function of the Co amount within the 60 and 110nm diameter nanodisks.

The difference was normalized to the maximum value of the imaginary part of the dielectric constant when no magnetic field was applied ($\varepsilon''_0$). Moreover, the spectral dependence was referenced with respect to the frequency ($\omega_0$) at which the maximum of $\varepsilon''_0$ is observed for each sample, as the spectral position of the LSP resonance depends on the aspect ratio and composition of the nanodisks. From Figure 4.20a, the relative variations for the Au/Co/Au nanodisks are in the range of 0.5%, whereas in the case of the Au nanodisks the relative variations are much lower (in fact, this spectrum is multiplied by a factor of 50). Moreover, for the nanodisks containing Co, the spectra exhibit a peak structure, whereas for the nanodisks without Co the spectra have an s-like structure. For the 110nm diameter nanodisks (Figure 4.20b), a similar behavior is found although the relative difference is larger.
Figure 4.20 - Experimental relative difference of the right and left-handed imaginary parts of the dielectric constant. The spectra are normalized to the maximum value of the imaginary dielectric constant when no magnetic field is applied ($\varepsilon'''_0$). (a) Relative difference corresponding to an array of 60nm diameter Au/Co/Au nanodisks and several Co concentrations (from 0 to 30%). The spectrum obtained for the pure Au nanodisks has been multiplied by a factor of 50. (b) The same as in (a) but for 110nm diameter nanodisks.

The change in the shape of the spectra can be explained through a simple model system. Let’s consider a layer composed of spherical metallic nanoparticles embedded in a dielectric medium under the influence of a magnetic field. If the dimensions of the nanoparticles are much smaller than the wavelength of light, the optical properties of this layer can be expressed by means of an effective dielectric constant (see Appendix 3):

$$
\varepsilon^\pm_{ef} = \frac{\varepsilon_d}{1 \pm f \frac{2(\varepsilon^\pm_m - \varepsilon_d)}{\varepsilon^\pm_m + 2\varepsilon_d}} \approx \varepsilon_d \left(1 + 3f \frac{(\varepsilon^\pm_m - \varepsilon_d)}{\varepsilon^\pm_m + 2\varepsilon_d}\right)
$$

being $\varepsilon^\pm_m = \varepsilon_{xx} \pm \varepsilon_{xy}$ the dielectric constants for the right and left-handed circularly polarized light respectively of the metallic nanoparticle, $\varepsilon_d$ the dielectric constant of the surrounding medium, and $f$ the nanoparticles concentration. From [4.10], we can obtain an expression for the difference ($\varepsilon^+_1 - \varepsilon^-_1$). For simplicity, we restrict to the imaginary part:

$$
\text{Im}(\varepsilon^\pm_{ef}) = (\varepsilon^\pm_{ef})_{im} = 9\varepsilon_d^2 f \frac{(\varepsilon^\pm_{im})^2}{(\varepsilon^\pm_{im})^2 + (2 + \varepsilon^\pm_{re})^2}, \quad \left\{ \begin{array}{l} 
\varepsilon^\pm_{re} = \varepsilon_{re} \mp \varepsilon_{xy}^v \\
\varepsilon^\pm_{im} = \varepsilon_{im} \pm \varepsilon_{xy}^v 
\end{array} \right. \quad [4.11]
$$
yielding, subsequently:

\[
\left( \varepsilon_{ef}^+ - \varepsilon_{ef}^- \right)_{im} \approx 18 \varepsilon_d^2 f \left[ \frac{(2 \varepsilon_d + \varepsilon_{re})^2 - (\varepsilon_{im})^2}{(2 \varepsilon_d + \varepsilon_{re})^2 + (\varepsilon_{im})^2} \varepsilon_{re}^{xy} + 2 \varepsilon_{im} \left( 2 \varepsilon_d + \varepsilon_{re} \right) \varepsilon_{im}^{xy} \right]
\]

where \( \varepsilon_{re} \) (\( \varepsilon_{im} \)) and \( \varepsilon_{re}^{xy} \) (\( \varepsilon_{im}^{xy} \)) are the real (imaginary) optical and MO dielectric constants of metal respectively. If the metal has very low absorption losses (\( \varepsilon_{im} \ll \)), the differences between the effective dielectric constants of the left-handed and right-handed circularly polarized light may be expressed as:

\[
\left( \varepsilon_{ef}^+ - \varepsilon_{ef}^- \right)_{im} \approx 18 \varepsilon_d^2 f \left[ \frac{\varepsilon_{re}^{xy}}{(2 \varepsilon_d + \varepsilon_{re})^2} + \frac{2 \varepsilon_{im} \varepsilon_{im}^{xy}}{(2 \varepsilon_d + \varepsilon_{re})^3} \right]
\]

From this expression, it can be observed that if the imaginary part of the MO constant of the metal is much higher than the real part (\( \varepsilon_{re}^{xy} \ll \varepsilon_{im}^{xy} \)), the second term of the Equation [4.11] dominates and the difference (\( \varepsilon_1^+ - \varepsilon_1^- \)) has an s-shape structure (this term presents a change of sign in the denominator when approaching \( \varepsilon_{re} = -2 \varepsilon_d \), which is precisely the condition of LSP excitations). On the other hand, if the real part of the MO activity is much higher than the imaginary part (\( \varepsilon_{re}^{xy} \gg \varepsilon_{im}^{xy} \)), the first term of the expression dominates and the difference has a peak structure.

Therefore, the difference in shape has its origin in which part of the MO dielectric constant is dominant. Going back to the Au and Au/Co/Au systems, Figures 4.27a and b show the real (continuous line) and imaginary (dashed line) parts of the MO constants for cobalt (black) and gold (red) [Note: due to the diamagnetic behaviour of gold, the constants have been multiplied by -1 in order to keep positive the real part]. It can be observed that the real part of the non-diagonal constant of cobalt is much larger than the imaginary part. On the contrary, the behaviour found for the gold constants is the opposite.
Figure 4.21 - Real and imaginary parts of the MO dielectric constants of (a) cobalt and (b) gold. (c) Ratio of the imaginary vs. real part of the MO constants.

Figure 4.21c also shows the ratio between the imaginary and real parts of the MO constants for gold and cobalt. For the gold nanodisks, this ratio is $\varepsilon_{im}^{xy}(Au)/\varepsilon_{re}^{xy}(Au) \approx 12$ in the frequency region of the LSP excitation, therefore the expected spectra of the difference $(\varepsilon_1^+ - \varepsilon_1^-)$ should present an s-like shape, as observed. In the case of the Au/Co/Au, the behaviour of gold and cobalt is the opposite (for pure cobalt, the ratio shows that $(\varepsilon_{im}^{xy}(Co))/\varepsilon_{re}^{xy}(Co) < 1$, so the spectra should present a peak shape). Since the MO activity of cobalt is far more pronounced than that of gold, the MO response is totally governed by the ferromagnetic material, and thus presenting a peak.

In order to corroborate these results, theoretical calculations were performed for the difference $(\varepsilon_1^+ - \varepsilon_1^-)$ of the right and left-handed circularly polarized light. The results are presented in Figure 4.22, finding a good agreement between the experimental spectra and the calculations. The difference in intensity between the 60 and 110nm diameter nanodisks can be explained taking into account that the spectral location of the LSP resonance is localized at lower energies if the nanodisks diameter increases. Since the MO dielectric constants of the materials involved (see Figure 4.27) are larger for lower energies, this makes the MO activity to increase and therefore increasing the overall relative variation as well.
Figure 4.22 - Relative difference of the right and left-handed imaginary parts of the dielectric constant. The spectra are normalized to the maximum value of the imaginary dielectric constant when no magnetic field is applied ($\varepsilon_0$). (a) Relative difference corresponding to an array of 60nm diameter Au/Co/Au nanodisks and several Co concentrations (from 0 to 30%). The spectrum obtained for the pure Au nanodisks has been multiplied by a factor of 50. (b) The same as in (a) but for 110nm diameter nanodisks.

4.5 Conclusions

Along this chapter the influence of LSP excitations on the MO response of a series of nanostructures containing exclusively noble-metals (Au nanodisks) or a combination of these materials and ferromagnetic metals (Au/Co/Au nanodisks) has been studied. Such nanostructures have shown MO activity enhanced by LSPs. The results obtained point out that the optical and MO properties are governed by the LSP resonance of the nanoparticles and can be tuned by modifying the size, shape or amount of MO active material of the nanoparticles.

Focusing on the pure Au nanostructure, it has been shown that despite the weak MO activity gold presents, it can be enhanced by means of LSP resonances. This result indicates that the plasmonic excitation itself provides the stimulus necessary to increase the MO response of any metallic system. On the other hand, for the Au/Co/Au nanostructure, it has been shown that the enhancement of the electromagnetic field in the cobalt layer induced by the LSP makes the MO signal to present a large increase. Due to the presence of noble metals, it is also worth noticing a narrowing of the spectral region where the MO signal is enhanced due to the LSP, when compared to the Ni nanostructures. Moreover, the intensity of the MO signal is similar to that of pure ferromagnetic metals, despite the smaller amount of MO active material. As a result, these structures may become very suitable to develop new types of active optical devices based on LSP resonances. The extensive simulations performed using both the SMM method and the TMM method together with
EM approximation, have reproduced the experimental results qualitatively in both kinds of nanostructures.

Finally, when comparing the MO activity between the pure Au nanodisks and the Au/Co/Au nanodisks, it was found significant differences in the shape of the observed MO spectra. These differences are due to the different physical mechanisms that give rise to magnetism in gold and cobalt, and they were explained through a simple theoretical model. These results were confirmed by numerical simulations.

On the other hand, in the same way it has been proposed in Chapter 3, there are some open questions. For example, the fabrication process provides no arrangement to the nanodisks, and because of that the effect of the interaction between nanodisks on the MO response could not be analyzed. Therefore, other fabrication processes are being employed to produce samples in which the nanodisks are arranged in a periodic fashion, being thus able to set the distance between disks arbitrarily. Moreover, since the electromagnetic field distribution when exciting the LSP is not uniform, the MO response of the nanostructure may depend on the position of the Co layer within the Au/Co/Au nanodisk. At present, numerical simulations are ongoing to study this effect.
Chapter 5

Nonreciprocity effects in Combined Noble-Metal/Ferromagnet Layered Structures

5.1 Motivation

So far, the studies discussed along the previous chapters had two main characteristics in common. First, the nanostructures were composed of nanoparticles, allowing the study of surface plasmons in isolated structures. As a consequence, the plasmon resonances were localized. The excitation condition for this kind of plasmons requires the proper wavelength for the incident light, and therefore no dispersion relation is available. Second, the focus has been the influence of the surface plasmons on the MO activity. However, the connection between plasmons and magnetism is bidirectional, i.e., the magnetic field also affects the plasmon properties. In this chapter we will use propagating surface plasmons (SPPs) to address the enhancement of the MO response as well as the modification of the SPP dispersion relation when a magnetic field is applied.

Therefore, Chapter 5 is organized as follows. The first part will introduce the fundamentals properties of SPPs. Moreover, a brief description of the main excitation methods is explained. Thereafter, it will be studied the MO response of Au/Co/Au continuous trilayers. Once described the fabrication process, morphological and magnetic characterization, we will analyze the MO activity in presence and absence of SPP excitation. We will show that the origin of the enhanced MO activity is mainly governed by the optical response. Then we will demonstrate that the plasmon wave vector can be modulated by the application of a magnetic field in the transverse configuration (see Chapter 2).
5.2 Fundamentals of Propagating surface plasmons (SPPs)

As it was quoted in the introduction chapter of this thesis, surface plasmons are collective oscillations of the conduction electrons of a metal. In the particular case of propagating surface plasmons (SPPs), these modes take place when considering flat surfaces like those of continuous films. They are characterized as non-radiative propagating waves along the metal/dielectric interface, whose propagation length is controlled by the properties of the metal and the dielectric. In general, SSPs present propagation lengths between several hundred of nanometers and a few microns. Depending on the configuration of the layered system the SSPs present different properties. For simplicity, only the single interface and thin film cases are analyzed.

5.2.1 SPPs at a single metal/dielectric interface

The physical properties of SPPs can be obtained by applying the Maxwell’s equations and looking for evanescent solutions. The most important ones are the following. First, these modes can only be excited by a perpendicular to the surface electric field, which induces a surface charge density at the interface. Therefore, SPPs exist only for TM polarization, i.e., no surface modes exist for TE polarization. On the other hand, the confinement of the EM field requires that $\text{Re}[\epsilon_1] < 0$ if the dielectric constant of the second medium is positive $\epsilon_2 > 0$, so that SPPs exist only at interfaces between materials with opposite signs of the real part of their dielectric constants.

![Diagram of a surface plasmon propagating along a metal-dielectric interface.](image)

**Figure 5.1** - Scheme of a surface plasmon propagating along a metal-dielectric interface. The plasmon propagates along the x-direction and decays exponentially (in the z-direction) at both sides of the interface.

The dispersion relation of SPPs propagating at a single metal-insulator interface is [1]:

$$k_z = k_0 \sqrt{\frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2}}$$  \hspace{1cm} [5.1]
This expression may be represented graphically in order to study the wavelength dependence of the wave vector. To do so, a simple Au/Air interface is considered. The results are depicted in Figure 5.2. In (a) the real part of the wave vector appears, whereas (b) shows the imaginary part of the wave vector that is related to the propagation length of the SPP (defined as \( L = \frac{1}{2\Im \left(k_x\right)} \)).

