UNIVERSITAT AUTÒNOMA DE BARCELONA

DOCTORAL THESIS

Diffractive multifunctional plasmonic systems

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Declaration of Authorship

Gervasi Herranz Casabona, Tenured Scientist at the Institut de Ciencia de Materials de Barcelona (ICMAB-CSIC) and Jordi Sort Viñas, ICREA Research Professor at the Universitat Autònoma de Barcelona (UAB).

Certify,

that Rafael Cichelero Bachelor in Physics and Master in experimental Physics from the Universidade Federal do Rio Grande do Sul (UFRGS-BR), carried out under their supervision the thesis entitled "Diffractive multifunctional plasmonic systems". This work has been developed within a Ph.D. program in Materials Science at the Universitat Autònoma de Barcelona at the department of Physics.

For that record they sign the certificate.

Signed (Gervasi Herranz Casabona):

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Abstract

Doctor in Materials Science

Diffractive multifunctional plasmonic systems

by Rafael Cichelero

In this Thesis we have investigated the modulation of the optical properties of photonic crystals sustaining plasmonic excitations. Such properties can be tuned or enhanced by incorporating functional materials in the crystals, including magnetic, ferroelectric or piezoelectric compounds, which add versatility to the control of light at the nanoscale. To probe the interplay between the optical properties and external magnetic and electric fields, we used grating couplers as the basic device structure for this analysis, with emphasis on the combined effects of diffraction and plasmonic resonances in the enhancement of magneto-optic responses. We started our work with magnetoplasmonic crystals grown on commercial digital disks (DVD, CD, Blu-ray), showing the potential of these platforms for the development of magnetoplasmonic devices. We generalized the results found in these systems to magnetoplasmonic crystals with engineered optical responses, where the interplay between diffraction, plasmonics and magnetism paves the way to a versatile way to engineer the properties of photonic crystals that goes beyond the particular case of grating couplers. In particular, we have explored how, under some circumstances, light at off-normal incidence can excite unidirectional plasmonic propagating modes with particularly large magneto-optic responses. Finally, we close the Thesis with the study of ferroelectric magnetoplasmonic crystals, where the interplay between magnetization and ferroelectric polarization enables a way to modulate electrically the optical properties of grating couplers. In the following, a list is given of the most relevant outcomes described in this Thesis:

i - Observation in magnetoplasmonics crystals of an unexpectedly large transverse magneto-optical Kerr effect at quasi-normal incidence, where, by geometry, the intrinsic TMOKE amplitude tends to zero.

ii - Use of angle-resolved Fourier reflectance spectroscopy to analyze diffractive modes of light and plasmons, from which the interplay between magnetoplasmonic modes and diffraction has been revealed, associated with the amplification of the transverse magneto-optical Kerr effect at plasmonic resonances on the diffracted light.

iii - Geometrical conditions of grating couplers for the simultaneous maximization of reflectance and TMOKE, relevant for applications where signals are read out optically.

iv - Excitation of unidirectional forward- and backward- SPP propagating modes using grating couplers, with particularly large associated TMOKE responses.

v - An advanced lithography design was developed and implemented, which allowed the optical access and in-situ ferroelectric and magnetic measurements. We used this methodology to analyze the effects of the ferroelectric polarization on the TMOKE response: under certain conditions, the sign of TMOKE can be reversed by electric fields.

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List of Abbreviations

WSF	**W**hat (it) **S**tands **F**or
SPP	**S**urface **P**lasmon **P**olariton
TMOKE	**T**ransverse **M**agneto-**O**ptic **K**err **E**ffect
DMOKE	**D**iffracted **M**agneto-**O**ptic **K**err **E**ffect
TE or s	**T**ransverse **E**lectric
TM or p	**T**ransverse **M**agnetic
PLD	**P**ulsed **L**aser **D**eposition
EBE	**E**lectron **B**eam **E**vaporation
ALD	**A**tomic **L**aser **D**eposition
EBL	**E**lectron **B**eam **L**ithography
SEM	**S**canning **E**lectron **M**icroscopy
LRM	**L**eakage **R**adiation **M**icroscopy
N.A.	**N**umerical **A**perture
e.m.c	**e**lectro **m**agnetic **c**oil
BTO	**B**arium **T**itanate
STO	**S**trontium **T**itanate
LSMO	**L**anthanum **S**trontium **M**anganite
BHF	**B**uffered **H**ydro**F**luoric acid
FE	**F**erro**E**lectric
FM	**F**erro**M**agnetic
FDTD	**F**inite **D**ifference **T**ime **D**omain
ac	**a**lternate **c**urrent
dc	**d**irect **c**urrent
1D	**One D**imensional
ΔNF(M)	Δ**Variations** of **N**ear-**F**ield distribution as a function of **M**agnetization

List of Symbols

ε_d	Dielectric function of a dielectric layer
ε_m	Dielectric function of a metallic layer
ω	Angular frequency
λ	Light wavelength
λ_{SPP}	SPP wavelength
k_0	Wavevector of light at free-space
k_{SPP}	SPP wavevector
Ω	Larmor frequency
γ	Gyromagnetic ratio
θ_c	Critical angle
θ	Incidence angle
ϕ	Emission angle
ψ	Azimuthal angle
n	Refractive index
m	Diffraction order
η	Plasmonic order
Δ	Photonic crystal periodicity
Γ	filling fraction
τ	TMOKE amplitude
χ	Normalized average intensity
B	Magnetic field induction
H	Magnetic field
M	Magnetization
B_{ij}	Indicatrix
r_{ijk}	Electro-optic coefficients
p_{ijk}	Elasto-optic coefficients

Tales of a single Photon.

In the loneliness of the universe.
Lost in space, just passing by.
-I'm sorry, -"I do not interact".
You can tell, it does even not "wait, neither weight".

A little boy once asked, -Mr Photon why such a pace?
-I don't know, -I just want to "see".
-Life is too short, not at "c".

-arrrgh

-Sr. Photon are you well?

-I just collapse, into a "quantum well"!
-Am I trapped? -What are these things?!

-"Electrons and protons", not your "siblings".
. . .

Rafael Cichelero

Chapter 1

Introduction

1.1 Motivation

Moore's law [1] has been the greatest semiconductor industry's blessing: its predictions have been kept up by deep-scale device miniaturization as well as multi-core architectures [2, 3]. Yet, this progression may slow down soon. In a nutshell, the transmission of data across on-chip connections should improve in speed, energy efficiency and immunity against electromagnetic interferences if optical –instead of electrical– interconnects are used to shift data around. Consequently, photonic or hybrid electronic/photonic circuits are considered to be a promising perspective for the next-generation of integrated devices [4, 5].

This brings up the need for devices that control the flux of light at small scales [6]. For instance, optical isolators are considered key elements, as they exploit nonreciprocity to allow propagation of waves along one direction and block the reverse propagation [7, 8]. Indeed, optical isolators are crucial to build immunity against noise, increase the signal-to-noise ratio and ensure device stability (e.g., in laser sources integrated into circuitry) [9]. Unsurprisingly, nonreciprocal devices are a mainstay of any communication technology based on light (for instance, Faraday isolators and circulators are used for data transmission along conventional optical fibers) [10]. Facing this need, there is an emergent activity that aims at achieving unidirectional light propagation along integrated optical waveguides [11].

Additionally, next generations of optical interconnects will require optical modulation and routing capabilities with high performance and, therefore, optical modulators and switches are among the most relevant components of photonic integrated circuits [12]. In this sense, optoelectronic devices are investigated, in which modulation and switching capabilities can be accomplished by electric control of the amplitude and phase of electromagnetic waves, using architectures based either on Mach-Zehnder interferometers or microring resonators [13, 14]. Therefore, the control of the properties of devices by electric fields is an active research field in nanophotonics.

In optical communications, optical isolation can be achieved by Faraday rotators, which control the flux of light exploiting magneto-optical effects [15, 16, 17]. The latter arise because of the breaking of time-reversal symmetry in magnetic materials, giving way to non-reciprocal propagation of light [18, 19]. The same principle may be applied to integrated circuits. However, as the relevant device length is reduced to very small scales, the magneto-optic activity –which is proportional to the amount of material–, is drastically reduced [20, 21]. It is therefore necessary to compensate the large reduction in size and magneto-optic activity with a large enhancement of the intrinsic response.

Facing this challenge, a substantial research has been devoted to boost the intrinsic magneto-optical by coupling magnetic materials to photonic/ plasmonic devices. In this line, magnetophotonic crystals have been intensively investigated, in which nonreciprocal optical effects and large magneto-optical responses at stop-band edge frequencies have been reported [22, 23, 24, 25]. The interest in magnetic materials has also been extended to plasmonics, giving way to the field of magnetoplasmonics [26]. Beyond conventional approaches based on noble metals, a series of studies on the plasmonic properties of structures containing ferromagnetic materials has been carried out. These studies include materials such as Au/Co/Au multilayers [27], periodically patterned ferromagnetic films and noble metal/ ferromagnet structures [28, 29], magnetic nanowires [30], or nanodisk arrays [31, 32, 33, 34]. In such a large variety of systems, remarkable magneto-optical enhancements have been observed at wavelengths where surface plasmons are

excited. In the same vein, metallic diffraction gratings have been also studied in the context of magnetoplasmonic crystals fabricated out of magnetic metals [19] or noble metals in contact with magneto-optical garnet materials [35, 36, 37]. Using such gratings, control over SPP excitation conditions by an external transverse magnetic fields was demonstrated [19, 35, 38].