![Graph](image)

**Figure 5.2** - Real (a) and imaginary (b) parts of the surface plasmon wave vector calculated for a single Au/Air. The continuous black line represents the corresponding light line for air.

The properties of the SPP modify as a function of the energy considered. For low energies, the real part of the wave vector is close to the light line whereas the imaginary part is small. Thus, SPP highly extends in the dielectric and propagates a long distance. It is worth to note that the dispersion relation lies at the right side of the light line at these energies. As a consequence, SPPs are non-radiative modes. This makes the excitation of SPPs by direct illumination to be impractical, requiring specific techniques that will be discussed at the end of this section. As the energy increases, the wave vector departs from the light line and increases its absorption. However, for certain energy the wave vector bends back, approaches and crosses the light line. This point indicates that the SPP has lost its plasmonic character and has become a radiative mode.

### 5.2.2 SPPs at thin film systems

If instead of a single interface, thin metallic films are considered, some differences are found. In principle, each metal/dielectric interface is able to sustain PSPs. However, when the separation between adjacent interfaces is comparable or smaller than the decay length of the modes, SPPs begin to interact and give rise to coupled modes. There are two different geometries that may present such mode coupling: IMI (insulator-thin metal film-insulator) or MIM (metal-thin film insulator-metal) structures. We restrict to the first ones, as these are the structures employed along this thesis. For simplicity, we start considering the same dielectric constants for the upper and bottom dielectric mediums.
Applying Maxwell’s equations and the proper continuity of the tangential fields, a pair of two implicit expressions for the dispersion relation is obtained:

\[
\left( \frac{k_{z_2}^2 + k_{z_1}^2}{\varepsilon_2} \right) = \left( \frac{k_{z_2}^2 - k_{z_1}^2}{\varepsilon_1} \right) e^{-2ik_d} \rightarrow \tanh k_{z_2}d = \frac{k_{z_2}e_1}{k_{z_1}e_2} \tag{5.2a}
\]

\[
\left( \frac{k_{z_2}^2 + k_{z_1}^2}{\varepsilon_2} \right) = -\left( \frac{k_{z_2}^2 - k_{z_1}^2}{\varepsilon_1} \right) e^{-2ik_d} \rightarrow \tanh k_{z_2}d = -\frac{k_{z_1}e_2}{k_{z_2}e_1} \tag{5.2b}
\]

, where \( d = z_2 - z_1 \) is the thickness of the metallic film. Equations 5.2 do not admit an explicit solution for the dispersion relation of the SPP wave vector and need to be numerically solved. [Note that for an infinite thickness this expression reduces to the equations of two uncoupled modes that can be regarded as single PSPs modes excited at independent metal/dielectric interfaces].

It can be shown that [5.2a and b] give rise to a mode with a symmetric (SM) and anti-symmetric (AM) electromagnetic field distribution respectively (see Figure 5.4a). These modes present different dispersion relations and behavior with respect to the thickness of the metallic film. For instance, the SM mode has the property that upon decreasing metal thickness, the confinement of the coupled mode to the metal decreases leading to an increased propagation distance. The AM mode shows the opposite behavior; their confinement to the metal increases when decreasing the metal thickness, resulting in a reduction of the propagation length. An example of the dispersion relations obtained for an Air-Au-Air layered system is depicted in Figure 5.3. The thickness of the Au layer was set to 70nm.

**Figure 5.3** - Real (a) and imaginary (b) parts of the surface plasmon wave vector calculated for a 70nm Au thin film with air on top and bottom as background medium. The blue and cyan lines account for the symmetric (SM) and anti-symmetric (AM) modes, respectively. The continuous black line represents the corresponding light line for air.
Figure 5.3a and b shows the real and imaginary parts of the SPP wave vector respectively for the SM mode (blue line) and the AM mode (cyan line). The continuous black line corresponds to the light line of air. For lower energies, the SM mode appears closer to the light line and presents a smaller imaginary part compared to the AM mode. This confirms that the SM mode presents a larger propagation length due to the confinement of the electromagnetic field mostly in the dielectric. Moreover, the dispersion relation of the SM mode does not present a sharp bend back as the AM does. On the contrary, it approaches gradually to the light line while increasing energy.

Figure 5.4 - (a) Magnetic field $H_y$ and intensity $|H_y|^2$ distribution for the odd and even plasmonic modes allowed in an insulator-metal-insulator (IMI) system. (b) Same as in (a) but in this case one of the insulators presents a different refractive index. (Note: the modes in (a,b) can also be found in metal-insulator-metal (MIM) systems)

If the refractive index of the dielectric mediums at each side of the metallic film is different, the plasmonic solution to the Maxwell’s equations presents a different form:

$$\left(\frac{k_{z2}}{\varepsilon_2} + \frac{k_{z1}}{\varepsilon_1}\right)\left(\frac{k_{z3}}{\varepsilon_3} + \frac{k_{z2}}{\varepsilon_2}\right) + \left(\frac{k_{z3}}{\varepsilon_3} - \frac{k_{z1}}{\varepsilon_1}\right)\left(\frac{k_{z3}}{\varepsilon_3} - \frac{k_{z2}}{\varepsilon_2}\right)e^{-4ikzd} = 0 \quad [5.3]$$

Two different modes arise from this expression, showing a modified behavior and electromagnetic field distribution respect to the previous SM and AM modes. In this case, it is no longer proper to label these modes “symmetric” and “anti-symmetric”. These are called the Low and High Index modes (LIM and HIM from now), depending on if the electromagnetic field is mainly concentrated at the interface with smaller or larger refractive index, respectively. Despite this difference, these modes present similar properties to the SM and AM modes previously analyzed. For example, the electromagnetic field of the LIM mode
extends more in the dielectric than in the metal, such as the SM mode does. The opposite behavior is found for the HIM mode, in analog to the AM mode. For comparison, a schematic field distribution of these modes has been also represented in Figure 5.4b.

**Figure 5.5** - Real (a) and imaginary (b) parts of the surface plasmon wave vector calculated for a 70nm Au thin film with air and glass as background mediums. The blue and cyan lines account for the low index (LIM) and high index (HIM) modes, respectively. The continuous and dashed black lines represent the corresponding light lines for air and glass.

The dispersion relations of these modes show more similarities to the SM and AM. They are depicted in Figure 5.5a and b for the same 70nm gold thin film with air and glass as the dielectric mediums. In (a) the real part of the wave vector for the LIM (blue line) and HIM (cyan line) modes is represented. The continuous and dashed black lines correspond to the light lines of air and glass respectively. The dispersion relation of each mode lies around each light line. This indicates that despite these are hybrid modes, they are closer to almost independent SPPs, which is the proper behavior if the thickness of the metallic layer increases. On the other hand, the imaginary part depicted in Figure 5.5b of the HIM mode is far larger than that of the LIM mode. As a consequence this mode propagates a shorter distance, similar to what is observed for the SM and AM modes.

### 5.2.3 Excitation methods of SPPs

There are several techniques that are used in the excitation of SPPs: for instance, those based on the impact of charged particles [2]. Others, like those used along this thesis, employ light in order to couple photons to the surface plasmon. However, the excitation of the SPPs with light is more difficult that it seems in theory. As it has been aforementioned, the dispersion relation of surface plasmons lies always at the right of the light line, whether metal/dielectric or dielectric/metal/dielectric systems are considered. This means that the wave vector of light is always smaller than that of SPPs and needs to be increased. As a consequence, excitation by direct light illumination is impractical and
therefore it is necessary the use of particular techniques. The most usual are the following:

![Figure 5.6 - Schemes of different techniques employed for the excitation of PSPs by light.](image)

**Kretschmann Configuration:** This technique (also called attenuated total internal reflection ATR) is employed for the excitation of surface plasmons in thin metallic films. The excitation method consists in attaching a semicircular prism to the bottom metal/dielectric interface of the system (see the scheme in Figure 5.6a). Its refractive index ($\varepsilon_p$) must present a larger dielectric constant than that of the upper dielectric medium ($\varepsilon_d$). Then, the angle of the incident light is increased to the critical value where total internal reflection occurs ($\theta_c = \arcsin(\sqrt{\varepsilon_d / \varepsilon_p})$). As it has been previously quoted, the wave vector of the surface plasmon is always larger than the x-component of the light wave vector. However, beyond this critical angle, the wave vector of light exceeds that of the surface plasmon, and the evanescent field from the bottom interface excites the surface plasmon at the upper interface.

\[
k_{sp} > k_x^d = k_0 \sqrt{\varepsilon_d \sin \theta}, \quad \forall \theta \\
k_{sp} < k_x^p = k_0 \sqrt{\varepsilon_p \sin \theta}, \quad \theta > \theta_c = \arcsin\left(\sqrt{\varepsilon_d / \varepsilon_p}\right)
\]

[5.4]

This method can only be applied to systems in which the overall thickness is small enough to assure that the evanescent electromagnetic field reaches the opposite metal/dielectric interface.

**Otto Configuration:** This is a similar technique in which the prism is separated from the metal film a small distance (see Figure 5.6b). The total internal reflection takes place at the prism/dielectric interface, and the SPPs are excited via the evanescent field again. This configuration is used when not considering thin metallic films or when direct contact with the metal is unwanted. It is worth to note that surface plasmons excited through both the Otto and Kretschmann configurations are leaky waves, i.e. they lose energy due to the leakage of radiation consequence of the de-excitation of the SPPs into light. The
origin of this behavior is found in the larger dielectric constant of the prism, which allows exciting the plasmon at the metal/dielectric interface but at the same time allows such de-excitation.

**Grating Coupler:** The difference in the wave vector of the photons and the SPPs can also be overcome by patterning a grating on top of the metallic surface. For simplicity let’s consider a simple one-dimensional grating of groves with lattice constant \( a \) (see Figure 5.6c). If the light wavelength is similar to the lattice constant of the grating, diffraction takes place. This makes the wave vector of light to increase an extra amount \( \Delta k \), which is given by:

\[
    k_x = k_0 \sqrt{\varepsilon_d} \sin \theta + \Delta k = k_{sp}, \quad \Delta k = \frac{2\pi m}{a}, m = \pm 1, \pm 2, \ldots \quad [5.5]
\]

, where \( m \) is the order of the diffracted beam. Therefore, controlling the light incident angle it is possible to excite the SPP at the metal/dielectric interface. The reverse process can also occur, i.e. the de-excitation of SPPs into light and radiate. For this technique, the design of the grating parameters is a critical point that may influence the strength of the light coupling to the SPPs or even their propagation direction.

### 5.3 MO activity in Au/Co/Au trilayers

In this section we analyze the MO response of a structure consisting in Au/Co/Au continuous trilayers. The reason for choosing these structures is the same that was pointed out in the previous chapter: the combination of noble-metals and ferromagnets may lead to an enhanced MO activity in a narrow spectral region. Several works have already shown the enhancement of the MO activity when exciting SPPs \([3,4,5]\). All of them described the increase in the MO response as a pure optical effect. However, we will show that the application of a magnetic field also modulates the SPP wave vector, and that this is responsible for the spectral shape of the TMOKE signal.

#### 5.3.1 Structure design and fabrication

The fabrication of the samples was performed by molecular beam epitaxy (MBE) of metals. The Co thickness was varied to analyze the effect of the amount of cobalt on the MO response and on the plasmon wave vector modulation. Complete details of the deposition conditions and optimization of the structure can be found in [6].
Table 5.1. - Nominal vs. X-Ray reflectometry measurements for the upper gold, middle cobalt and buffer gold thicknesses. In brackets, measured roughness for each layer.

As it was previously mentioned, the Kretschmann configuration uses evanescent waves to couple light to the SPP, imposing thus a limitation to the overall thickness of the trilayer. A large thickness would prevent the EM field to reach the interface where the SPP can be excited. Because of that, a rough estimation of the maximum thickness for good plasmon coupling is 40nm. This thickness decreases as the Co amount increases due to the higher absorption of cobalt with respect to gold. Another limit in the fabrication process comes from the thickness of the upper gold layer, which has to be thicker than 6nm. This obeys to the fact that a thinner layer may not prevent the cobalt layer from oxidation.

Figure 5.7. - AFM images of a 16nm Au/ 7.7nm Co/6nm Au trilayer after the fabrication process. (a) Topographic image (b) Roughness profile of the sample along the grey line in (a).

Taking into account all these restrictions to the parameters, the structure chosen for the Au/Co/Au trilayers consisted in a 16nm gold buffer layer and a 6nm gold upper layer. The thickness of the ferromagnetic cobalt layer was systematically varied between 0 and 10nm nominally. The actual thickness and
roughness of the trilayers were obtained by means of X-ray reflectometry measurements, which are presented in Table 5.1. An example of the sample roughness is shown in Figure 5.7, where the surface morphology of a 7.7nm Co Au/Co/Au trilayer has been obtained by atomic force microscopy (AFM). As it can be observed, the trilayer presents some corrugation with hills and valleys of several nanometres height. The average roughness of the samples is close to 1-2nm, similar to what it has been analyzed [6].

5.3.2 Optical Characterization. SPP vs. No SPP Excitation

To analyze the optical response, let us focus on a trilayer with a Co thickness of 5.5nm. Reflectivity measurements were performed in two different configurations, with and without SPP excitation. To excite the surface plasmon we used a Kretschmann configuration where the sample is optically interfaced to a half-cylinder glass prism by a matching index fluid, placing the glass substrate in contact with the prism and leaving the Au capping layer in contact with air. In this way it is possible to excite the low index SPP mode (see section 5.2) at the Au/Co/Au/glass structure with a maximum of the EM field at the Au/Air interface. To obtain comparative results without any SPP excitation, the signal was also measured with the same experimental geometry but without the glass prism.

Figure 5.8 - Experimental setup scheme used in the Transverse Kerr system for (a) the SPP excitation (Kretschmann configuration) and (b) for the no SPP excitation.

Figures 5.9a and b show the angular dependence of the optical response for an Au/Co/Au trilayer with 5.5nm of Co thickness with and without SPP excitation. In absence of plasmon excitation a monotonous evolution of the reflectivity curve can be observed. If the Kretschmann configuration is used, we can see that the reflectivity reaches an almost 100% for an angle close to 41°. This angle corresponds to the total internal reflection condition, which can only exist when the refractive index of the incident medium (glass) is larger than that of the final medium (air). Beyond this angle, the reflectivity shows a minimum
close to 44°. This is the characteristic dip that appears when the surface plasmon at the gold/air interface is excited.

![Experimental reflectivity curves corresponding to an Au/Co/Au trilayer with 5.5nm Co thickness without (a) and (b) with SPP excitation respectively.](image)

**Figure 5.9** - Experimental reflectivity curves corresponding to an Au/Co/Au trilayer with 5.5nm Co thickness without (a) and (b) with SPP excitation respectively.

To get a deeper insight of the SPP excitation we have calculated the EM field distribution |H_y|^2 in the system at different angles by means of a Transfer matrix method (see Appendix 1). The optical constants employed in the calculations were obtained experimentally through reflectometry measurements. The results are displayed in Figure 5.10, where it can be observed that in absence of plasmon excitation (Figure 5.10a, b, and c), the field intensity is almost independent on the incident angle. It is concentrated at the interface where the light comes in and decreases along the system. Moreover, it can be observed that the field intensity at the final medium (glass) is constant, which points out that the light at the end of the system is a propagating wave. In the Kretschmann configuration, when the incidence angle is 40.65°, close to the total internal reflection condition (Figure 5.10d), the field intensity distribution follows a similar behavior to the observed in absence of plasmon excitation. However, for larger angles of the incident light (figures 5.10e and f), this distribution is completely different. The EM field concentrates at the last (Au/Air) interface of the system instead of the first. It can also be observed that the field intensity exponentially decays at both sides of the interface, which is the expected behavior if a surface plasmon is excited.
Figure 5.10 – Calculated electromagnetic field distribution for a 16nm Au/5nm Co/6nm Au trilayer at different angles $40.65^\circ$, $42.55^\circ$, and $44.11^\circ$. The left panel is for the case without SPP excitation and the right panel is when SPP excitation takes place. The arrows point out the medium from which the light hits the sample.

The calculations of the EM field in the system of Au/Co/Au trilayers shows an interesting effect of the ferromagnetic material on the optical activity. The angle at which the minimum of the reflectivity is reached ($44.11^\circ$) has been related to the optimum excitation of the surface plasmon [1,2]. As a consequence, the EM field in the Au/Air interface should reach a maximum value. However, in the trilayer system this maximum is found at a slightly different angle as it can be observed in Figures 5.10e and f. The origin of this behavior lies in the role that absorption plays in the system. In systems composed of low absorption materials (for instance gold or silver), the decrease in reflectivity is mainly due to the
excitation of SPPs. As a result, the minimum of reflectivity and the maximum of the EM field are located at the same angle. In contrast, if the absorption of the system is large (because of cobalt in our case), the reflectivity decreases due to both absorption and the SPP excitation. Since absorption also depends on the incidence angle, the minimum of reflectivity and the maximum of the EM field appear at different angular positions.

5.3.3 MO response. Surface SPP vs. No SPP Excitation

Before obtaining the MO response of the Au/Co/Au trilayer system, we are going to perform a study of the magnetic behavior of the samples. Series of MO Kerr hysteresis loops were obtained through a Polar Kerr spectrograph at same light wavelength ($\lambda=530\text{nm}$). Figure 5.11 shows the results obtained for the samples fabricated. It can be observed that, for thin Co thicknesses (Figures 5.11a and b), the hysteresis loops present a large remanence and the magnetic field needed to saturate the samples is small (less than 500 Oe). This is a clear evidence of an easy axis of magnetization perpendicular to the sample plane. As we increase the Co thickness (Figures 5.11c, d, e, f, g, and h), the remanence decreases and the saturation field grows up to 16,000 Oe. The origin of such modification is the anisotropy change from perpendicular to parallel to the sample plane. As a result, these samples will be easily saturated if the magnetic field is applied parallel to the sample plane, which is precisely the orientation in the Transverse Kerr system.
Figure 5.11 - Polar Kerr loops at $\lambda=530\text{nm}$ for Au/Co/Au trilayers for (a) 0.9nm (b) 0.8nm, (c) 1.7nm, (d) 2.4nm, (e) 2.7nm, (f) 4.7nm, (g) 5.5nm, and (h) 7.7nm Co thickness. Note the evolution of the hysteresis loops as the anisotropy of the Co layer changes from out of plane to in plane when the Co thickness increases.

To analyze the MO activity of the Au/Co/Au trilayers, the samples were measured in and out of SPP excitation condition in the TMOKE setup (see Chapter 2). Figure 5.12 (blue dots) shows the experimental results in the absence of SPP excitation, that is, when the incident light comes from the gold/air interface (see experimental scheme in Figure 5.8). From (a) to (d) the experimental angular dependence of the reflectivity $R_{pp}$ in the demagnetized state and TMOKE signal for different Co amounts are displayed.
Figure 5.12 - Evolution of the reflectivity $R_{pp}$ (left column) and TMOKE signal (right column) as a function of the incidence angle and of the Co thickness in the trilayer samples under no SPP excitation conditions. The Co thicknesses are 1.7nm, 4.7nm, 5.5nm, and 7.7nm for (a), (b), (c), and (d), respectively. Cyan lines represent theoretical calculations obtained by means of the Transfer Matrix method.
For a particular sample, the variation of both $R_{pp}$ and TMOKE signal as a function of the incident angle $\vartheta$ shows a smooth featureless evolution. If the amount of Co inside the trilayer is then increased it can be observed that the $R_{pp}$ curve slightly modifies but its intensity remains still the same. On the other hand, the TMOKE signal presents an enhancement of the signal as the Co thickness increases. This behavior is fully compatible with the standard Transverse Kerr effect [7], in which its signal is proportional to the thickness of the MO active material. As the incident light can penetrate several nanometers through metallic layers, a thicker Co layer provides a large distribution of the EM field in the MO active material and thus increasing the transverse Kerr signal.

Next, the MO response of the samples was analyzed under surface plasmon excitation conditions. Figure 5.13 shows the experimental (dots) and theoretical (lines) angular dependence of $R_{pp}$ and TMOKE signal (left and right columns respectively). Again, from (a) to (d) the curves are displayed as a function of the Co thickness within the trilayer. The excitation of a surface plasmon manifests itself as a structure in both $R_{pp}$ and TMOKE for incident angles in the range of 40°-50° degrees.

As it was previously mentioned, the reflectivity reaches the total internal reflection condition around 41° degrees and then a minimum related to the plasmon excitation at 44° degrees approximately. It can be observed that the minimum of $R_{pp}$ does not behave proportionally to the Co thickness. As it increases, the angular position of the reflectivity shifts towards smaller angles. In contrast, the intensity of the $R_{pp}$ minimum decreases almost to 0 as the Co reaches 5.5nm and then increases again. It is also worth noticing that the reflectivity curve broadens as the Co thickness increases, as expected from the large absorption of the ferromagnetic metal.

The transverse Kerr signal is also very different with respect to the case without plasmon excitation. It can be observed an s-like structure whose intensity grows until 5.5nm of Co thickness, decreasing afterwards. The TMOKE signal shifts to smaller angles as a function of the Co amount. Its intensity is several orders of magnitude larger than without plasmon, emphasizing the effect of the surface plasmon on the enhancement of the MO activity. Comparing the optical and the MO results, it can be noticed that the maximum of the MO activity coincides with the minimum of the reflectivity, pointing out that the enhancement of the MO activity is related to a pure optical effect.
Figure 5.13 - Evolution of the reflectivity $R_{pp}$ (left column) and transverse Kerr signal (right column) as a function of the incidence angle and of the Co thickness in the trilayer samples under SPP excitation conditions. The Co thicknesses are 1.7nm, 4.7nm, 5.5nm, and 7.7nm for (a), (b), (c), and (d), respectively. Red lines represent theoretical calculations obtained by means of the Transfer Matrix method.
Introducing this layer to the program based in the transfer matrix method and employing the experimental thicknesses obtained for each layer by means of X-ray reflectometry, the optical and MO responses of the complete set of trilayers were calculated considering the no SPP and SPP excitation configurations, although some differences are appreciated especially when the Co thickness is close to 5.5nm. Thus, despite that the theoretical model does not reproduce exactly the experimental results, it is possible to employ this method in order to study the evolution of the MO activity and compare it to the case of no SPP excitation.

**Figure 5.14** - Experimental (dots) and theoretical (solid line) values of the Transverse Kerr signal intensity as a function of the Co layer thickness, without (a) and with (b) SPP excitation.

The comparison of the TMOKE signal with and without SPP excitation was performed by considering the maximum positive value of the MO activity from each sample. In Figure 5.14, the TMOKE signal without (a) and with (b) SPP excitation are displayed as a function of Co thickness in the range 0–10 nm. The experimental results appear as red dots. In the absence of SPP excitation, an almost monotonic increase in the MO signal for the range of Co thicknesses considered is found. Actually, this curve tends towards a limit value, since the incident light cannot penetrate deeper in the material due to absorption. In contrast, when SPPs are excited, the MO activity exhibits a very large enhancement as the Co thickness approaches to 6 nm.

In order to understand the evolution of the MO activity, the experimental results were compared with theoretical simulations performed by means of a transfer matrix method. The experimental thicknesses, optical and MO constants employed in the calculations were obtained through X-ray reflectometry and ellipsometry measurements. The results are plotted as cyan lines in Figures 5.12, 5.13 and 5.14. As it can be observed, there is a good agreement between theory and experiment.
The non-linearity presented by the transverse Kerr signal due to the SPP excitation may be further analyzed by obtaining a series of contour maps as a function of the light incidence angle and the Co thickness in the Au/Co/Au trilayer. In Figure 5.15 the results for (a) the optical reflectivity $R_{pp}$, (b) the variation of reflectivity with the magnetic field, which is defined as $\Delta R_{pp} = R_{pp}(+H) - R_{pp}(-H)$, (c) the TMOKE signal, and (d) the EM field intensity at the Au/Air interface $|H_y|^2$ are plotted. From the reflectivity map (Figure 5.15a), the total internal condition can be observed as a maximum. It tends to disappear when the thickness increases over 20nm, verifying that the high Co absorption prevents light from reaching the Au/Air interface. The angle at which this condition occurs is independent from the Co amount. On the other hand, the minimum of the reflectivity appears at larger angles, its angular position depending on the Co thickness. From small to large Co amounts, the $R_{pp}$ minimum location evolves from 45° to 43° degrees.

The transverse Kerr signal (Figure 5.15c) exhibits an extremely sharp dependence on both Co thickness and incidence angle. It reaches its maximum value at 44.1° and 6.1nm of Co thickness, which are precisely the values at where the minimum of $R_{pp}$ is reached. This result confirms the hypothesis that the enhancement of the MO response is mainly governed by the optical response.

On the other hand, the variation of reflectivity with the magnetic field $\Delta R_{pp}$ (Figure 5.15b) exhibits a very weak dependence on the Co thickness. The maximum of $\Delta R_{pp}$ slightly shifts to a lower angle when the Co thickness increases. The largest value of $\Delta R_{pp}$ is reached at an incidence angle of 42.7 deg and a Co thickness of 9.9 nm, far from the values at which the largest TMOKE is obtained.
Finally, attending to Figure 5.15d, it is possible to observe the effect of Co on the EM field intensity at the Au/Air interface. The maximum field intensity is observed when there is no MO active material in the trilayer, that is, for a pure gold system. This effect is mainly due to the increasing absorption of the system provided by the ferromagnetic material. For a specific Co thickness, the angle at which the maximum field intensity is reached does not coincide with the minimum of $R_{pp}$, pointing to that this minimum is due to the combination of the SPP excitation and Co absorption. However, comparing Figures 5.15b and d, it is worth to notice that the maximum of the EM field matches that of $\Delta R_{pp}$ for every Co thickness. Such correlation reveals that the variation of reflectivity is enhanced when the SPP is excited.
5.3.4 Magnetic modulation of the SPP resonance. $\Delta k_x$ Mechanism

We have just shown that the MO response of the trilayers is not only merely an optical effect but that the modification of the SPP wave vector due to the application of an external magnetic field plays a key role. In simple structures such as dielectric/ferromagnet and dielectric/ferromagnet/dielectric layers it is possible to obtain close expressions for this modification [8]. In the case of the Au/Co/Au trilayer, a simple closed-form expression of the magnetic field dependence of the SPP wave vector is not easy to obtain. However, a reasonably accurate representation can be obtained by considering a dielectric/metal structure in which a MO active thin film of thickness $h$ is embedded in the metal and at a distance $d$ (see Figure 5.16).