Yet, all these previous studies focused on the excitation of plasmons and its consequences on the optical properties, but did not exploit the phenomenon of diffraction. Indeed, diffraction in magneto-optically active systems is an important phenomenon that is used, e.g., in diffracted magneto-optical effect (DMOKE) to probe magnetization reversal dynamics of sub-micron-sized magnetic patterns [39, 40, 41, 42, 43]. However, up till now the mutual interplay of plasmonics, magneto-optics and diffraction has not been investigated in a single system. This Thesis is primarily concerned with the study of this interplay using the grating coupler structure as the basic device where to analyze the effect of diffraction and plasmonics on magneto-optic responses. We stress that, indeed, grating couplers are important devices in the perspective of integrated photonic applications. More specifically, grating couplers enable efficient coupling between light from optic fibers to on-chip waveguides [44, 45, 46] offering easy integration on wafers and enabling communication with external units via optical links.

In our study of magnetoplasmonic crystals based on gratings, one important aspect is the possibility of studying selectively surface plasmon polaritons propagating along backward or forward directions, enabling us to easily assess their non-reciprocal magnetic modulation. In addition to magnetism, we also show that plasmon propagation and magneto-optic responses can be modulated by electric fields after incorporation of ferroelectric materials into the structures. The combined action of magnetic and electric fields opens up fascinating avenues for multifunctional nano- photonic devices, which, as aforementioned, would enable the integration of active nanophotonic devices in integrated circuits. Additionally, we anticipate that our results can be generalized and used to design more complex diffractive elements, such as many plasmonic metasurfaces, where they could find use in designing non-reciprocal, isolating devices.

The following sections are devoted to give a brief introduction to the

main topics relevant to this Thesis. In the first place, we will give a concise description of surface plasmon polaritons. Secondly, we discuss briefly optical plasmon-mediated responses relevant in the context of this Thesis, including transverse magneto-optic Kerr effect (TMOKE) –where the propagation of plasmons is modulated by magnetic fields– as well as electro-optic and photoelastic effects –which arise from changes of the permittivity by electric fields–. Finally, the diffraction and interference phenomena and their relation to magnetoplasmonics will be overviewed.

1.2   Surface plasmon polaritons (SPPs)

Surface plasmons polaritons (SPPs) are electromagnetic excitations propagating at the interface between a conductor and a dielectric. These electromagnetic waves are originated by the collective oscillations of the electron plasma present in the conductor, generating an evanescent electromagnetic wave that propagates along the interface [47].

To capture the essential nature of SPPs, we focus on the simplest geometry, that of a single dielectric/metal interface, where we assume that waves propagate along the x-direction. The interface is defined between a dielectric, non-absorbing half space (along the positive z-direction $z > 0$), with positive real dielectric constant ε_d and an adjacent conducting half space ($z < 0$) described by a dielectric function $\varepsilon_m(\omega)$ with $\mathrm{Re}[\varepsilon_m] < 0$ -i.e., corresponding to the properties of metals at frequencies below the bulk plasmon frequency. Next, we use a modified Helmholtz equation –derived from Maxwell equations– which includes a complex propagation constant $\beta = k_x$, corresponding to the component of the wavevector along the direction of propagation. This is an important property of evanescent electromagnetic waves which, in contrast to free-space propagating waves, have a component of the wavevector along the direction of propagation [47, 48]. One can think about this longitudinal component as contributed by the longitudinal oscillations of the electronic plasma, which is the crucial ingredient that allows the possibility of evanescent propagation and also the possibility of breaking the Rayleigh diffraction limit [48].

The latter point can be understood by reminding that the Rayleigh diffraction limit can be derived from the uncertainty relations for photons, which gives:

$$\Delta_x \geq \frac{\lambda}{4\pi}. \tag{1.1}$$

Then, remembering that:

$$\Delta_x \geq \frac{1}{2\Delta k_x}, \tag{1.2}$$

and that

$$k = \sqrt{k_x^2 + k_y^2 + k_z^2}, \tag{1.3}$$

an increment in the wavevector component along x results in a higher confinement of light in this direction (see equation 1.2). However the possibility of confinement in the x-direction implies that, according to equation 1.3, a purely imaginary wavevector component has to be added along z, necessary to compensate the large wavevector along the x-direction. Introducing in a plane wave the purely imaginary wavevector $k_z = i\kappa_z$, where $\kappa_z \in \mathbb{R}$ results in $e^{ik_z z} = e^{-k_z z}$, giving an exponential decay along z, characteristic of evanescent waves.

We come back to the modified Helmholtz equation, which reads as follows for the electric field:

$$\frac{\partial \vec{E}(z)}{\partial t} + (k_0^2 - \beta^2)\vec{E} = 0, \tag{1.4}$$

with a similar equation for the magnetic field $\vec{H}$.

Now, we look at solutions of equation 1.4 for propagating waves confined to the interface, i.e. with evanescent decay in the perpendicular z-direction. It can be demonstrated that such solutions are forbidden for transverse electric (TE or s) modes, i.e., with only H_x, H_z and E_y being nonzero. Therefore, the excitation of SPPs can be only found for transverse magnetic (TM or p) modes, where only the field components E_x, E_z and H_y are

FIGURE 1.1: Schematics of the metal/dielectric interface.

nonzero (see the 1.1). This observation is relevant for experiments, since the excitation of SPPs can be tested by controlling the polarization state of the incident light. With hindsight, the fact that SPPs cannot be excited with s-polarized light is coherent with the observation that only p-polarized light can contribute to the wavevector along the propagation, which, as pointed out above, is a necessary condition for the excitation of evanescent waves.

For the abovementioned single dielectric/metal interface, it can be demonstrated that the TM (p-polarized) solutions to the propagation of SPP waves is determined by [47]:

The TM solutions for $z > 0$ can be written as:

$$H_y(z) = A_2 e^{i\beta x} e^{-k_2 z} \tag{1.5}$$

$$E_x(z) = iA_2 \frac{1}{\omega \varepsilon_0 \varepsilon_d} k_2 e^{i\beta x} e^{-k_2 z} \tag{1.6}$$

$$E_x(z) = -A_2 \frac{\beta}{\omega \varepsilon_0 \varepsilon_d} k_2 e^{i\beta x} e^{-k_2 z} \tag{1.7}$$

and for $z < 0$,

$$H_y(z) = A_1 e^{i\beta x} e^{k_1 z} \tag{1.8}$$

$$E_x(z) = -iA_1 \frac{1}{\omega \varepsilon_0 \varepsilon_m} k_1 e^{i\beta x} e^{k_1 z} \tag{1.9}$$

$$E_x(z) = -A_1 \frac{\beta}{\omega \varepsilon_0 \varepsilon_m} k_1 e^{i\beta x} e^{k_1 z} \tag{1.10}$$

Where ω, ε_0, ε_m and ε_d are the dielectric function of vacuum, metal and

dielectric, respectively. From these equations, one can derive the dispersion relation for SPPs, which is given by:

$$\beta = k_0 \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}}. \tag{1.11}$$

From the expression outlined in equation 1.11 one can infer that the wavevector of SPPs can be modulated by changes in the permittivity values of ε_m and ε_d. In the following, we analyze the two cases contemplated in this Thesis for the modulation of plasmons by external fields: firstly, the TMOKE generated from magnetic field-induced shifts of the plasmon resonances and, secondly, the modulation of the refractive index by electric fields, which include electro-optic and photo-elastic effects.

1.3 Modulation by magnetic fields: Transverse Magneto-Optic Kerr Effect mediated by plasmons

Magneto-optical effects arise from the interaction between light and matter, whenever the latter is subject to a magnetic field. In the case of magnetic ordered materials such as ferro- and ferri-magnetics, the effects are present even in the absence of any external magnetic field, being originated by the magnetization of the material. In the simplest scenario, the presence of a magnetic field (internal or external) changes the dispersion curves of the absorption coefficient, thus inducing a disparity in the optical anisotropy. Such disparity is a direct outcome of the splitting of the system energy levels under a magnetic field, which is called the Zeeman effect [49].