![Figure 5.16](image)

**Figure 5.16** - Scheme of the structure employed on the analytical deduction.

To obtain the modification of the plasmon wave vector we proceed in the same way as to obtain the plasmon dispersion relation (see Appendix 4). The non-zero solution to this system of equations can be obtained if the determinant of the matrix of coefficients is zero. Through this determinant, it is possible to derive an analytical expression for the SPP wave vector after assuming some approximations.

$$k_x = k_{x0} + \delta k_O + \delta k_{MO}$$  \[5.1\]

where $k_{x0}$ is the SPP wave vector for a pure metal/dielectric interface, and $\delta k_O$ and $\delta k_{MO}$ are the modifications of the SPP wave vector due to the insertion of the MO active thin film.

$$\delta k_O = \frac{ie^2 k_{z3} \left( k_{x0}^2 - k_0^2 \varepsilon_1 \right)}{k_{x0} \left( \varepsilon_1^2 - \varepsilon_2^2 \right)} \left( \frac{k_{z2}}{\varepsilon_2} \right)^2 - \left( \frac{k_{z3}}{\varepsilon_3} \right)^2 - h e^{2ik_{z3}d}$$  \[5.2a\]
\[ \delta k_{MO} = -\frac{2\varepsilon_2^2 (k_{\times 0}^2 - k_0^2 \varepsilon_1)}{\varepsilon_1^2 - \varepsilon_2^2} i\varepsilon_3^{-1} e^{2ik_{\times d}} \]  

[5.2b]

being \(d\) the distance and metal thickness between the interface and the MO active film, \(h\) the thin film thickness, \(\varepsilon_j\) the diagonal dielectric constants of medium \(j\), \(k_0\) the light wave vector, \(k_{\times 0}\) the SPP wave vector for a pure metal/dielectric interface, and \(k_{\times j}\) accounts for the \(z\)-component of the wave vector in each medium. It can be observed that both expressions depend linearly on the thickness of the MO active layer and exponentially on the distance to the metal/dielectric interface.

The properties of the two different contributions to the SPP wave vector are very different. \(\delta k_{\times j}\) is independent from the MO character of the layer. It only affects the wave vector if the refractive indexes of the metal and the thin film are different. Because of that, this contribution accounts for a purely optical variation of the SPP wave vector. On the other hand, \(\delta k_{\times 0}\) is proportional to the non-diagonal dielectric constant of the MO active material \(\varepsilon_3^{xx}\), and disappears for non-magnetic materials. Therefore, this expression accounts for a MO variation of the SPP wave vector.

If the magnetization direction is reversed, the SPP wave vector modification will be given by the difference between \(k_{\times}^+ - k_{\times}^-\). As \(k_{\times 0}\) and \(\delta k_{\times 0}\) remains the same for any magnetic field, this difference becomes:

\[ \Delta k_{\times} = 2\delta_{MO} = \frac{4\varepsilon_2^2 d_1 (k_{\times 0}^2 - k_0^2 \varepsilon_1)}{\varepsilon_1^2 - \varepsilon_2^2} i\varepsilon_3^{-1} e^{2i\alpha_{\times d}} \]  

[5.3]

This expression represents the modulation of the plasmon wave vector due to the presence of a magnetic field. Once this modulation has been deduced, we can now analyze its effect in the Transverse Kerr signal. The condition for the SPP excitation by light using the Kretschmann configuration must fulfill the following expression:

\[ k_\times = k_0 \sin \theta \]  

[5.4]

where \(k_0\) is the wave vector of the light and \(\theta\) its incident angle. If a magnetic field is applied, the SPP wave vector changes a quantity given by Equation [5.2a] and as a consequence the angle at which the SPP excitation is reached changes as well (see Figure 5.17).
As the wave vector modification is very small, this is equivalent to differentiate Equation [5.4] with respect to $\theta$, which gives rise to:

$$\frac{\partial k_x}{\partial \theta} = k_0 \cos \theta \rightarrow \Delta k_x = k_0 \Delta \theta \cos \theta; \quad \Delta \theta = \frac{\Delta k_x}{k_0 \cos \theta} \quad [6.5]$$

The angular shift $\Delta \theta$ obtained is then proportional to the wave vector variation of the SPP excitation condition induced by the magnetization inversion. Since the angular shift of the plasmonic excitation is the responsible of the variation of reflectivity $\Delta R_{pp} = R_{pp}(+H) - R_{pp}(-H)$ with the magnetic field, $\Delta R_{pp}$ can also be expressed as the derivative of the reflectivity $R_{pp}$ with respect to the light incident angle $\theta$:

$$TMOKE = \frac{R_{pp}(+H) - R_{pp}(-H)}{R_{pp}(+H) + R_{pp}(-H)} \approx \frac{\Delta R_{pp}}{2R_{pp}(0)} = \frac{\Delta \theta}{2R_{pp}} \frac{\Delta \theta}{\partial \theta} 2R_{pp} \quad [6.6]$$

To illustrate this behavior, Figure 5.18 shows the experimental angular dependence of TMOKE (blue dots) and $(\partial R_{pp}/\partial \theta)(\Delta \theta/2R_{pp})$ (cyan circles) for different samples with different Co thicknesses: (a) 1.7 nm, (b) 4.7 nm, (c) 5.5 nm, and (d) 7.7 nm. Both curves appear nearly identical in the region of SPP excitation, showing a good agreement in shape and in intensity. On the contrary, the derivative $(\partial R_{pp}/\partial \theta)$ close to the total internal reflection condition generates a very intense dip, which does not show any correlation with the TMOKE angular dependence.
Figure 5.18 - TMOKE signal (blue dots) and the derivative of the reflectivity (cyan circles) for different samples with (a) 1.7nm, (b) 4.7nm, (c) 5.5nm, and (d) 7.7nm Co thicknesses. The sharp dip observed in the derivative of the reflectivity at 40 deg corresponds to the onset of total internal reflection.

Equation [6.6] also tells us that the shape of the TMOKE signal is provided by the angular derivative of $R_{pp}$, whereas the MO enhancement comes from the ratio between the angular modulation and the optical reduction of $R_{pp}$. The experimental values of $\Delta \theta$ were obtained from adjusting the amplitude of $(\partial R_{pp}/\partial \theta)/R_{pp}$ to that of TMOKE. This angular shift is presented in Figure 5.19 as a function of Co thickness (circles) together with the theoretical dependence (lines) that was calculated from the theoretical curves of $R_{pp}$ for the two opposite magnetization directions.

Figure 5.19 - Angular shift of the minimum in reflectivity as a function of Co thickness. The circles correspond to the experimental values of $\Delta \theta$ whereas the line represents a theoretical calculation.
5.4 Conclusions and Outlook

This chapter has been devoted to the study of the optical and MO responses of a continuous film made of combined noble metal and ferromagnetic materials (Au/Co/Au trilayers). In such structures it has been observed a strong increase of magnitude of the TMOKE signal when the surface plasmon is excited. This effect is observed at low magnetic fields and depends on the magnetization of the Co layer and can be tuned by varying the Co layer thickness. It does not have a monotonic increase as we increase the Co thickness, but reaches a maximum very close to the observed value for optimum plasmonic excitation and then decreases again, exhibiting a sharp dependence on the incidence angle and on the CO thickness.

It has been also presented experimental evidence of SPP wave vector modulation due to the application of a magnetic field. This result proved that the external magnetic field not only acts on the MO activity of the structure, but also on the plasmon excitation itself. It was also found that this is the last responsible of the Transverse Kerr effect, being the optical reduction coming from the SPP excitation responsible of its intensity.

The applications of these results may be multiple. For example, the ability to modulate the SPP reflectivity signal by means of an external magnetic field could lead to an enhanced sensitivity and to improved sensitivity biosensors. In that sense, a new MO surface plasmon resonance biosensor is being developed [9]. Moreover, novel structures are being considered in order to improve the MO response, like Ag/Co/Ag layered systems, which present a narrower plasmon resonance [10]. Finally, the modulation of the SPP wave vector is being used in plasmonic interferometers, in order to control its response by means of magnetic fields [11].
Appendix 1

The Transfer Matrix Method (TMM)

In this appendix we describe the numerical method employed to calculate the MO activity of systems involving only continuous layers. This is the so-called Transfer Matrix method (TMM) [1,2].

Let’s consider an incident wave impinging at an angle $\Phi_a$ over a multilayered system with parallel interfaces. It can be described by its p and s polarizations ($A_p$ and $A_s$) and wave vector $k_w$. Then the p and s components of the reflected ($B_p$ and $B_s$) and transmitted ($C_p$ and $C_s$) waves can be related in terms of the Transfer matrix:

$$
\begin{pmatrix}
A^0_p \\
B^0_s \\
A^0_s \\
B^0_p
\end{pmatrix}
= T
\begin{pmatrix}
C^0_p \\
0 \\
C^0_s \\
0
\end{pmatrix}
\Rightarrow
\begin{pmatrix}
T_{11} & T_{12} & T_{13} & T_{14} \\
T_{21} & T_{22} & T_{23} & T_{24} \\
T_{31} & T_{32} & T_{33} & T_{34} \\
T_{41} & T_{42} & T_{43} & T_{44}
\end{pmatrix}
$$

From this Transfer matrix it is easy to obtain the reflection and transmission coefficients of the system under consideration. For instance, to obtain the complex Kerr rotation as it has been defined in Chapter 2:

$$
r_{ps} = \frac{B_s}{A_p}_{A_p=0} = \frac{T_{13}T_{21} - T_{11}T_{23}}{T_{13}T_{31} - T_{11}T_{33}}
$$
\[ r_{pp} = \left( \frac{B_p}{A_p} \right)_{A_p = 0} = \frac{T_{13} T_{41} - T_{11} T_{43}}{T_{13} T_{31} - T_{11} T_{33}} \]  

[A1.2b]

The only step missing is the calculation of the Transfer matrix itself. Let’s express the Maxwell equations in terms of the in-plane components of the electric and magnetic field:

\[ \partial_z \Psi(z) = i k_0 \tilde{\Delta} \Psi(z), \quad \Psi(z) = \left( E_x, E_y, H_x, H_y \right)^T (z) \]  

[A1.3]

where we have assumed harmonic time dependence for the electromagnetic fields and also a homogeneous non-diagonal dielectric tensor (containing the polar and transversal MO dielectric constants), being the \( \tilde{\Delta} \) matrix defined as:

\[ \tilde{\Delta} = \left( \begin{array}{cccc}
    \frac{k_x}{\varepsilon_{xz}} & 0 & 0 & 1 - \frac{k_x^2}{\varepsilon_{zz}} \\
    0 & -1 & 0 & 0 \\
    \varepsilon_{xy} & k_x^2 - \varepsilon_{xx} & 0 & 0 \\
    \varepsilon_{xx} + \frac{k_x^2}{\varepsilon_{zz}} & \varepsilon_{xy} & 0 & -k_x \frac{\varepsilon_{xz}}{\varepsilon_{zz}} \\
\end{array} \right) \]  

[A1.4]

Now we need to use the matching conditions at each interface:

\[ \Psi(z + d) = e^{io\Delta d/c} \Psi(z) = T_p \Psi(z) \]  

[A1.5]

The matrix \( T_p \) relates the in-plane components of the electric and magnetic fields at a given interface. Applying the Cayley-Hamilton theorem [3], \( T_p \) can be expressed in a series expansion up to the power of \( n-1 \), being \( n \) the rank of the matrix:

\[ T_p = e^{io\Delta d/c} = \beta_0 \tilde{I} + \beta_1 \tilde{\Delta} + \beta_2 \tilde{\Delta}^2 + \beta_3 \tilde{\Delta}^3 \]  

[A1.6]

where \( \beta_i \) must fulfill the following condition:

\[ e^{ioQ_kd/c} = \sum_{j=0}^{3} \beta_j Q^j_k, \quad k = 1, \ldots, 4 \]  

[A1.7]
being \( q_i \) the eigenvalues of the matrix \( \tilde{\Delta} \). Therefore, the total transfer matrix \( T \) can be constructed simply by a sequential multiplication of the partial transfer matrices at each interface:

\[
\begin{pmatrix}
A^i_s \\
B^s_p \\
A^i_p \\
B^p_p
\end{pmatrix}
= T^{i+1} \begin{pmatrix}
C^i_s \\
D^i_s \\
A^0_p \\
B^0_p \\
C^i_p \\
D^i_p
\end{pmatrix}
= T_{p}^{0,1} \begin{pmatrix}
A^0_s \\
B^0_s \\
A^0_p \\
B^0_p
\end{pmatrix}
= T_{p}^{0,1} T_{p}^{1,2} = \ldots = T_{p}^{0,1} \ldots T_{p}^{n-1,n}
\]

\[
T = L_{a}^{1} \left[ \prod_{i=1}^{n} T_{p} \right] L_{f} \quad [A1.8]
\]

where \( L_{a} \) and \( L_{f} \) are the projection of the in-plane components of the incident/reflected and transmitted waves through to the first and last interfaces:

\[
L_{a}^{1} = \frac{1}{2} \begin{pmatrix}
0 & 1 & -n_{a}^{-1} \cos^{-1} \Phi_{a} & 0 \\
0 & 1 & n_{a}^{-1} \cos^{-1} \Phi_{a} & 0 \\
\cos^{-1} \Phi_{a} & 0 & 0 & n_{a}^{-1} \\
-\cos^{-1} \Phi_{a} & 0 & 0 & n_{a}^{-1}
\end{pmatrix} \quad [A1.9]
\]

\[
L_{f} = \begin{pmatrix}
0 & 0 & \cos \Phi_{f} & 0 \\
1 & 0 & 0 & 0 \\
-n_{f} \cos \Phi_{f} & 0 & 0 & 0 \\
0 & 0 & n_{f} & 0
\end{pmatrix} \quad [A1.10]
\]

A particular application of the transfer matrix algorithm is the determination of the MO constants of a magnetic layer. To do so, it is necessary to know the optical dielectric constants of each layer and the experimental rotation and ellipticity spectra of the system. The determination is based in the fact that, in a first approximation, the MO response of the layer system is linearly proportional to the real and imaginary parts of the MO constants, i.e., the MO activity can be expressed as:

\[
\theta_{k}(\lambda) = A(\lambda) \varepsilon_{xy}^{-}(\lambda) + B(\lambda) \varepsilon_{xy}^{+}(\lambda) \quad [A1.11a]
\]

\[
\phi_{k}(\lambda) = C(\lambda) \varepsilon_{xy}^{-}(\lambda) + D(\lambda) \varepsilon_{xy}^{+}(\lambda) \quad [A1.11b]
\]

where the factors \( A, B, C \) and \( D \) are wavelength dependent and express the contribution of the real and imaginary parts of \( \varepsilon_{xy} \) to the rotation and ellipticity.
These expressions represent a system of equations with two unknowns, which may be solved if the latter factors are inferred.

It is worth noticing that this approximation is valid only if these factors are small enough. One way to demonstrate the linearity is to calculate several MO responses of the system, in which the MO constants are: exclusively real ($\varepsilon_{xy} = 0.1$), exclusively imaginary ($\varepsilon_{xy} = 0.1i$), or a combination of them ($\varepsilon_{xy} = 0.1 + 0.1i$). If the sum of the two first MO responses is equal to that of the last one, then the approximation is valid. If not, it indicates that the nonlinear terms are important.

**Figure A1.2** - Calculated (a) Kerr rotation and (b) ellipticity spectra for an 11nmAu/10nmCo/11nmAu trilayer system and considering the following MO constants of cobalt: $\varepsilon_{xy} = 0.1$ (black dots), $\varepsilon_{xy} = 0.1i$ (red dots), $\varepsilon_{xy} = 0.1 + 0.1i$ (blue open circles). The line represents the sum of the two first spectra.

An example can be seen in **Figure A1.2**, where we show the calculated Kerr rotation and ellipticity spectra of an 11nmAu/10nmCo/11nmAu thick trilayer system considering the following MO constants of cobalt: (a) $\varepsilon_{xy} = 0.1$ (black dots), (b) $\varepsilon_{xy} = 0.1i$ (red dots), and (c) $\varepsilon_{xy} = 0.1 + 0.1i$ (blue open circles). The line represents the sum of the spectra for the exclusive real and imaginary MO constants. It can be observed that the linear dependence of the rotation and ellipticity on the real and imaginary parts of $\varepsilon_{xy}$ is verified.

Under these conditions we have a way to obtain the spectral dependence of the A, B, C and D factors. As they depend on the structure, but not in the MO activity, they may be obtained through the calculated spectra for the $\varepsilon_{xy} = 0.1$ and $\varepsilon_{xy} = 0.1i$ MO constants:

$$A(\lambda) = \frac{\theta_k(\varepsilon_{xy} = 0.1)}{0.1} \quad \text{[A1.12a]}$$

$$B(\lambda) = \frac{\theta_k(\varepsilon_{xy} = 0.1i)}{0.1} \quad \text{[A1.12b]}$$

$$C(\lambda) = \frac{\phi_k(\varepsilon_{xy} = 0.1)}{0.1} \quad \text{[A1.12c]}$$

$$D(\lambda) = \frac{\phi_k(\varepsilon_{xy} = 0.1i)}{0.1} \quad \text{[A1.12d]}$$
Finally, once these factors have been determined, it is straightforward to obtain the real and imaginary parts of the MO dielectric constants using Equation [A1.11].

\[
\varepsilon_{xy}^r(\lambda) = \frac{B\varphi_k - D\theta_k}{BC - AD} \quad \text{[A1.13a]}
\]

\[
\varepsilon_{xy}^i(\lambda) = \frac{C\theta_k - A\varphi_k}{BC - AD} \quad \text{[A1.13b]}
\]
Appendix 2

The Scattering Matrix Method (SMM)

The experimental results that have been obtained along this thesis come from systems composed of a specific number of layers, whether they are nanostructured or not. In order to perform a theoretical comparison respect to these results, it is necessary a computational tool able to calculate the electromagnetic fields in each one of the system layers. Among the wide range of algorithms available, our choice is the Scattering Matrix method (SMM).

This method presents the advantage of being more stable with respect to other methods like those based in transfer matrices (see Appendix A1) [1]. For the particular requirements of this thesis, the SMM method was applied to systems in which some or all of the layers are periodically nanostructured, i.e., the nanoparticles are perfectly ordered. To do so, it was followed the formalism presented by Whittaker and Culshaw [2] based in the earlier work of Ko and Inkson [1]. Moreover, this formalism was adapted to consider MO effects in the polar configuration, that is, when the magnetic field is applied perpendicular to the sample plane [3], in order to obtain the MO response of the system.

![Diagram of the scattering matrix method](image)

**Figure A2.1** - Scheme of the SMM method. The S-matrix relates the incoming waves to the outgoing waves of a layered system, and from which it is possible to obtain the optical and MO responses.

For a layered system (see Figure A2.1), the SMM method consists in relating the outgoing waves to the incoming waves of such system. Mathematically, this relation is expressed as:

\[
\begin{pmatrix}
a_n \\
b_0
\end{pmatrix} = \mathbf{S}(0,n) \begin{pmatrix}
a_0 \\
b_n
\end{pmatrix} = \begin{pmatrix}
S_{11} & S_{12} \\
S_{21} & S_{22}
\end{pmatrix} \begin{pmatrix}
a_0 \\
b_n
\end{pmatrix}
\]

[A2.1]
The S-matrix \( S(0,n) \) relates the vectors of the amplitudes of forward \( a_i \) and backward \( b_i \) going waves between the initial and final layers of the structure. It contains all the information of the system, from which it is possible to obtain its optical and MO responses.

The coefficients \( S_{ij} \) of the S-matrix can be obtained from the matching conditions for the wave functions and derivatives at the \((n+1)\)th interface, conditions that can be expressed in the following matrix notation:

\[
\begin{pmatrix}
  a_n \\
  b_n
\end{pmatrix} = I(n,n+1)
\begin{pmatrix}
  a_{n+1} \\
  b_{n+1}
\end{pmatrix} =
\begin{pmatrix}
  I_{11} & I_{12} \\
  I_{21} & I_{22}
\end{pmatrix}
\begin{pmatrix}
  a_{n+1} \\
  b_{n+1}
\end{pmatrix}
\]

where \( I(n,n+1) \) is the matrix representing the interfacial coupling. This equation may be used to replace the \( a_n \) and \( b_n \) coefficients from Equation (AX1.1), yielding a new S-matrix for the \((n+1)\)th layer.

\[
\begin{pmatrix}
  a_{n+1} \\
  b_0
\end{pmatrix} = \begin{pmatrix}
  -S_{22} I_{21} & 1 \\
  I_{11} - S_{12} I_{21} & 0
\end{pmatrix}^{-1}
\begin{pmatrix}
  S_{21} & S_{22} I_{22} \\
  S_{11} & S_{12} I_{22} - I_{12}
\end{pmatrix}
\begin{pmatrix}
  a_0 \\
  b_{n+1}
\end{pmatrix}
\]

Therefore, through an iterative process it is possible to calculate the \( S(0,n) \) matrix by consecutively calculating \( S(0,1), S(0,2), \ldots, S(0,n-1) \) [Note that \( S(0,0)=I \)]. This iterative process is not as easy to use as the common transfer matrix methods, which consists in a simple product of matrices, but the enhancement of the stability compensates the increasing computing time.

The calculation of the \( I_{ij} \) coefficients first needs the knowledge of the electromagnetic fields in each layer of the system. If the layer is homogeneous this is straightforward, as they may be described as a linear combination of a single forward and a backward wave. However, for patterned layers these fields are more difficult to obtain. In order to do so, let’s consider the Maxwell’s equations in absence of electric currents, where harmonic time dependence has been supposed:

\[
\nabla \cdot \mathbf{\varepsilon}_0 \mathbf{E} = 0 \quad \left[ A2.4a \right] \quad \nabla \cdot \mathbf{H} = 0 \quad \left[ A2.4b \right]
\]

\[
\nabla \times \mathbf{E} = -i \omega \mu_0 \mathbf{H} \quad \left[ A2.4c \right] \quad \nabla \times \mathbf{H} = -i \omega \mathbf{\varepsilon}_0 \mathbf{E} \quad \left[ A2.4d \right]
\]

Due to this harmonic time dependence, the first and second equations are automatically satisfied by the third and fourth by means of \( \nabla \cdot (\nabla \times \mathbf{F}) = 0, \forall \mathbf{F} \). This allows considering only the last two Maxwell’s equations.
The dielectric function will be consider periodic in the xy plane \( \varepsilon(r) = \varepsilon(r+a) \), but not in the z direction. Because of that, it is convenient to express the in-plane fields in terms of Bloch modes over reciprocal lattice vectors:

\[
H(r, z) = \sum_{G} \hat{H}_k(G, z)e^{ik \cdot r} \tag{[A2.5]}
\]

In order to satisfy Equation [A1.4b], \( \hat{H}_k(G, z) \) must be expressed in a particular basis states:

\[
H(r, z) = \sum_{G} \left\{ \phi_x(G) \begin{pmatrix} u_x - \frac{1}{q} (k_x + G_x) u_z \end{pmatrix} + \phi_y(G) \begin{pmatrix} u_y - \frac{1}{q} (k_y + G_y) u_z \end{pmatrix} \right\} e^{i(k \cdot G) \cdot r + iqz} \tag{[A2.6]}
\]

where \( u_x, u_y \) and \( u_z \) are the unit vectors along the x, y and z orthogonal directions, and \( \phi_x(G), \phi_y(G) \) are expansion coefficients.

Due to the arbitrary dependence of this expansion on the number of lattice vectors, and in order to present a more compact notation, a momentum representation may be introduced in Maxwell’s equations and fields [Note: Maxwell’s equations have been rescaled in the form \( \omega \varepsilon_0 E \rightarrow E \) and \( \sqrt{\mu_0 \varepsilon_0 \omega} = \omega/c \rightarrow \omega \].

\[
\begin{pmatrix} 0 & -\hat{\partial}_z & i\hat{k}_y \\ \hat{\partial}_z & 0 & -i\hat{k}_x \\ -i\hat{k}_y & i\hat{k}_x & 0 \end{pmatrix} \begin{pmatrix} h_x \\ h_y \\ h_z \end{pmatrix} = -i \begin{pmatrix} \hat{\varepsilon}_{xx} & \hat{\varepsilon}_{xy} & 0 \\ \hat{\varepsilon}_{yx} & \hat{\varepsilon}_{yy} & 0 \\ 0 & 0 & \hat{\varepsilon}_{zz} \end{pmatrix} \begin{pmatrix} e_x \\ e_y \\ e_z \end{pmatrix} \tag{[A2.6]}
\]

\[
\begin{pmatrix} 0 & -\hat{\partial}_z & i\hat{k}_y \\ \hat{\partial}_z & 0 & -i\hat{k}_x \\ -i\hat{k}_y & i\hat{k}_x & 0 \end{pmatrix} \begin{pmatrix} e_x \\ e_y \\ e_z \end{pmatrix} = -i\omega^2 \begin{pmatrix} h_x \\ h_y \\ h_z \end{pmatrix} \tag{[A2.7]}
\]

Here, \( \hat{k}_i \) are diagonal matrices with \( \hat{k}_i(G, G) = k_i + G_i \) and \( N_G \times N_G \) dimensions. In addition, it has been also introduced the Fourier expansion of the dielectric tensor \( \hat{\varepsilon} \), which depends on the periodicity of the layer structure. It is worth noticing that this dielectric tensor presents non-diagonal elements \( \varepsilon_{xy} \), allowing the patterned layer to have MO activity in the polar configuration [3].
denotes differentiation with respect to the z coordinate. Through this notation the fields adopt the following expression:

\[ h(z) = e^{iqz} \left\{ \phi_x \hat{u}_x + \phi_y \hat{u}_y - \frac{1}{q} \left( \hat{k}_x \phi_x + \hat{k}_y \phi_y \right) \hat{u}_z \right\} \]  