From a phenomenological point of view, the conventional magneto-optical effects are explained qualitatively by the changes in the medium refraction index n_+ and n_-, which correspond to the material response to the magnetic fields along opposite directions, H^+ and H^-, respectively. In the case of magnetic metals, one can understand intuitively the changes in refractive index as induced by changes in the Larmor frequency $\Omega = -\gamma B$,

which consists in the frequency of the electron spin precessing around a external applied magnetic field B, where γ is the gyromagnetic ratio $\gamma = -\frac{eg}{2m_e}$, where e, g and m_e are the electron- charge, g-factor and mass, hence, under external magnetic field the Larmor frequency can be written as:

$$\Omega_\pm = \Omega_0 \pm \frac{eH}{m_e c},$$ (1.12)

where, c and H are the light speed and the magnetic induction. The appearance of two resonance frequencies (Ω_+ and Ω_-) induces a slitting in the absorption, resulting in the displacement in the dispersion response $n_+(\Omega)$ and $n_-(\Omega)$, resulting in:

$$n_\pm(\Omega) \approx n(\Omega) \pm \frac{eH}{2mc}\frac{dn}{d\Omega},$$ (1.13)

where $n(\Omega)$ is the refractive index in the absence of magnetic field H and $\frac{dn}{d\Omega}$ is the variation of refractive index based on the Zeeman slitting.

This Thesis is focused on the observation of transverse magneto-optical Kerr effect (TMOKE), in which changes in reflectance are measured as a function of the magnetic field. In TMOKE, the external magnetic field is applied parallel to the sample surface and the perpendicular to the light plane of incidence (see figure 1.2).

FIGURE 1.2: The changes in total intensity exerted by the external magnetic field are demonstrated in a hysteresis cycle.

The observation of TMOKE is based on physical mechanisms similar to the ones described previously. More specifically, the observation of TMOKE

in magnetic multilayers can be described by the modification of the permittivity by a transverse magnetic field H. In the case of magnetoplasmonic gratings, this, in turn, modifies the wavevector k_{SPP} of the SPPs, which results in a magneto-optic effect that arises from the magnetic modulation of the SPP excitation condition. The change in k_{SPP} depends on the off-diagonal permittivity ε_{xy} of the metallic layer and is given by [50, 51, 52].

FIGURE 1.3: Induced displacement in the SPP resonance frequency by a magnetic field.

$$k_{SPP}(M) = \pm k_{SPP}\left(1 \mp \frac{M\varepsilon_{xy}}{\beta}\right), \qquad (1.14)$$

where $\beta = \sqrt{\varepsilon_m \varepsilon_d}(1 - \varepsilon_m^2/\varepsilon_d^2)$, 1.14, the plasmon wave vector k_{SPP} and the magnetization M are preceded by $\pm$ and $\mp$ signs to account for the two possible directions of the SPP propagation and the magnetization. Their mutual relationship is determined by the symmetry of the system. The magnetization of the grating locally breaks the time-reversal symmetry of the system. Consequently, a reversal of the SPP propagation direction is equivalent to a reversal of the magnetization direction. Thus the degeneracy between forward and backward propagating modes is lifted and their wavevectors are distinct (see figure 1.3), which results in changes in the diffracted light intensity as the function of magnetic field. To quantify these changes, the magnetization of the sample is cycled to record a hysteresis loop from which variation of the reflected and diffracted intensities as function of magnetization can be extracted. In chapters 3, 4 and 5 we describe in detail the mechanisms that lead to a large enhancement of TMOKE responses in gratings, driven by the excitation of SPPs.

1.4   Modulation by electric fields: electro-optic and photoelastic effects

1.4.1   Linear electro-optic effect

In electro-optic materials the refractive index can be modulated by electric fields. This property can be exploited to develop nano-optical filters, modulators and switches that control the amplitude and phase of electromagnetic waves in integrated communication systems. Fully polarized ferroelectric materials exhibit the linear electro-optic effect -also called the Pockels electro-optic effect-, where the light intensity can be controlled by altering the birefringence of with an external electric field. The Pockels effect is only present in non-centrosymmetric point groups, which include ferroelectric materials.

The linear electro-optic response is defined in terms of the changes induced by the electrical field in the optical indicatrix [53]:

$$B_{ij}(E) - B_{ij}(0) = \Delta B_{ij} = r_{ijk}E_k, \tag{1.15}$$

where the variation in the indicatrix ΔB_{ij} determines the changes in the refractive relative to the crystal orientation, while r_{ijk} are the electro-optic coefficients. The indicatrix, can be reduced to a third rank polar tensor known as Pockels effect, being written as:

$$\Delta B_i = r_{ij}E_j \quad (i = 1, 2, ..., 6. \quad j = 1, 2, 3). \tag{1.16}$$

The complexity associated to the indicatrix imposes a previous analysis of the crystallography and optical orientation of the crystal, since the optical effects are entangled to the electro-optic coefficients. The changes in the refractive index induced by the external electric field can be simplified in some cases as:

$$\Delta n(E) = \frac{-n^3}{2}(\delta B) = -\left(\frac{-n^3}{2}rE\right), \tag{1.17}$$

where, r and E are the electro-optic coefficient and the external electric field, being in the general case in the order of ($r = 10^{-11}$m/V and $E =$

10^6V/m) respectively, resulting in a refractive index modulation in the order of $\Delta n(E) \approx 10^{-5}$.

1.4.2 Photoelastic effects

Alternatively, the refractive index can be changed by electric fields through the converse piezoelectric effect, so that the state of mechanical stress can be modulated. This effect is also important in acousto-optic devices, optical switches and modulators [53, 54]. The photoelastic effect can be described mathematically by the tensorial expression [53]:

$$\Delta B_{ij} = p_{ijkl}x_{kl}, \tag{1.18}$$

where ΔB_{ij} corresponds to the changes in the indicatrix, which, as mentioned above, determines the magnitude of the refractive index relative to the orientation in a crystal-, p_{ijkl} is the elasto-optic tensor and x_{kl} is the acting strain vector.

In ferroelectrics, the application of electric fields may result in different microscopic arrangements of ferroelectric domains that may give way to different strain states, resulting in changes of the refractive index through photoelastic effects. As discussed in chapter 6, the modulation of the TMOKE responses by electric fields and, under appropriate conditions, the reversal of the sign of TMOKE induced electrically, can be rationalized by photoelastic effects in magnetoplasmonic crystals that include ferroelectric materials.

1.5 Diffraction and interference of electromagnetic waves

Interference results from the superposition of two or more waves. Historically, the phenomenon of interference was instrumental in establishing the wave nature of light [55] and later, the double-slit experiment –either with photons or material particles– has become the emblematic cornerstone experiment of quantum mechanics [56].

During the course of this Thesis, we used the diffraction phenomena as a tool to probe and amplify SPP effects in the magneto-optic response of magnetoplasmonic crystals, composed of magnetic metallic multilayers. A one-dimensional photonic crystal, i.e. a grating coupler, can be modeled as a se-

FIGURE 1.4: Double slit experiment.

ries of equispaced slits (grating periodicity (Δ)). In the grating, the impinging wavefront is diffracted into different beams that, depending on the slit separation, are emitted at specific angles ϕ, determined by the difference in optical paths between successive beams, which is given by $\delta = 2\pi\Delta\sin(\phi)/\lambda$ (see figure 1.4). The far-field response of the nth slit can be calculated as:

$$E_j(\theta) = Ae^{i(j-1)\delta},\tag{1.19}$$

with $j = 1, 2, ..., q$, where q is the total number of slits, and, thus the total electric field can be computed as:

$$E_j(\phi) = \sum_{j=1}^{q} E_j(\phi) = A\sum_{j=1}^{q}(e^{i\delta})^{j-1},\tag{1.20}$$

which can be simplified to:

$$E(\phi) = A\frac{1 - e^{iq\delta}}{1 - e^{i\delta}},\tag{1.21}$$

resulting in the total intensity:

$$I = I^0\left[\frac{sin^2(q\frac{\delta}{2})}{sin^2(\frac{\delta}{2})}\right],\tag{1.22}$$

where I^0 is the individual slit intensity.

Chapter 2

Experimental methods and sample fabrication

This chapter includes a brief description of the deposition, lithography and optical characterization techniques that have been used in this Thesis. The chapter is divided in two subsections, where the sample fabrication is described first, followed by a description of the methods used for optical characterization.

2.1 Sample fabrication

The sample fabrication is described in different subsections, in which deposition and lithography techniques are presented. We start with the description of the deposition techniques.

2.1.1 Deposition techniques

The materials studied in this Thesis were either metals (mostly Au and Co) or oxide perovskites (e.g., $BaTiO_3$). For the growth of the former we used electron-beam evaporation, whereas for oxides pulsed laser deposition (PLD) was used instead.

Electron-beam evaporation

The electron-beam evaporation (EBE) method consists in a high vacuum deposition technique where electrons are accelerated through a static electric

potential V in the order of 10 kV, providing them with high kinetic energy, so these electrons are directed against the material to be deposited. Whenever an electron collides against the target its energy is transferred to the material, mostly as thermal energy. Once the temperature and vacuum levels are high, the material evaporates coating the sample. The process can be controlled by the amount of current I provided to the electron gun, that is used to tune or cease the evaporation rate.

A schematic representation of the EBE is shown in figure 2.1, where a current I flows through an electron gun, generating electrons which are accelerated through and electric potential V, reaching the crucible that contains the desired material. In this process, the material that is evaporated from the crucible is deposited over the sample, while the total amount of deposited material is obtained through a quartz balance.