[A2.8]

where \( \phi_i = [\phi_i(G_1), \phi_i(G_2), \ldots]^T \) represents the vector of the expansion coefficients. Introducing this expression into Equations [A2.6] and multiplying by \( i \hat{n} \), being \( \hat{n} = \varepsilon^{-1} \) the Fourier expansion of the inverse dielectric tensor, the electric field reads:

\[
e(z) = \frac{1}{q} e^{iqz} \hat{n} \left\{ + \left( \hat{k}_y \hat{k}_x \phi_x + (q^2 + \hat{k}_y \hat{k}_y) \phi_y \right) \hat{u}_x 
- \left( \hat{k}_x \hat{k}_y \phi_y + (q^2 + \hat{k}_x \hat{k}_x) \phi_x \right) \hat{u}_y 
+ q \left( \hat{k}_y \phi_x - \hat{k}_x \phi_y \right) \hat{u}_z \right\}
\]  

[A2.9]

Inserting these fields into [A2.7], two final equations are obtained (the third is a linear combination of the others). In matrix form:

\[
\begin{bmatrix}
\hat{n}_{yy} & -\hat{n}_{yx} \\
-\hat{n}_{xy} & \hat{n}_{xx}
\end{bmatrix}
\begin{bmatrix}
q^2 & 0 \\
0 & q^2
\end{bmatrix}
+ \begin{bmatrix}
\hat{k}_y \hat{k}_x & \hat{k}_x \hat{k}_y \\
\hat{k}_x \hat{k}_y & \hat{k}_y \hat{k}_y
\end{bmatrix}
\begin{bmatrix}
\phi_x \\
\phi_y
\end{bmatrix}
= \omega^2 \begin{bmatrix}
\phi_x \\
\phi_y
\end{bmatrix}
\]  

[A2.10]

From this set of equations the \( q \) eigenvalues can be calculated. This will allow obtaining the in-plane fields in the patterned layer. In order to obtain such eigenvalue equation, this expression needs to be operated, so let’s put it in a more compact form:

\[
\mathcal{H}(q^2 + K) + \mathcal{K} \phi = \omega^2 \phi \quad \rightarrow \quad \mathcal{E}(\omega^2 - \mathcal{K}) + K \phi = q^2 \phi
\]  

[A2.11]

where \( \mathcal{H} \), \( K \), \( \kappa \), and \( \mathcal{E} \) accounts for the matrix containing components of the inverse dielectric tensor \( \hat{n} \), the \( \hat{k}_i \hat{k}_j \forall i,j \) product, the \( \hat{k}_i \eta_{zz} \hat{k}_j \forall i,j \) product, and the inverse of the \( \mathcal{H} \) matrix, respectively.
Once the \(q_n\) eigenvalues and the \(\phi_x^n, \phi_y^n\) expansion coefficients have been obtained, the in-plane electric and magnetic fields can be expressed as a sum of \(n\) forward and backward propagating waves:

\[
\begin{align*}
\begin{pmatrix} h_x(z) \\ h_y(z) \end{pmatrix} &= \sum_n \begin{pmatrix} \phi_x^n \\ \phi_y^n \end{pmatrix} \left( a_n e^{iq_n z} + b_n e^{iq_n (d-z)} \right) \\
\end{align*}
\]  

\[\text{[A2.12a]}\]

\[
\begin{align*}
\begin{pmatrix} -e_x(z) \\ e_x(z) \end{pmatrix} &= \sum_n (\omega^2 - \kappa) \cdot \begin{pmatrix} \phi_x^n \\ \phi_y^n \end{pmatrix} \frac{1}{q_n} \left( a_n e^{iq_n z} + b_n e^{iq_n (d-z)} \right) \\
\end{align*}
\]  

\[\text{[A2.12b]}\]

where \(a_n\) and \(b_n\) represent, respectively, the forward and backward wave coefficients for each \(q_n\) eigenvalue. Making \(e_\parallel = [-e_y, e_x], h_\parallel = [h_x, h_y]\), \(\hat{f}(z) \rightarrow \hat{f}(z)_{mn} = e^{iq_n z}\) and defining a matrix \(\Phi\) whose columns are the vectors \(\phi_x^n, \phi_y^n\), the fields expressions are given by:

\[
\begin{align*}
\begin{pmatrix} e_\parallel(z) \\ h_\parallel(z) \end{pmatrix} &= \begin{pmatrix} (\omega^2 - \kappa) \Phi \hat{q}^{-1} & -(\omega^2 - \kappa) \Phi \hat{q}^{-1} \end{pmatrix} \begin{pmatrix} \hat{f}(z) \alpha \\ \hat{f}(d-z) \beta \end{pmatrix} \\
&= M \begin{pmatrix} \hat{f}(z) \alpha \\ \hat{f}(d-z) \beta \end{pmatrix}
\end{align*}
\]  

\[\text{[A2.13]}\]

The final matrix \(M\) obtained contains all the information of the electromagnetic field distribution in a periodically nanostructured layer, and may be used to obtain the interface matrix \(I(n, n+1)\). This step is accomplished by making equal the fields at the interface of two adjacent layers, and as a consequence:

\[
I(n, n+1) = M_n^{-1} M_{n+1} = \begin{cases} \\
\frac{1}{2} \hat{q}_n \Phi_n^T \left( \omega^2 - \kappa_{n+1} \right) \Phi_{n+1} \hat{q}_{n+1}^{-1} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \\
+ \frac{1}{2} \Phi_n^T \left( \omega^2 - \kappa_n \right) \Phi_{n+1} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} 
\end{cases}
\]  

\[\text{[A2.14]}\]

The final process in this development consists in calculating the Fourier expansion of the dielectric tensor \(\hat{\varepsilon}\) of the nanostructured layer. As it was previously quoted, it will be assumed a periodical distribution in the xy plane, \(\hat{\varepsilon}(r) = \hat{\varepsilon}(r+a)\). Under such conditions, the momentum representation of the dielectric constant may be expressed, in Cartesian coordinates as:

\[
\hat{\varepsilon}(\mathbf{G}) = \frac{1}{S} \int_S \varepsilon(\mathbf{r}) e^{i \mathbf{G} \cdot \mathbf{r}} d\mathbf{r} = \frac{1}{S} \int_{x_1}^{x_2} \int_{y_1}^{y_2} \hat{\varepsilon}(x, y) e^{i G_x x + i G_y y} dx dy
\]  

\[\text{[A2.15]}\]
where S represents the unit cell area of the lattice. Depending on the periodicity of the nanostructure, the expression of the dielectric constant in the momentum representation is completely different and needs to be calculated in each case. In this thesis, most of the nanostructures are composed of cylindrical entities, presenting an hexagonal arrangement.

![Image](image.png)

**Figure A2.2** - Schematic example of the system under consideration. The hexagonal arrangement of the nanostructure translates into a particular momentum representation of the dielectric distribution.

To obtain the Fourier expansion of the dielectric function let us consider the fundamental unit cell of a hexagonal arrangement of circles presenting a different dielectric tensor with respect to the embedding matrix (see Figure A2.2). The dielectric distribution in the cell may be mathematically expressed in polar coordinates as:

\[
\tilde{\varepsilon}(\rho) = \tilde{\varepsilon}_0 + (\tilde{\varepsilon}_1 - \tilde{\varepsilon}_0) H(r_1 - \rho) + \ldots + (\tilde{\varepsilon}_n - \tilde{\varepsilon}_{n-1}) H(r_n - \rho)
\]  \hspace{1cm} [A2.16]

where \( H(x) \) represents the Heaviside step function whose value is zero or one for negative or positive arguments, respectively. The distribution has also considered an arbitrary number \( n \) of concentric cylinders with different dielectric tensors and diameters. In polar coordinates Equation [A2.15] reads:

\[
\tilde{\varepsilon}(\mathbf{G}) = \frac{1}{S} \int_0^{2\pi} \int_0^{\rho} \rho \tilde{\varepsilon}(\rho) e^{iG \rho \cos \theta} d\theta d\rho
\]  \hspace{1cm} [A2.17]

Introducing Equation AX1.16 into this expression, the momentum representation of the periodical dielectric tensor gives, depending on if the reciprocal vector \( \mathbf{G} \) is zero or not:

\[
\tilde{\varepsilon}(0) = \tilde{\varepsilon}_0 + \frac{\pi r^2 (\tilde{\varepsilon}_1 - \tilde{\varepsilon}_0)}{S} + \ldots + \frac{\pi r^2 (\tilde{\varepsilon}_n - \tilde{\varepsilon}_{n-1})}{S}
\]  \hspace{1cm} [A2.18]
\[ \tilde{\epsilon}(G) = \frac{2\pi (\tilde{\epsilon}_1 - \tilde{\epsilon}_0) r_1}{S} \frac{J_1(G r_1)}{G} + \ldots + \frac{2\pi (\tilde{\epsilon}_n - \tilde{\epsilon}_{n-1}) r_n}{S} \frac{J_1(G r_n)}{G} \]  

[A2.19]

where \( S \) accounts for the unit cell area, \( G = \sqrt{G_x^2 + G_y^2} \) represents the modulus of the reciprocal vector \( G \), and \( J_i \) is the Bessel function of the first kind. This expression may be finally inserted into the latter development performed to obtain the fields in a periodical nanostructured system.
Appendix 3

The Effective Medium Approximation (EMA)

In most cases analyzed along this thesis, the elements composing the nanostructures were randomly distributed. This represents a problem since, to theoretically analyze the experimental results, it is necessary to consider the most accurate description of the system. However, a disordered arrangement of nanoparticles usually makes such description very difficult due to the large computing time or memory resources required.

There is a way to overcome this problem if the dimensions of the nanoparticles are much smaller than the wavelength of the light. If this condition is fulfilled, the light behaves as if the nanostructure was a homogeneous medium, being its refractive index a function of that of the particles and of the surrounding medium. Therefore, it is possible to calculate an effective dielectric tensor including the optical (and MO) properties of the disordered particle ensemble and their embedding medium and use it in the theoretical calculations. This method is the so-called Effective Medium Approximation (EMA).

![Diagram](image)

**Figure A3.1** - The Maxwell-Garnett approximation calculates an effective dielectric tensor, function of the dielectric tensors of the surrounding medium and the different parts of the nanoparticles (core and shells)

There are many different EMAs (Maxwell-Garnett [1], Bruggeman [2], Looyenga[3], Sheng [4], Monecke [5], etc.), each of them being more or less accurate depending on the problem at hand [6]. For the purpose of this thesis, the Maxwell-Garnett approximation is a suitable option. The approximation is only valid for small nanoparticle concentrations, as it underestimates interactions and it does not take into account the size of the nanoparticle, just the aspect ratio [7]. As a consequence, it cannot consider effects that are related to size, as for instance the retardation effect that shifts to lower energies the surface plasmon resonance of relatively big particles [8].
The formalism to obtain the effective dielectric tensor follows that developed by M. Abe and T. Suwa [9], which is based on the earliest work of D. Stroud [10]. The particles have ellipsoidal shape and an arbitrary number of shells (core) n (see Figure A3.1). Both the nanoparticles and the embedding mediums are supposed to present MO activity, so that their dielectric tensors can be expressed as:

$$\varepsilon_i = \begin{pmatrix} \varepsilon_{i}^{xx} & \varepsilon_{i}^{xy} & 0 \\ \varepsilon_{i}^{yx} & \varepsilon_{i}^{yy} & 0 \\ 0 & 0 & \varepsilon_{i}^{zz} \end{pmatrix}$$  \[A3.1\]

For simplicity, all the mediums were supposed to be isotropic so that $\varepsilon^{xx} = \varepsilon^{yy} = \varepsilon^{zz}$. In addition, only the polar configuration (marked in red) has been considered for the applying magnetic field, although the approximation is easily extensible to the transversal and longitudinal configurations.

In order to obtain the effective dielectric tensor, it is necessary to calculate the electric fields and polarizabilities induced in the core and shells of a single nanoparticle [9]. The external applied field $F_0$ is no longer equal to $E_0$ in the matrix or $E_m$ in the mth-shell or core, because $F_0$ induces electric dipole moments inside the nanoparticle. Thus, for the mth-shell, the total electric field can be expressed as:

$$E_{mi}^i = \frac{F_{mi}^i}{a_m b_m c_m} + \frac{C_m N_m}{2}$$  \[A3.2\]

where the first term $F_{mi}^i$ is the electric field within the mth-shell, and the second represents the mth-component of the dipole field. This second term depends on the principal radius of the mth-ellipsoid $a_m$, $b_m$, $c_m$ (see Figure A3.1), on a constant $C_m$ to be adjusted to satisfy the boundary conditions, and on the depolarization factor $N_m^i$. The depolarization factor represents how much the internal field within the ellipsoid is weakened by the polarization, and is closely related to the shape of the ellipsoids. It is defined as:

$$N_m^x = \frac{1}{2} a_m b_m c_m \int_0^\infty (s + a_m^2)^{\frac{\gamma_x}{2}} (s + b_m^2)^{\frac{\gamma_x}{2}} (s + c_m^2)^{\frac{\gamma_x}{2}} ds$$

$$N_m^y = \frac{1}{2} a_m b_m c_m \int_0^\infty (s + a_m^2)^{\frac{\gamma_x}{2}} (s + b_m^2)^{\frac{\gamma_x}{2}} (s + c_m^2)^{\frac{\gamma_x}{2}} ds$$  \[A3.3\]

$$N_m^z = \frac{1}{2} a_m b_m c_m \int_0^\infty (s + a_m^2)^{\frac{\gamma_x}{2}} (s + b_m^2)^{\frac{\gamma_x}{2}} (s + c_m^2)^{\frac{\gamma_x}{2}} ds$$
Once the expression of the electric field is known, and the boundary conditions at each mth-interface set, it is possible to obtain a recurrent formula relating the electric field of the mth and the mth+1 shell:

\[
\begin{bmatrix}
F_{m-1} \\
C_{m-1}
\end{bmatrix} = \begin{bmatrix}
\tilde{T}_m \\
\tilde{C}_m
\end{bmatrix} \begin{bmatrix}
F_m \\
C_m
\end{bmatrix} \rightarrow F_m = \tilde{S}_m F_0
\]  

[A3.4]

\[
\tilde{S}_m = \left( \tilde{T}^{(1,1)}_{m+1} \cdot \tilde{T}^{(1,1)}_{m+2} \cdots \tilde{T}^{(1,1)}_n \right) \left( \tilde{T}^{(1,1)}_1 \cdot \tilde{T}^{(1,1)}_2 \cdots \tilde{T}^{(1,1)}_n \right)^{-1}
\]

from which it is possible to express the electric field in the mth-shell \( F_m \) in terms of the external field \( F_0 \). Here \( \tilde{T}_m \) and \( \tilde{S}_m \) represent a supermatrix and its (1,1) sub-block which depends on the dielectric tensors of the nanoparticles and host medium and on the depolarization factor.

This result can be used to calculate the total induced polarizations, electric fields, and electric flux densities for the whole composite:

\[
\langle P \rangle = \left( \sum_{m=1}^{n} t_m P_m \right) = \left( \sum_{m=1}^{n} t_m (\tilde{\varepsilon}_m - \tilde{\varepsilon}_0) F_m \right)
\]  

[A3.5a]

\[
\langle E \rangle = \left( \sum_{m=0}^{n} t_m E_m \right) = (1 - f) F_0 + f \sum_{m=1}^{n} t_m F_m
\]  

[A3.5b]

\[
\langle D \rangle = \langle \tilde{\varepsilon} \rangle \langle E \rangle = \tilde{\varepsilon}_0 \langle E \rangle + f \langle P \rangle
\]  

[A3.5c]

Substituting Equations [A3.5a]-[A3.5b] into Equation [A3.5c], and solving, the expression for the effective dielectric tensor we get:

\[
\langle \tilde{\varepsilon} \rangle = \tilde{\varepsilon}_0 + f \left[ \sum_{m=1}^{n} t_m (\tilde{\varepsilon}_m - \tilde{\varepsilon}_0) \tilde{S}_m \right] \left[ (1 - f) \tilde{I} + f \sum_{m=1}^{n} t_m \tilde{S}_m \right]^{-1}
\]  

[A3.6]

In general, this expression cannot be reduced to a simpler formula. Nevertheless, there are some cases in which it is possible. For instance, in the simplest case of a system composed of non-magnetic and non-shelled oblate nanoparticles, the effective medium tensor reduces to:

\[
\varepsilon_{\text{ef}}^i = \varepsilon_0 + \frac{f (\varepsilon_1 - \varepsilon_0) \varepsilon_0}{\varepsilon_0 + (1 - f)(\varepsilon_1 - \varepsilon_0) N_1^i}
\]  

[A3.7]

which is the simplified form of the expression (22) obtained by S. Ahn et al. [11]. Here, \( \tilde{\varepsilon}_1 \) represents the dielectric tensor of the oblate particle, \( \tilde{\varepsilon}_0 \) that of the
embedding medium, \( f \) the particle concentration, and \( N_m^i \) the depolarization factor.

If the nanoparticle presents MO activity and only the linear terms of the non-diagonal constant are taken into account, then the expression obtained for the effective MO dielectric constant reads:

\[
\varepsilon_{ef}^{xy} = \varepsilon_0^{xy} \frac{f \varepsilon_0^2}{\varepsilon_0 + (1 - f)(\varepsilon_1 - \varepsilon_0)N_1^x}^2
\]

Despite these cases, the effective dielectric tensor can only be obtained via numerical calculations. That is in part due to the depolarization factor, since the integrals in Equation [A3.3] cannot be analytically solved. However, throughout this thesis, most of the nanoparticles to be simulated present a prolate (wires) or oblate (disks) shape, i.e., they present two identical radiuses (in particular, \( a_m = b_m \)). Under such circumstances, the integrals admit a solution which depends on the beta \( B(x,y) \) and the Gauss hypergeometric \( {}_2F_1(\alpha,\beta;\gamma;z) \) functions, making the calculation considerably faster:

\[
N_m^{x,y} = \frac{1}{2}a_m^{-1}c_m B\left(\frac{3}{2},1\right) {}_2F_1\left(\frac{3}{2},1,\frac{5}{2};\frac{c_m^2-a_m^2}{c_m^2}\right)
\]

\[
N_m^{z} = \frac{1}{2}a_m^{-1}c_m B\left(\frac{3}{2},1\right) {}_2F_1\left(\frac{1}{2},1,\frac{3}{2};\frac{a_m^2-c_m^2}{a_m^2}\right)
\]
Appendix 4

Plasmonic Dispersion Relation for Multilayered Systems

In Chapter 5, it has been shown that the plasmonic dispersion relation for a single metal/dielectric interface is quite simple. For IMI or MIM symmetric systems the dispersion relation becomes a little bit more complicated. On the opposite, the dispersion relation is very hard to obtain for multilayered systems; even more since the MO activity of the layers have to be considered.

In this sense, some works have attempted to analyze the dispersion relation of multilayered systems [1], or systems presenting MO activity [2]. However, none of them have considered both characteristics at the same time. Moreover, the dielectric constants used in these studies did not take into account absorption, so that the dispersion relations obtained cannot be contrasted to a real system. For all these reasons, this appendix is devoted to the deduction of plasmonic dispersion relations in multilayered magneto-optically active systems with real dielectric constants.

Figure A4.1 - Scheme of a multilayered system. $a_n$ and $b_n$ are the forward and backward wave coefficients respectively. For the calculation of the plasmonic modes, no incoming waves are allowed. Moreover, a component of the electric field must be perpendicular to the system plane (z direction). As a consequence, only TM electromagnetic fields are allowed (characterized by $E_x$, $E_y$, and $H_y$). Bottom: Example of the EM field intensity distribution corresponding to the excitation of a plasmonic mode.

To do so, let’s consider a system composed of $n$ continuous layers arranged in the z-direction. Each layer is able to present MO activity in the transversal configuration, so that the dielectric tensor is no longer diagonal and $\varepsilon_{xz} \neq 0$. In this configuration, the Maxwell’s equations still admit a separation between the
TE and TM polarizations (the last characterized by $E_x$, $E_z$, and $H_y$). As it was pointed in Chapter 5, the excitation of propagating surface plasmon needs an electric field component perpendicular to the surface layer. This makes to be considered only TM modes, so this development will be restricted to them.

The solutions of the TM Maxwell’s equations for a given layer are electromagnetic fields which may be expressed as a linear combination of forward and backward waves:

\[
H_{y,i} = \left[ a_i e^{-\alpha_i(z-z_{i-1})} + b_i e^{+\alpha_i(z-z_{i})} \right] e^{i(k_z - \omega t)} \quad [A4.1a]
\]
\[
E_{x,i} = \left[ a_i F^+_i e^{-\alpha_i(z-z_{i-1})} + b_i F^-_i e^{+\alpha_i(z-z_{i})} \right] e^{i(k_z - \omega t)} \quad [A4.1b]
\]

, being $F_i^\pm$ the factor that allows expressing the electric field $E_x$ in terms of the magnetic field $H_y$ for the forward (+) and backward (-) waves:

\[
F_i^\pm = -i \frac{k_x^2 - k_0^2 \varepsilon_{xx,n}}{k_0 \mp \alpha_n \varepsilon_{xx,n} - ik_z \varepsilon_{xz,n}} \quad [A4.2]
\]

In addition, $\alpha_n = -ik_{zn}$ represents the $z$-component of the wave vector, which can be expressed in terms of the $x$-component as:

\[
k_z^2 = k_0^2 \left( \varepsilon_{xx} + \frac{\varepsilon_{xx}^2}{\varepsilon_{xx}} \right) - k_x^2 \quad [A4.3]
\]

For the existence of plasmonic modes, it must be imposed the condition of no incoming waves to the multilayered system (see Figure A4.1). As a consequence, the magnetic fields for the first and last semi-infinite layers must read as (the electric fields are modified in a similar manner as well):

\[
H_{y,1} = b_1 e^{+\alpha_1(z-z_{1-1})} e^{i(k_z - \omega t)} \quad [A4.4a]
\]
\[
H_{y,n} = a_n e^{-\alpha_n(z-z_{n-1})} e^{i(k_z - \omega t)} \quad [A4.4b]
\]

It is worth to note that there are no restrictions for the wave propagation at these two limiting layers. The absence of incoming waves and sources in the system assures the exponential decay of the electromagnetic fields outside it, and therefore, the only existence of surface plasmon modes as solutions.

The knowledge of the fields allows establishing their continuity at each interface $z = z_i$. The overall boundary conditions represent a system of $2n$ equations with $2n$ propagation coefficients which has non-zero solution just if the determinant is
zero. For a \( i = 1, \ldots, n \) layered system, this determinant presents the following form:

\[
\begin{vmatrix}
0 & 0 & \cdots & 0 \\
0 & 0 & \cdots & 0 \\
\vdots & \vdots & \ddots & \vdots \\
0 & 0 & \cdots & 0
\end{vmatrix} = 0
\]

where \( d_i = z_{i+1} - z_i \) represents the thickness of the \( i \)th layer. C. A. Ward et al. [3,4] have shown that it may be described through a general fully condensed expression, but restricted to non-magnetic multilayered systems. In spite of this, the determinant becomes more and more complicated as the number of layers considered increases, being very difficult to find an analytical expression for the dispersion relation. Only some specific cases, as the one described in Chapter 6, allow a particular approximation. In general, the solution of the determinant requires a numerical calculation.

An example of the plasmonic dispersion relation of a multilayered system may be appreciated in Figure A4.2. The system considered consists in an Au/Co/Au trilayer (15, 10, and 15nm thicknesses respectively) where the refractive indexes of the top and bottom semi-infinite mediums are air \((n=1.0)\) and BK7 glass \((n=1.5)\). Such a system must present two different plasmonic modes, as it was pointed out in Chapter 2. These are the low and high index modes (LIM and HIM).

![Figure A4.2](image)

**Figure A4.2** - Real (a) and imaginary (b) parts of the surface plasmon wave vector calculated for a 15nmAu-10nmCo-15nmAu with air and glass as background mediums. The blue and cyan lines account for the LIM and HIM modes, respectively. The dashed and dotted lines represent the corresponding lightlines for air and glass. The grey zone marks the region of the leaky modes (see text).

**Figure A4.2a** and **b** shows the real and imaginary parts of the plasmonic dispersion relation calculated for this system. The blue and cyan lines represent
the LIM and HIM modes respectively, whereas the dashed and dotted lines accounts for the air and glass lightlines. As it can be observed, for low energies the real part of the surface plasmon wave vectors remains larger than the lightlines till 2.5eV approximately (LIM respect to air and HIM respect to glass). Simultaneously, the imaginary parts increase, highlighting the increasing losse of the modes. From this point, the dispersion relations bend back and cross the lightlines, indicating that the modes have lost their plasmonic character. The grey zone shows the “leaky modes” region [5,6], where the plasmonic modes can decouple to light and reradiate. The origin of this behavior may be explained by attending to the LIM mode. Despite its wave vector is larger than that of the air lightline, it is at the same time smaller than the one of glass, and therefore there is a chance for this mode to decouple to light.

![Energy vs. Wavelength](image)

**Figure A4.3** - Real relative variation of the SPP wave vector with the magnetic field for the same system considered in **Figure A4.2**. The blue and cyan lines account for the low and high index plasmonic modes (LIM and HIM), respectively.

For the latter system, the LIM and HIM dispersion relations show a very similar profile when considering an applied magnetic. In order to see the modulation of the plasmonic wave vector, **Figure A4.3** shows the real relative variation of the wave vector for these modes. As it can be observed, this variation is very small (∼10⁻³%), although it appears larger for the HIM than for the LIM mode. The origin of this difference might be found analyzing the electromagnetic field distribution of these modes.