FIGURE 2.1: Schematics of e-beam evaporation. Electrons, which are generated by a filament, are accelerated against a target material through an electric potential V. Whenever a critical current I is achieved (melting point), the material in the crucible evaporates to the sample holder.

This technique was used specially for the growth of gold (Au), cobalt (Co) and chromium (Cr). A more detailed description of the sample preparation and protocols are given in the sections below.

Pulsed laser deposition

Pulsed laser deposition (PLD) was the technique used in the growth of ferroelectric thin films (BaTiO$_3$). It consists in a high power pulsed laser that is

focused on the material target located in a chamber with controllable oxygen pressure. The material then sublimates from the target, coating the substrate. The process occurs in the presence of an oxygen atmosphere in which the pressure is adjusted in order to have materials with optimal stoichiometry. In addition to pressure, other relevant parameters are the laser fluence -usually measured in Joules/cm^2- the repetition rate and the temperature at which the substrate is kept during the growth [57].

In this Thesis, we report on experiments carried out in magnetoplasmonic systems that incorporate ferroelectric BaTiO$_3$ layers. For the deposition of this material, we took advantage of Dr. F. Sánchez (ICMAB) expertise in high-quality BaTiO$_3$ thin films with optimal ferroelectric properties [58, 59].

FIGURE 2.2: Schematics of pulse laser deposition. A pulsed laser light is focused on the target, sublimating the material on the substrate.

2.1.2 Lithography techniques

We used microfabrication methods based on lithography [60] to develop advanced photonic micro- and nano-structures, including the construction of nanophotonic devices with ferroelectrics, which required optical access

with in-situ applied electric fields. We used two different lithographic techniques, namely, optical lithography, used to pattern structures on the micron scale and electron-beam lithography for sub-micron scale structures.

Optical lithography

A standard optical lithography process is shown in figure 2.3. The process of micro-fabrication starts when a light-sensitive polymer is coated over the desired substrate (a), and then it is annealed until the polymer solidifies. The exposition is done through ultraviolet light ($\lambda = 385$ nm) (b), exposing the desired marked areas. During the exposition, the UV light breaks the molecular links between the polymer chains, allowing the removal of the polymeric resist through a developer (see figure 2.3 (c)). Subsequently, the sample goes through a deposition step (d), where the substrate and photoresist are coated (e). Conventionally, in the final step, a lift-off is performed, where the photoresist is etched away by a solvent, usually acetone. The final structure consists of patterned metallic layers deposited over the substrate.

FIGURE 2.3: Standard photo-lithography scheme: In (a) a photo- or electron- sensitive polymer is spin coated on the substrate, followed by an exposition to light or an electron-beam (b). Subsequently, the mask is etched away by a developer in (c), after which the sample is coated with a metal (d). Finally, a liftoff is performed revealing the patterned structure (e).

Electron-beam lithography

Aiming for high precision and control, the electron-beam lithography offers nanoscale resolution. Electrons are collimated and accelerated against an

electron-sensitive resist, allowing the production of nanostructures in the order of tens of nanometers. A conventional method to define patterns using e-beam lithography follows a similar protocol to the one described by figure 2.3, where the photon-sensitive polymer and exposition are replaced by an electron-sensitive resists and exposition to electron-beams. More technical details about the lithography process illustrated in figure 2.3 can be found in Appendix A.

2.1.3 Patterning of the magnetoplasmonic crystals

In the last subsection, we defined how patterns can be made through a lithography process. In particular cases, such patterns can be used to protect specific regions of the sample from chemical or physical attacks done by dry or wet etching techniques. For metallic layers dry etching proves to be extremely efficient, having very high etching rates. For some cases, as is the case of Alumina (Al_2O_3 as discussed below) and $BaTiO_3$, dry etching fails, because the etching rates drop dramatically, and therefore a chemical etching is necessary.

Ion Milling

For the dry etching, ion milling was used. The technique is based on the acceleration of argon ions that are accelerated against the target. After hitting the latter, the energy from the impinging ions is used to eject atoms from the target surface. For conventional metals, high etching rates in the order of 60 nm per minute or higher, are easily achieved. However, the etching rates for oxides drops to just a few nanometers per minute, which makes ion milling impractical as it could cause both damage to the material and etching of the resist. In this case it is preferred to use a different etching technique, as discussed in the next section.

Wet etching

In this method, the sample is immersed in a bath of a certain etchant. A wet etching process is designed to corrode a specific material, in which a specific etchant is selected, thus reacting exclusively with the desired material. This

technique is extensively used in the industrial scale, where for instance sili-
con oxide wafers are etched by buffered hydrofluoric acid (BHF). Generally,
for the case of complex oxides, semiconductors and some metals the wet
etching process is not well studied, and only few reviews and compilations
can be found [61, 62]. In this work BHF was used to corrode Al_2O_3, having
an etching rate of 60 nm per minute at a concentration of 4%, leaving the
photoresist intact.

Although dry or wet etching are a convenient way to create patterns,
they are not exempt of some drawbacks arising mostly from anisotropic
etching along the walls of the patterns, which is schematized in figure 2.4.
For example, some of the milled material can be accumulated at the borders
of the wall, as shown in figure 2.4 (a). On the other hand, during the wet
etching the acid penetrates isotropically, including the regions under the re-
sist, as shown in figure 2.4 (b), digging small cavities at side of the structure.
Thus, the induced anisotropic by wet or dry etching potentially can limit
the scalability of the device. For instance, wet etching cannot be used for
an extended period of time, since the corrosion can dissolve the base of the
nano-structure. A similar problem happens also for ion milling, where nar-
row structures can present a large shape anisotropy due to reabsorption of
material near the borders. The ion-milling induced anisotropy can be min-
imized through rotation of sample during the process, thus reducing the
anisotropic etching. Unfortunately, this method could not be applied in the
wet etching, so these effects could not be reduced in that case.

FIGURE 2.4: Schematics for dry- (a) and wet- (b) etching,
showing the effects of anisotropic etching.

2.1.4 Device fabrication

In the following we discuss the details regarding the fabrication of different magnetoplasmonic crystals -grown on digital disks or on conventional substrates- and in combination with ferroelectrics.

Magnetoplasmonic crystals on top of patterned commercial disk media

This subsection is focused on the preparation of magnetoplasmonic systems, where commercial optical disks were used as substrates. Even though having a fixed periodicity, commercial optical disks such as as CD, DVD and Blu-ray can be easily used as photonic crystals in massive scale at very low cost.

FIGURE 2.5: Schematics of an optical disk. (a) Initially, the patterned polycarbonate surface is protected by an additional polycarbonate layer. After mechanical dismantling, the protective layer is removed and the exposed patterned surface is coated by a magnetic metallic multilayer, as shown schematically in (b).

The fabrication of magnetoplasmonic crystals on top of commercial disks requires that the structure of the optical disk (see figure 2.5 (a)) is first dismantled mechanically. Subsequently the exposed patterns can be easily identified since light is diffracted by the gratings. The surface has to be cleaned carefully, to ensure the full removal of all the optical disk coatings. In the case of DVD disks, the grating structure can be easily cleaned using a scotch tape followed by a bath in ethanol, in which the polycarbonate is immersed for a few minutes. Afterwards, the sample is ready for the metallic coating and finally the photonic crystal is ready to be measured

Magnetoplasmonic crystals on unpatterned substrates

The aforementioned protocol allows the use of conventional optical disks as a basis for photonic nano-devices. However, the geometry of the nano-devices is constrained to the topography of the surface patterns of the optical disk. Aiming for full flexibility in defining the magnetoplasmonic crystals, we require using of electron-beam lithography to define the magnetoplasmonic systems. The protocol described here was used for the construction of the photonic crystals presented at chapters 4 and 5.

We used a process that included a combination of optical- and e-beam-lithography. This is illustrated in the schematics of figure 2.6, where a magnetoplasmonic base (red) hosts photonic structures (salmon), in which the base has the finality to provide optical access to the photonic structures.

After the evaporation of the magnetic multilayers, an electron-beam lithography is performed (shown by the salmon areas), in which a mask is set over the metallic layers. The final step of the process consists in the milling of the metal not covered by the electron sensitive polymer down to the required thickness. At the end of the process, magnetoplasmonic crystals are prepared with tailored geometry, in which periodicity, width and depth of the structures are adjusted at will.

FIGURE 2.6: Design of the combination of optical- and electron-beam lithography. Red areas indicate where the photonic structures are located, depicted in salmon. Crosses are used for alignment.

Magnetoplasmonic crystals with ferroelectrics

This subsection is dedicated to an advanced lithography method, which allows the construction of devices that simultaneously are able to switch a ferroelectric (FE) material while allowing optical measurements on multifunctional photonic crystals, enabling the modulation of optical properties by external electric fields. Ferroelectrics are a good option, as they have

electro-optic and photoelastic properties, by which the refractive index can be varied linearly with electric fields.

Yet, ferroelectric materials that are scaled to into smaller dimensions present serious challenges that must be addressed. For instance, ferroelectric thin films are not fully insulating, as they may present leakage currents that may hinder the switching of electric polarization, especially when films are scaled to small sizes, typically on the micron scale. This, in turn, puts a constraint on the ferroelectric contact size, which should be large enough to enable electrical contact with wires. Besides that, the switching areas have to be accessed optically, which means that electrodes have to be contacted away from region that is measured with light.

The process starts with the deposition of the ferroelectric layer. In our case, we deposited $BaTiO_3$ (BTO) grown on top of $SrTiO_3$ (STO). $BaTiO_3$ (BTO), is selected because their bulk crystals present large electro-optic Pockels coefficients [63, 64, 65] and photoelastic properties [54, 66]. As a bottom electrode, grown between BTO and the substrate, we used $La_{0.7}Sr_{0.3}MnO_3$ (LSMO), which is a metallic oxide perovskite, with crystal structure compatible with BTO, making easier its integration with the ferroelectric [58, 59].

The patterning process starts once the substrates are fully covered by BTO/LSMO bilayers of thickness typically 200 nm/10 nm. The lithographic process is summarized schematically in 2.7, which is based on several nanofabrication steps, including lithography, wet-etching, ion-milling and metallic evaporations, described in the next paragraphs.

The construction of ferroelectric magnetoplasmonic crystals follows a building-block hierarchy, starting with the inner structure that consists of a metallic Au/Co magnetoplasmonic multilayer in the areas shown in red in figures 2.7 and 2.8. This step is done through optical lithography, with the active contacts and the alignment crosses shown in red and green respectively in figures 2.7 and 2.8. The areas shaded in red are then covered with magnetoplasmonic multilayers patterned into gratings of different periodicities, as shown later in figure 2.10.

FIGURE 2.7: Schematics of ferroelectric magnetoplasmonic crystals as defined by lithography. Ferroelectric active areas and alignment patterns are displayed in red and green colors, respectively. Blue areas correspond to top electrodes (Au) that contact the ferroelectric layer.

As aforementioned, ferroelectric thin films present the problem of leakage currents, which may preclude the ability to switch the electric polarization. To minimize the effect of leakage currents, the area where the ferroelectric is contacted has to be reduced to about less than about $< 10000 \ \mu m^2$. In addition, the top electrodes -usually defined by evaporating Au/Al- have to be designed so that the zone that is contacted electrically is far away ($\approx$ 4 mm) from the region that is accessed optically (see the blue areas in figure 2.7). Taking into account these constraints, the reduction of leakage currents was ensured by using a dielectric passivation layer grown before the deposition of top-electrode contacts (shown in blue). Note that the top electrode only contacts a small fraction of the active contact (shown in red), since these are the regions that are accessed optically see the red and gray areas in 2.7

and 2.8. As a passivation layer, we used Alumina (Al$_2$O$_3$) grown by atomic layer deposition (ALD). We found that, in order to be effective, the Al$_2$O$_3$ layer should have a minimum thickness of 300 nm, otherwise the leakage currents are not reduced enough. Ferroelectric polarization-electric field loops measured in devices defined by this lithography are shown later in chapter 6. Finally, as mentioned above, the top electrode is in direct contact with the ferroelectric exclusively inside small regions (gray areas in figures 2.7 and 2.8 i.e. the interconnection bridges).

FIGURE 2.8: Schematics from the motive micro-structure, in which the long-range contacts, ferroelectric active areas, alignment patterns and cladding bridges are shown in blue, red, green and gray respectively.

After the passivation layer has been deposited, the whole sample is covered by the Al$_2$O$_3$ layer including the active contacts (shown in red). Therefore, a wet etching was necessary to unblock the connection with the active layers shown in figure 2.9 (a). For that purpose, an additional photolithography mask has been placed, marking the regions where the passivation layer has to be removed, inside the red areas shown in figures 2.7 and 2.8. The process has been done through chemical attack, using an acid solution of BHF (Buffered Hydrofluoric Acid) with concentration of 4%, hence etching the Al$_2$O$_3$ layer at a rate of 60 nm per minute. A side view of the device structure can be seen in figure 2.9 (a), in which the passivation layer is

observed covering the plasmonic active layers. The corrosion stops once the acid dissolves the uncovered Al_2O_3 layer, as seen in 2.9 (b). The resulting layer structure can be seen in 2.9 (c), which shows how the passivation layer covers the whole sample with the exception of the metallic contacts that are in direct contact with BTO.

FIGURE 2.9: Cross-sectional views of: (a) the plasmonic contacts after sample passivation; (b) after the passivation layer is attacked by BHF; (c) the final structure with unblocked connections to the magnetoplasmonic layers.

Once the active contacts are unobstructed, a new optical lithography has been performed creating the long-range contacts and bridges for the ferroelectric switching. The long-range switching structure can be identified in figures 2.7 and 2.8 by the layers in blue and gray colors. Finally a metallic evaporation was performed, in which relative thick (200 nm) metallic layer is grown, thus establishing the connection between bridge and contacts. A thick metallic layer is essential to overcome the gap between the ferroelectric and the long-range contacts, since the difference in height between these two sets of contacts depends on the thickness of the passivation layer, as shown in the cross-sectional view of figure 2.10.

With the macro- and micro- motive structure finished, the last lithography process consists in the patterning of photonic crystals into the magnetoplasmonic active layers. This process was done through e-beam lithography, followed by an ion-milling, where the metal not protected by the electron-sensitive polymer has been removed by the argon atoms, thus shaping the photonic crystal as intended. The cross-sectional schematics shown in figure 2.10 displays the final structure of the magnetoplasmonic crystals in the active areas that are accessed optically.

The full process list can be listed as:

- Plasmonic active contacts were deposited on the ferroelectric thin film through optical lithography.

- Deposition of the passivation layer through atomic layer deposition.

- Wet etching, opening the access to the plasmonic contacts.

- Macro contacts assembly through optical lithography.

- Photonic crystal construction through electron-beam lithography and dry etching.

The aforementioned structure provides a stable and reliable way to integrate magnetoplasmonic devices with ferroelectric materials. On the other hand the amount of lithography steps entails a long development time. In order to accelerate the process, a simplified process, which is described in the next section, is based on the replacement of the Al_2O_3 passivation layer by a photoresist. Doing so, the number of nano-fabrication steps is reduced, but it comes at a price of mechanical and chemical instability.

A simplified construction

As discussed above, the simplified process consists in the substitution of the Al_2O_3 passivation layer by a layer of photoresist, which is annealed at high temperatures, thus solidifying the polymer. This baking process, although granting an extra resistance against solvents, does not allow prolonged exposures to them, as it could be corroded.

Similarly to the methodology described previously, the process starts with the definition of the main contacts and alignment patterns using optical lithography. Subsequently, the sample is annealed at 150 degrees for 30 minutes. An extra layer of photoresist is then deposited at the top of the hardened photoresist, and the long-range connections, bridges, alignment pattern and electro-active plasmonic contacts were exposed. Finally a metallic evaporation was performed, covering all the exposed areas with a magnetoplasmonic multilayer. During the liftoff (short process) only the

second layer of photoresist is susceptible to the solvent etching, thus, leaving the first photoresist layer intact, therefore acting as a passivation layer. Afterwards, the nano-patterning is performed by e-beam lithography and ion-milling as discussed in the previous case.

Thus, the process can be summarized as:

- A photoresist exposed and then hardened, acting as a passivation layer.

- A second layer of photoresist exposed followed by a metallic evaporation, however during the liftoff only the second layer was removed.

- Photonic crystal construction through electron-beam lithography and dry etching.

A qualitative comparison from the previously presented methods can be seen in figure 2.10. The comparison between the two passivation layers is shown in table 2.1.4.

FIGURE 2.10: Cross-sectional plots showing the full layered structure, using as passivation layers either Al_2O_3 (a) or photoresist (b).

Lithography method	Al_2O_3	Photoresist
Lithography steps	5-6	3
Evaporations	2-3	1
ALD depositons	1	0
Ferroelectric switching	good	good
Stability	good	poor
Corrosion resistance	good	poor

As indicated in table 2.1.4, both protocols prove to be useful to reduce leakage currents, enabling an effective switching of the ferroelectric polarization.

2.2 Optical characterization

This section describes the experimental methodology adopted for the optical characterization of the magnetoplasmonic crystals. It embraces different experimental techniques. We first describe leakage radiation microscopy, which aims at obtaining the leakage radiation generated by evanescent waves, e.g., from surface plasmons polaritons (SPPs). The section also includes Fourier optics spectroscopy, which allows the acquisition of the band structures of photonic systems. We have relied on these approaches to develop methods to explore how magnetic effects influence SPPs absorption or propagation. Alternatively, we have also developed methods to analyze individually the signatures of light or plasmonic diffraction modes of specific orders.