With this aim, the wave vector must be inserted into **Equations [A4.1]**, allowing to obtain the $a_i$ and $b_i$ propagation coefficients in each layer. The condition to exist of plasmonic modes forces the determinant to be equal to zero, and as a consequence, only $2n-1$ of the total $2n$ equations may be employed to calculate the propagation coefficients. Since there are $2n$ unknowns, the system is consequently undetermined allowing one of the unknowns to be parameterized. In the particular case of the latter system considered, the coefficient to parameterize is that of the first interface Air/Au ($b_1$), as it is where the LIM plasmonic mode is excited and where the electromagnetic field shows a maximum (For HIM modes, the coefficient to parameterize is that related to the
opposite Au/Air interface). For simplicity, its value is set to one. From this coefficient, the rest is calculated via a semi-recurrent formula:

\[
b_1 = 1
\]
\[
a_2 = b_1 \left( 1 + \frac{(C^a_2 F^+_2 - C^b_3)}{(C^b_3 - C^a_3 F^-_2)} e^{2\alpha_2 d_2} \right)^{-1}
\]
\[
\vdots
\]
\[
b_j = a_j \left( \frac{(C^a_{j+1} F^+_j - F^a_{j+1})}{(C^b_{j+1} - C^a_{j+1} F^-_j)} \right) e^{\alpha_j d_j}
\]
\[
\vdots
\]
\[
b_{n-1} = a_{n-1} \left( \frac{F^+_n - F^-_n}{F^-_n - F^-_{n-1}} \right) e^{\alpha_{n-1} d_{n-1}}
\]
\[
a_n = a_n e^{\alpha_{n-1} d_{n-1}} + b_{n-1}
\]

where \( C^a_k \) and \( C^b_k \) are defined as:

\[
C^a_k = \left( \frac{F^+_k - F^-_{k+1}}{F^-_{k+1} - F^-_k} \right) e^{2\alpha_k d_k} + 1
\]
\[
C^b_k = \left( \frac{F^+_k - F^-_{k+1}}{F^-_{k+1} - F^-_k} \right) F^-_k e^{2\alpha_k d_k} + F^+_k
\]

Depending on the energy of the mode, the distribution of the fields may vary. For instance, taking the LIM and HIM wave vectors at 1.5eV such distribution may be appreciated in Figure A4.4, where it has been depicted the intensity of the transverse magnetic field \( |H_y|^2 \). (a) Shows the fields for the layered system and its behavior in the course of ±1μm, whereas (b) represents a zoom of the fields within the Au/Co/Au trilayer. The fields have been normalized to the total area beneath the curves.
Figure A4.4 - (a) Normalized electromagnetic field distribution $|H_y|^2$ for the LIM and HIM plasmonic modes at 1.5eV energy. (b) Zoom of this field distribution to the Au/Co/Au trilayer. The dashed part of the line corresponds to the EM field inside the MO active material.

As it can be observed, both modes present a strong decay of the field intensity within the metallic trilayer and a smooth decay in the dielectrics. The decay is far more pronounced in the HIM mode, as it concentrates the field in the metallic structure. As a consequence, the field intensity in the MO active layer (Co) becomes larger in this mode, making in turn the variation of the wave vector with the magnetic field larger as well.
Appendix 5

Effect of Local Changes of the Refractive index on the MO response

In order to get a deeper insight about the localization of the EM field at the plasmonic resonance (see Chapter 3), we analyzed the effect of local dielectric changes on the MO response. To do so, we considered a cylindrical shell, surrounding the nanowire with a refractive index (n=1.4) smaller than that of the (n=1.7) background. In Figure A5.1a we show the $|r_{ps}|$ spectra calculated for a 15nm shell thickness, simultaneously with the $n=1.7$ and $n=1.4$ uniform backgrounds. Calculating the average refractive index obtained from the shell and background it is obtained $n=1.64$, so therefore the $|r_{ps}|$ spectral position and intensity was expected to be very similar to the $n=1.6$ case shown in Chapter 3. Indeed, the $|r_{ps}|$ peak position of the shelled nanowires shifts to higher energies, in accordance with the ratio of the dielectric refractive indexes. On the contrary, its intensity is much smaller than that of the background mediums.

![Figure A5.1 - Theoretical spectra of the (a) polarization conversion $|r_{ps}|$ and (b) average EM field intensity within the Ni nanowires for the latter shelled system. The black and purple lines correspond to different surroundings mediums ($n=1.7$ and $n=1.4$ refractive indexes, respectively), whereas the cyan line represents a system where the wires are surrounded by a 15nm thick shell ($n=1.4$) and embedded in a continuous $n=1.7$ medium.](image)

We compared these results with the corresponding calculation of the EM field within the nanowires. The results are displayed in Figure A5.1b, and show a very similar trend. The EM field calculated inside the shelled nanowire presents a large decrease with respect to that of the uniform backgrounds. Thus, in order to unravel the origin of this decrease, several spectra with different shell thicknesses were achieved, from zero (no shell) to half the lattice constant (neighbouring shells in contact). For each shell width, a full $|r_{ps}|$ activity...
spectrum was calculated. We show in Figures A5.2a and b the intensity and spectral position of the peak as a function of the shell thickness.

**Figure A5.2** - Cyan dots represent the (a) intensity and (b) spectral position of the $|r_{ps}|$ peak related to the plasmon resonance as a function of the $n=1.4$ shell thickness. The black and purple lines correspond to the values for the $n=1.7$ and $n=1.4$ uniform backgrounds respectively.

From these figures it can be observed that, as the shell thickness is increased, the curves depart from that associated to the $n=1.7$ continuous background and tend to that of a continuous background $n=1.4$. The spectral position of the peak presents an almost linear behaviour, whereas the evolution of its intensity does not appear to happen in a linear way. For example, for a 5nm shell around the wires a strong decrease of the intensity for the peak related to the plasmon resonance can be observed. A 20nm shell leads to the maximum decrease, and beyond these thickness the value of $|r_{ps}|$ approaches quickly to that of the $n=1.4$ uniform dielectric medium.

The origin of these differences might be due to the localization of the EM field at the metal-dielectric interface induced by the plasmon excitation. If the EM field were uniformly distributed throughout the system, the evolution of the $|r_{ps}|$ peak should be similar to that found in the continuous embedding medium case. However, as it is extremely localized, any variation of the index of refraction in the vicinity of the wires affects a larger amount of the electromagnetic field than in the uniform case and thus induces a bigger perturbation of the MO response.

This study was extended to perforated membranes. The environmental effects on the MO response to local variations of the dielectric medium in the holes was analyzed by considering inner shells within the pores with a different refractive index.
Figure A5.3 - Theoretical spectra of the (a) polarization conversion $|r_{ps}|$ and (b) average EM field intensity within the nickel for the latter shelled system. The black and purple lines correspond to different surrounding mediums ($n=1.7$ and $n=1.4$ refractive indexes, respectively), whereas the blue and cyan lines represent a system where the $n=1.7$ filled pores contain respectively a 10 and 20nm thick shell ($n=1.4$).

Figure A5.3a shows the $|r_{ps}|$ spectra calculated for a 10 and 20nm shell thickness, together with the $n=1.7$ and $n=1.4$ uniform backgrounds. The average refractive indexes obtained from the shell and background are $n=1.57$ and $n=1.48$ for the 10 and 20nm shells respectively. When comparing to the $n=1.6$ and $n=1.5$ previous background cases (see Chapter 3), the $|r_{ps}|$ spectral position and intensity for the shells is observed to be almost identical. Figure A5.3b shows the calculations of the average EM field intensity within the Ni membrane for the same shells and backgrounds as in Figure 3.20a. It can be appreciated the same evolution of the curves with the shell thickness. These results highly contrast with those obtained for the nanowires when considering plasmonic excitations, and are due to the small pore diameter considered in the calculations, which makes the EM field to present an almost uniform profile at the hole.
Bibliography

Chapter 1


Chapter 2


Chapter 3


Chapter 4


Chapter 5


Appendix 1


Appendix 2


Appendix 3


Appendix 4


List of Publications


Concluding Remarks

The most important conclusions that can be extracted from the studies performed all over this thesis are the following:

1.- Plasmon effects on the MO properties

- Nanostructures composed exclusively of ferromagnetic materials.

Two types of nanostructures were analyzed, nickel nanowires embedded in an alumina matrix, and perforated nickel membranes, both exhibiting MO activity and plasmonic properties simultaneously. We have shown that the excitation of plasmon-like modes leads to strong modifications in the MO response. In the plasmon excitation region, the nanostructured arrays presented similar Kerr rotation intensities despite the different Ni amount in the system. In the other spectral region, where no plasmon is excited, the intensity of the rotation turns out to be basically proportional to the amount of ferromagnetic material. This effect has found to be not only due to a reduction of the optical reflectivity of the system, but also to an increase of the polarization conversion due to the increase of the EM field within the MO active material. The spectral position of the enhancement could be tuned by changing the refractive index of the material.
surrounding (filling) the wires (pores) and/or changing the geometrical parameters.

- Nanostructures involving noble metals

The idea is to improve the performance by the incorporation of noble metals, since their plasmonic excitations are better defined due to the smaller absorption when compared with ferromagnetic metals. The influence of plasmon excitations on the MO response was analyzed in nanostructures composed exclusively of noble-metals (Au nanodisks) and in nanostructures made of a combination of these materials and ferromagnetic metals (Au/Co/Au nanodisks). Such nanostructures have shown MO activity enhanced by localized surface plasmons (LSPs). For pure noble metals nanostructures, we have shown that the plasmonic excitation allows having a measurable MO response of any metallic system at low magnetic fields. On the other hand, for the Au/Co/Au nanostructure, it has been shown that the enhancement of the electromagnetic field in the cobalt layer induced by the LSP makes the MO signal to present a large increase. Due to the presence of noble metals, we observed a narrowing of the spectral region where the MO signal is enhanced due to the LSP, when compared to the Ni nanostructures. Moreover, the intensity of the MO signal is similar to that of pure ferromagnetic metals, despite the smaller amount of MO active material. In addition, when comparing the MO activities between these two structures, significant differences in the shape of the observed MO spectra were found. These differences are due to the different physical mechanisms that give rise to magnetism in gold and cobalt, and they were explained through a simple theoretical model. These results were confirmed by numerical simulations.

2.- MO effects on Plasmon properties

We have studied how magnetic fields affect the properties of surface plasmons. In particular, we have studied the optical and MO responses of a continuous film made of combined noble metal and ferromagnetic materials (Au/Co/Au trilayers). In such structures we showed experimental evidence of SPP wave vector modulation due to the application of a magnetic field. This result proved that the external magnetic field not only acts on the MO activity of the structure, but also on the plasmon excitation itself.
Conclusiones Generales

Las conclusiones más importantes que pueden extraerse de esta tesis son las siguientes:

1.- Efectos de los Plasmones en las propiedades MagnetoÓpticas

- Nanoestructuras compuestas exclusivamente por materiales ferromagnéticos

Dos nanoestructuras diferentes fueron analizadas, hilos de níquel embebidos en una matriz de alúmina, y membranas perforadas de níquel, ambas exhibiendo simultáneamente actividad MO y propiedades plasmónicas. En este análisis hemos demostrado que la excitación de modos plasmónicos da lugar a grandes modificaciones en la respuesta MO. En la región de excitación del plasmon, la intensidad de la rotación Kerr en los sistemas nanoestructurados presentan intensidades similares a las del níquel masivo, a pesar de que la cantidad de material MO activo es sensiblemente menor. En la región de no excitación del plasmon en cambio, la intensidad de la rotación Kerr es básicamente proporcional a la cantidad de níquel en el sistema. Además, hemos demostrado que este efecto no se debe tan sólo a la disminución de la reflectividad óptica del sistema, sino también al aumento de la conversión de polarización, debido al
aumento del campo electromagnético en el interior del material MO activo. Asimismo, hemos visto que la posición espectral del aumento de la respuesta MO puede ser ajustada arbitrariamente cambiando el índice de refracción del material rodeando (o llenando) los nanohilos (poros) y/o cambiando los parámetros geométricos del sistema.

- Nanoestructuras que involucran metales nobles.

En este tipo de estructuras la idea era mejorar la respuesta MO del sistema incorporando los llamados metales nobles, cuyas propiedades plasmónicas están mucho mejor definidas debido a la menor absorción que presentan respecto a los materiales ferromagnéticos. Así pues, analizamos la influencia de las excitaciones de plasmones en la respuesta MO en nanoestructuras compuestas por metales nobles exclusivamente (nanodiscos de oro) y en nanoestructuras compuestas por una combinación de metales nobles y ferromagnéticos (nanodiscos de Au/Co/Au). Tales estructuras mostraron un aumento de la actividad MO debido a la excitación de plasmones localizados. Para las nanoestructuras compuestas exclusivamente de metales nobles, se vio que las excitaciones plasmónicas permiten obtener actividad MO medible a bajos campos magnéticos. Por otro lado, en las estructuras de Au/Co/Au se vio que el aumento del campo electromagnético en el cobalto inducido por la excitación plasmónica, inducía a su vez un aumento de la señal MO.

También se observó que la presencia de los metales nobles en estas estructuras produjo una disminución del ancho espectral donde se encuentra el aumento MO debido a los plasmones. Igualmente se observó que la intensidad de la señal MO era similar a la de las estructuras compuestas tan sólo por metales ferromagnéticos. Finalmente, comparando los discos de oro con los de oro-cobalto, se observó que la respuesta MO presentaba diferencias significativas en la forma de los espectros. Estas diferencias son debidas a los diferentes mecanismos físicos que dan lugar al magnetismo en el oro y el cobalto, y fueron explicadas mediante un modelo simplificado y confirmadas mediante simulaciones numéricas.

2.- Efectos Magnéticos en las propiedades Plasmónicas

En esta tesis también se studiaron cómo los campos magnéticos pueden afectar las propiedades de los plasmones superficiales. En particular, se estudiaron la respuesta óptica y MO de capas continuas compuestas por metales noble y ferromagnéticos (tricapas de Au/Co/Au). En tales estructuras se encontró evidencia experimental de que el vector de onda del plasmon propagante era modulado mediante la aplicación de un campo magnético. Este resultado demostró que un campo magnético no sólo actúa en la actividad MO de la estructura, sino también en la propia excitación plasmónica.