2.2.1 Experimental setup

The main structure and components of the experimental setup can be seen in figure 2.11, which, in the following, we accompany with a brief description of its components. A xenon lamp is used to provide a continuous white light source that is sent through a monochromator. The monochromatic beam is then collimated through an objective lens 10x N.A. 0.4 (Obj, 1), and after going through a linear polarizer and a 50:50 beam splitter, the beam is directed towards the sample surface, the monochromatic light is focused on the sample focal plane by an oil immersion objective lens 63x N.A. 1.4 (Obj, 2), and the reflected light is collected by the same lens and directed to a set of condenser lenses (Cond, 1 and Cond, 2) after which it is projected on two different detectors. One of them is a CCD camera aiming to ensure that the sample is placed in the focal plane, providing

a monochromatic real space image. The second detector consists of a sC-MOS camera placed in the Fourier focal plane, which allows the detection of the photonic band structure of a given photonic material or crystal. The experimental setup also holds two step motors (Step M, 1 and Step M, 2), which allow a precise alignment, focus, shaping and steering of the light beam. The setup also allows the placement of electromagnetic coils (e.m.c), which enable the measurement of magneto-optic hysteresis loops.

FIGURE 2.11: Description of the experimental setup. A xenon lamp provides a continuum spectra of white light, which is directed to a monochromator. The beam, after collimated (Obj, 1) and polarized, goes through a beam splitter (B-S) to an oil immersion objective lens (Obj, 2). The reflected beam is collected through a set of condenser lenses (Cond, 1) and (Cond, 2), finally arriving at the detectors (CCD) and (sCMOS) cameras where, real and reciprocal space images can be extracted.

The setup presented in figure 2.11 is the building block for all the measurements taken and presented during this work. Below we give a brief description of the experiments that were performed using the setup shown in figure 2.11.

2.2.2 Real space imaging

One way of probing and excite SPPs is through a technique called leakage radiation microscopy (LRM), [67, 68]. This technique is based on the excitation of plasmons in Kretschmann configuration [69], which are excited at the metal/dielectric interface. The SPPs excited through Kretschmann configuration are coupled into plasmons when the impinging light satisfies a coupling condition, i.e. whenever the angle of incidence is at critical angle θ_C as presented in figure 2.12 (a). Yet,

SPPs can also be excited through corrugated/rough surfaces [70] (seen in the next chapters).

FIGURE 2.12: In (a) the standard Kretschmann configuration is shown, in which a dielectric prism allows the coupling of SPPs, whenever the incoming light has an angle of incidence equal to the critical angle (θ_C). In (b) an oil immersion objective lenses allows the excitation of SPPs at the metal/oil interface, whenever the critical angle condition is satisfied.

The leakage radiation microscopy can be thought as exploiting the Kretschmann configuration, where the dielectric prism is replaced by an oil immersion objective lens as shown in figure 2.12 (b), where the immersion oil allows SPPs to be excited under matching with the refractive index.

A conventional illumination scheme is shown in figure 2.13 (a), where the back aperture of an oil immersion objective lens is fully illuminated. Since surface plasmons (SPPs) are only coupled at the critical angle, the beam size can be reduced from the full width of the lens aperture (6 mm) to near 0.5 mm, thus narrowing down substantially the range of angles of incidence. In these illumination conditions, the location of the beam spot in the back aperture can be steered as shown in 2.13

FIGURE 2.13: Depiction of the beam illumination and position at the back aperture of an oil immersion objective lens. In (a) a full aperture is exposed by the beam. In (b) the beam size is reduced and displaced to angles of incidence near the critical angle θ_C. Panel (c) shows the SPPs excited through Kretschmann configuration, which is characterized by an spectral leakage radiation emission.

(b). For instance, if the displaced beam shown in (b) satisfies the critical angle condition, SSPs are excited and their leakage radiation is collected, as shown schematically by figure 2.13 (c).

The formed image in (c) consists in the reflected beam together with the leakage radiation. The radiation pattern is, in general, wavelength dependent, i.e. longer wavelengths are able to couple SPPs that can propagate longer than short wavelengths. Generally, losses in the noble metals are smaller for longer wavelengths, which explains the larger propagation at these frequencies.

2.2.3 Reciprocal space imaging in the Fourier plane

Beyond microscopy techniques, Fourier optics [55, 71] provides access to the band structure of photonic crystals. The working principles of the Fourier optics are based on the collimation of the light coming from a point source schematically represented in figure 2.14, so that a plane wave is projected into the detector (camera sCMOS in figure 2.11). As shown by figure 2.14 the detector is placed in a specific position called the Fourier plane, in which the reflected light projects the reciprocal space response. Hence the Fourier plane provides the band structure of photonic structures, where the response is projected into the pixel arrays of the sCMOS camera. In this configuration, the angular information can be recovered through a space-angular conversion, which is discussed in section 2.2.3.

FIGURE 2.14: Schematics of the principles of Fourier optics spectroscopy. The ensemble of angles generated by a point source is collected and collimated, converting the original angle of emission ϕ into a position in the Fourier plane.

Scanning Fourier method

A Fourier optics arrangement can use a spectrometer as the main detector, which senses the light intensity collected from the sample through an optical fiber. This detection scheme is depicted in figure 2.15 (a), which shows schematically a motorized stage that moves an optical optical fiber with aperture ds, which scans across the whole space with steps dx and dy, as shown at figure 2.15 (b). Whenever a full scan along x or y is completed, the band structure of a given photonic structure can be reconstructed. The light polarization axis is in general set along the scanning directions, as shown in 2.15, where, for instance, the p- polarized mode is set along the x scanning axis.

FIGURE 2.15: Sketch of Fourier optics scanning method. An optical fiber with aperture ds is moved by a stepper motor capable of moving in $\hat{x}$ and $\hat{y}$ directions, corresponding to p- and s- polarization. The light is collected by the fiber and delivered through an optical fiber to a spectrometer.

Photonic band structures: Angle-resolved spectroscopy.

The projection of the Fourier plane (as seen in figure 2.14) can be analyzed and calibrated, allowing the conversion from space to angles of incidence, which provides a proper description of the photonic band structure. The calibration from positions in the Fourier plane and angles from the point source can be done using commercial 1D photonic crystals (in our case a DVD optical disk) with well known periodicity. The corresponding Bragg diffraction

provides accurately the angular position, enabling, after a proper rescaling in the Fourier plane, the reconstruction of diffracted and plasmonic modes.

FIGURE 2.16: (a) The light impinging on a one-dimensional photonic crystal is decomposed into two diffracted beams shown in blue. The well-known periodicity of the crystal allows a proper calibration, so that plasmonic dispersion lines (shown in (b) can be correctly identified from scans in the Fourier plane. The lines in (b) are calculated from the equation 2.2.

This process can be understood as follows. The light impinging on a one-dimensional photonic crystal is decomposed into diffracted modes, which are emitted from the sample surface as shown in figure 2.16 (a). However, light can be also converted into SPPs, if wavevector matching conditions are found between light and plasmons. The conversion of light into SPPs is visible by a dip in reflectance caused by the absorption of light at specific resonance wavelengths, as shown in figure 2.16 (b), where a well defined crossing between two SPP bands can be identified.

The dispersion lines for diffracted and plasmonic modes can be defined according to the Bragg condition, as explained below. Using these equations, the calculated plasmon dispersion lines were included in figure 2.16 (b). The Bragg law relates the incident wavevector k_{in} to the one of the reflected wave k_{dif} by the expression:

$$k_{in}(\lambda) = k_{dif}(\lambda) \pm m\Delta, \tag{2.1}$$

from which the wavevectors can be written as $k_{in} = \frac{n\omega}{c}\sin\theta_{in}$, $k_{dif} = \frac{n\omega}{c}\sin\theta_{ref}$ and the grating component $\Delta = \frac{2\pi}{d}$, where c are ω the light speed

and frequency, θ_{in} and θ_{ref} are the angle of incidence and reflection, d is the grating periodicity, m is a integer corresponding to the different diffraction modes and n is the refractive index. In what follows, we will call the modes indexed by m as simply "diffracted modes". Next we assume that light initially coupled into SPP, is then diffracted by the 1D photonic crystal. Thus the equation 2.1 can be written as:

$$k_{SPP} = k_x \pm \eta \frac{2\pi}{\Delta}, \tag{2.2}$$

where k_{SPP}, k_x and η, are, respectively, the SPP wavevector, the interface component of the wavevector of the incident light $k_x = k_{in} \sin\theta$, and the plasmonic orders. Oftentimes we will refer in this Thesis to the diffracted SPPs as simply "plasmon dispersion lines of order η", or similar.

Monochromatic Fourier Microscopy

An alternative way to perform the Fourier analysis consists in replacing the spectrometer by a fast camera (sCMOS camera), where the full Fourier plane can be detected at once in few milliseconds, without the help of scanning motors. In this method, the resolution is given by the camera pixel size, where in our apparatus it corresponds to 12 μm. As a result

FIGURE 2.17: (a) A real space image is shown, from a commercial grating (Thorlabs GH13-24V). (b) We show the reciprocal space image obtained from the Fourier plane for a given wavelength ($\lambda = 530$ nm). The initial polarization state of the incoming light is defined by the guided arrows s' and p'. (c) The dotted lines represent iso-angular circumference with similar values of Θ. The red-shaded area corresponds to the region to be integrated for the data acquisition for the p-light component. (d) Shows the reconstruction of the reflectance plot from a 1D photonic crystal normalized by the reflectance of an Al (aluminum) mirror.

of the combination of pixel size and camera speed, imaging of direct or reciprocal space can be speed up by about two orders of magnitudes (e.g., an image of $600x600$ pixels can be taken in about 2 minutes instead of about 150 minutes using the spectrometer).

In turn, this increase of speed comes with a massive increase of volume of data, typically by about two orders of magnitude. For instance, the acquisition of an image using the spectrometer requires the storage of about 1 megabytes of information, while using the sCMOS camera the volume increases to about 200 megabytes. This amount of data demands a big storage capacity and very efficient data analysis.

The methodology behind the Fourier microscopy can be explained with the help of figure 2.17. Panel (a) shows the image in real space of a commercial GH13-24V photonic crystal from Thorlabs. To obtain this image, the focal plane was adjusted to the plane corresponding photonic crystal surface, so that all light coming from that plane was collected and detected. Panel (b) shows the image of the same photonic crystal in the reciprocal space for a particular wavelength. In this image, the arrows s′ and p′ indicate the initial orientation of the s- and p-polarization of the incident light. The frame obtained in (b) contains two sets of angular coordinates of the Fourier plane, one corresponding to the angle of incidence θ and the other the azimuthal angle ψ, shown schematically in (c) as the dotted concentric circumferences and the in plane angle, respectively.

The method enables the imaging of the photonic band structure obtained with a particular state of the polarization of the incoming light. For instance, if we choose to select only the p-light component, we should collect the pixels corresponding to the red-shaded area shown schematically in figure 2.17 (c). In the conditions of the experiments, the angle ψ should not be larger than about 3 degrees. Finally, by collecting the data for the whole range of wavelengths, the full Fourier spectrum for the p-light polarization is obtained. Thus, figure 2.17 (d) shows the p-component Fourier spectrum of a calibration grating normalized by an Al mirror. A similar procedure can be done for incoming light with s-polarization, just by changing the azimuthal angle ψ, when ψ satisfies two conditions: $\psi < 93$ and $\psi > 87$ degrees.

Summarizing the measurement and analysis routine, the following steps must be done:

- Focusing the light in the photonic crystal plane (real space).

- Collecting monochromatic pictures of the Fourier plane (reciprocal space).

- The continuum of light spectrum is scanned.

- Performing the pixel-angular conversion.

2.2.4   Diffractive Fourier optics

By reducing the beam spot size in the objective back aperture, (see section 2.2.2), we could untangle the diffracted light independently of the specular reflection, which allowed us to explore and discover how plasmonic resonances affect the diffracted light. Beyond that, the diffraction Fourier optics is capable to reveal very weak surface plasmon effects, which would be unnoticed in the conventional approach.

FIGURE 2.18: Diffractive Fourier optics schematics: (a) The incidence light impinges the center of the back aperture of an objective lens. In the case of a continuous flat layer, the light specularly reflected. (b) The continuous layer is replaced by a diffraction granting, from which the decomposition of the diffracted modes can be identified individually in the Fourier plane.

We note that by decreasing the beam spot size, the angles of incidence of the light impinging the sample is narrowed down to a limited range of values. Therefore, for the case of a continuous metallic layer illuminated by a reduced ensemble of angles, just the pure reflection can be detected, as shown schematically in figure 2.18 (a). Yet, in the case of a periodically corrugated structure, the light intensity will be decomposed into diffracted modes, that will be emitted at specific angles, which depend on the periodicity of the corrugation. Hence, all the light upcoming from the diffracted modes is collected, and projected into the respective emission angles at the Fourier plane, as displayed schematically in figure 2.18 (b).

The diffracted Fourier optics proved to be also a suitable method for the detection of weak modifications in the light intensity. This approach has been used to characterize magnetoplasmonic effects in the reflected and diffracted light, presented later in chapter 4.

2.2.5   Magnetic and ferroelectric loops

Magneto-optic hysteresis loops were measured in multifunctional (magnetic or ferroelectric) photonic crystals. A conventional hysteresis loop is shown in figure 2.19 (a). We will see in chapter 6 that for specific studies it is convenient to measure the initial electric polarization curves. This can be achieved through ac depolarization processes. This approach consists in setting an ac electric field with an amplitude that is decreased gradually to zero. This method provides a versatile way of demagnetization or electric depolarization, although real-time measurements during the depolarization process are almost impossible due to the frequency of the ac fields (from the order of 0.001 Hz) [72].

As an alternative, a quasi-static ac demagnetization/depolarization has been also used, which allows in-situ measurements during the demagnetization/depolarization. The method is presented schematically in figure 2.19 (b), for the case of magnetic hysteresis loops. The red dots indicate specific values of the applied external field, whereas the red dashed lines indicate the path followed by the changes in the field polarity.

FIGURE 2.19: In (a) a conventional magnetic hysteresis loop is shown in blue, in with the magnetic field cycled around the loop. Panel (b) depicts a measurement using a quasi-static ac demagnetization, where the magnetic field polarity is reversed between consecutive data points, while the magnitude is decreased gradually. The red dots and dashed lines are a guide to the eyes, and display the magnetic field path over the demagnetization cycle. Finally at (c), the ac demagnetization states are shown by the black dots, representing the magnetic state at a given magnetic field. A peculiarity can be observed in the red shadowed dots, where irreversible magnetic states are found within the hysteresis loop, being part of minor loops indicated by the purple lines.

Initially (point 1°) a negative saturating field is applied, and the corresponding opto-electric response is recorded. Subsequently, the field is cycled around the loop, by reversing its polarity between consecutive measurements. The measurement is performed typically over many steps, until

the whole loop is measured.

The aforementioned demagnetization/depolarization protocol allows to measure signals along minor hysteresis loops, as shown schematically in figure 2.19 (c) by black dots.

Due to the magnetic shape anisotropy induced by the long axis of the 1D photonic crystals, a demagnetized state could not be properly induced. Yet, in the case of the ferroelectric magnetoplasmonic crystals, we will see in chapter 6 that the way the material is electrically depolarized plays a key role in the optical properties.

2.2.6 Ellipsometry

Ellipsometry spectroscopy enables to extract the permittivity of materials. In these experiments, a beam of linearly polarized light is projected on the sample with an angle of incidence θ. The reflected beam passes through an analyzer and is collected by a detector, as shown in the schematics 2.21. The experiments measure the complex reflectance ratio that corresponds to:

$$\varrho = \frac{r_p}{r_s} = \tan(\Psi)e^{i\delta}, \tag{2.3}$$

where, r_s and r_p are the light amplitude after reflection for $s-$ and $p-$ polarized light, $\tan(\Psi)$ is the amplitude ratio upon reflection and δ is the phase shift.

FIGURE 2.20: Schematics of an ellipsometry spectroscopy setup, in which a polarized light beam is projected on the sample surface, and its reflection reaches the detector after going though an analyzer.

To extract the optical constants, a model is required. As an illustrative example, we show in figure 2.21 the data corresponding to three different

samples, namely, pure Au and Co thin films, each with thickness 200 nm, and a magnetoplasmonic multilayer. The ellipsometry has been performed over three different composition, a pure Au and Co thin films with thicknesses of 200 nm, and a magnetoplasmonic multilayer composed by Au (16 nm) / [Co (14 nm) / Au (16 nm)] x4 / Co (14 nm) / Au (7 nm). All the aforementioned thin films were treated as a continuous layer of infinite thickness. Besides that, optical constants for countless compounds can be obtained from Palik [73]. The resulting model, provides the optical constant for Au, Co and the Au-Co multilayer shown in figure 2.21 (a), (b) and (c), where the real and imaginary refractive index components i.e. n and k_i, are displayed in red and blue respectively.

FIGURE 2.21: Spectral response from the complex refractive index, panels (a), (b) and (c) present the dielectric functions of a pure Au, Co and the plasmonic multilayer Au/Co.

2.2.7   Scanning electron microscopy (SEM)

The scanning electron microscopy (SEM) was used for the development and imaging of the photonic crystals, being used in two different ways: electron-beam lithography (EBL) and a conventional imaging technique SEM. The principle of operation can be observed in figure 2.22, which shows schematically how an electron gun injects those electrons in the system, accelerated by an external electric potential, being projected into the sample direction. A set of condenser lenses is used to collimate the beam, reducing aberrations and astigmatism. The electron-beam diameter is controlled by a tunable aperture. The aperture size plays an important role in the EBL, in which only extremely collimated electrons are desired to arrive at the sample surface, since small deviations in the electron-beam trajectory increase the exposition area.

After the impinging electrons interact with the sample, the response is collected by two distinct detectors: a secondary electrons detector, and back-scattered electrons. Secondary electrons are generated by the collisions between impinging electrons and the material under study. These electrons are in general used for high resolution imaging. On the other hand, backscattered electrons, are those electrons reflected from the sample by elastic scattering. As expected from elastic scattering, the total momentum is preserved, what makes the back-scattered electrons technique sensitive to the chemical composition of materials and, thus, can be used for the recognition of different elements in the sample.

electron-gun

condenser
lenses

aperture

sec. electron
detc.

sample

backscattered
electron dect.

FIGURE 2.22: Schematics of a scanning electron microscopy, in which electrons generated by an electron gun are accelerated against the sample. A set of condenser lenses and an iris with tunable aperture are used collimate and reduce the beam aberrations. Afterwards the beam interacts with the sample, which has its response detected by two detectors: a secondary electrons detector and a back-scattering detector.

Chapter 3

Magnetoplasmonic crystals based
on commercial digital disks

3.1 Abstract

Here we investigate the transverse magneto-optical Kerr effect (TMOKE) on large scale magnetoplasmonics crystals grown on top of conventional commercial optical disks. A full angle-resolved Fourier analysis reveals a complex band structure arising from the excitation of surface plasmons polaritons (SPPs) within a periodic lattice. The corresponding analysis over a broad range of wavelengths and angles of incidence reveals that the magneto-optic TMOKE response is enhanced substantially in the vicinity of the SPP resonances, providing a clear evidence of the interaction between the impinging light with the propagating surface plasmons, that results in a surprisingly large magneto-optical response.

Our results are in agreement with previous literature, in which plasmons have been shown to increase the magneto-optic responses in numerous previous works. However, the originality of the work presented here is that, under suitable conditions, an unexpected large TMOKE response is found at quasi-normal incidence, where, by symmetry, the intrinsic signal should be vanishingly small. The key towards this unexpected outcome is to engineer the geometry and permittivity of magnetoplasmonic crystals, so that first-order plasmon dispersion lines run up towards quasi-normal angles of

incidence. Incidentally, our results show the large potential of a standard commercial disk as platforms for enhanced magneto-optics devices.

3.1.1 Chapter highlights

- Use of a full angle-resolved Fourier reflectance spectroscopy reveals a band structure arising from the interplay between conventional light and surface plasmon polaritons, which are induced by coupling to magnetoplasmonic gratings.

- Observation of unexpectedly large transverse magneto-optical Kerr effect at quasi-normal incidence, where, by geometry the TMOKE amplitude tends to zero.

- The possibility of mass production of magnetoplasmonics crystals on top of standard commercial disks.

3.2 Introduction

Surface plasmons polaritons (SPPs) are collective oscillations of electric charges at metal/dielectric interfaces that are coupled to electromagnetic waves, thus generating confined electromagnetic fields into sub-wavelengths volumes that increase dramatically the density of electromagnetic energy. This effect drives a large magnification of light-matter interactions, enabling, e.g., the detection of single molecules through surface-enhanced Raman spectroscopy SERS [74, 75]. Localized surface plasmon resonances (LSPR), have been also used to enhance the imaging resolution, in which sub-micron objects can be seen [76]. Due to the extreme sensitive to changes in the permittivity, SPPs are nowadays vastly used in applications for probing and sensing [77, 78].

Unsurprisingly, many nanophotonic applications aim at the exploitation of the properties of surface plasmons. Here we put our focus in the particular case of magneto-optical applications, illustrated by the case of Faraday isolators. Such devices play nowadays a fundamental role in optical communications, where they isolate optical signals coming from back reflections that could have a detrimental effect on the signal quality. This property is used in Faraday isolators based on large-scale devices based on YIG [15, 16, 17, 79, 80]. However, for implementation of the same principles in integrated photonic circuitry, Faraday isolators should be downsized to much smaller scales, using thin film technology instead of bulk crystals. The dramatic device downscaling requires the magneto-optic signal -which is proportional to the optical path in the material- to be largely increased. Here is where surface plasmon resonances (SPR) in magnetoplasmonic systems may pave the way to increased magneto-optic activity required for applications in integrated nanophotonic devices.

Following these lines, it has been shown that metal/dielectric systems incorporating ferromagnetic thin layers support plasmonic resonances, leading to large increases of the magneto-optic Kerr [20, 81, 82], Faraday [16, 83, 84, 85] and transverse effects [86, 87, 88]. Therefore, the combination of plasmonic nanostructures with magneto-optic materials paves the way to the development of ultracompact nonreciprocal photonic devices.

These developments have spurred an interest to design and fabricate devices based on plasmonic nanostructures. This, in turn, has urged the need of inexpensive, reproducible and largely-scalable methods for the development of magnetoplasmonics nanodevices. An interesting platform consists of off-the-shelf commercial digital disks, where the surface is covered by a periodic patterned structure. The patterns on the surface of these disks act naturally as sub-wavelength optical gratings, providing the necessary phase-match conditions to couple the light into plasmons [47]. During the past years, digital disk media, such as CD, DVD or Blu-ray have been used as platforms for plasmonics systems, being used for biosensing or photo-catalytic applications [89]. Interestingly, a large increase of the transverse magneto-optic Kerr effect (TMOKE) can be observed in magnetoplasmonics crystals grown on commercial digital disks [90]. However, so far the possibility of controlling and tuning the magneto-optic enhancement on commercial digitals disks has not been fully addressed. Here, we used an angular-resolved reflectance spectroscopy, aiming to explore the effects of propagating surface plasmons into the enhanced transverse magneto-optic Kerr effect.

3.3 Experiment description

3.3.1 Sample preparation

To explore the transverse magneto-optical Kerr effect (TMOKE) response of magnetoplasmonic crystals grown on top of commercial disk gratings, we prepared the samples following the protocol described in section 2.1.4. Figure 3.1 (a) shows a schematics of the cross section of the final magnetoplasmonic system, while figure 3.1 (b) displays a SEM image taken from a coated DVD disk. The magnetoplasmonic multilayers were composed of Cr (4 nm) / Au (16 nm) / [Co (14 nm) / Au (16 nm)] x4 / Co (14 nm) / Au (7 nm), where Cr was selected as adhesion layer. The multilayer was deposited using electron beam evaporation with growth rates smaller than 0.1 nm per second. Such slow deposition rates are required to avoid the formation of crystallites, ensuring low roughness in the metallic layers.

FIGURE 3.1: In (a) the sketch of a commercial optical disk grating, in which the grating geometry is described by the parameters Δ, w, b and d, being the periodicity, width, base and depth of the grating. The panel (b) shows a SEM image from a coated DVD-disk.

The multilayer composition was previously optimized through FDTD simulations software, where the magneto-optical response was maximized by the aforementioned multilayer. Due to the presence of Co a trade-off between magneto-optical activity and the SPPs resonances was established, since the higher losses in the magnetic layer are harmful for the SPPs resonance conditions.

To characterize the magnetic behavior of the magnetoplasmonics crystals, magnetic hysteresis loops have been measured by a superconducting

quantum interference device (SQUID). The magnetic characterization revealed a shape-induced anisotropy along the grating long axis, i.e. lying parallel to the grating metallic wires. A detailed analysis of the magnetic response will be presented at the end of the chapter in figure (3.12).

3.3.2 Optical measurements

Using the monochromatic Fourier microscopy defined previously in section 2.2.3 provides access to a full set of angular and spectral information, allowing the reconstruction of magnetoplasmonic crystal band structure. The collimating nature of the objective lenses allows the projection of the p- and s-light polarization simultaneously, as shown in the schematic representation of the experimental setup (2.11). Note that in this Thesis we use the terms p- and s-polarization for transverse-magnetic (TM) and transverse-electric (TE) polarization states of light.

FIGURE 3.2: Schematics of the light cone after focusing through the objective lens. (a) By cutting the light cone of the incoming p-polarized light along different azimuthal angles, the optical responses for s- and p-polarized light can be obtained at different angles of incidence (purple and yellow shaded areas). Figures (b)-(c) show the projection on the sample surface of the initially p-polarized light. In (b), having $\psi = 0$, the polarization of the reflected light is contained in the plane of incidence, so that we map the optical response of p'-polarized reflected light. In (c) $\psi = 90$, and the projection of p-polarized light is perpendicular to the plane of incidence, enabling the measurement of the optical response for s'-polarization.

The experiments were carried out with the incoming light in a p-polarized state. The projection of the incoming beam through the objective lens onto the sample surface creates a cone of light, as displayed in green in figure 3.2 (a). The incident beam with initial p- light polarization is focused to single focal point. During this process, all the light is condensed, thus, the light polarization projection will be different for different incidence θ and azimuthal angles ψ. For instance, figure 3.2 (b) depicts the changes in the p-polarization axis, as a function of the angle of incidence, for a fixed azimuthal angle $\psi = 0^o$. In this particular case, the polarization of the reflected light is always contained in the plane of incidence and the only changes are with respect to the angles with the normal to the surface. Therefore, for the azimuthal angle $\psi = 0^o$, the polarization state of the reflected light is denoted by p'-polarization.

A different case is presented, however, when the initial p-polarized light is projected along the azimuthal angle $\psi = 90^o$, as shown in figure 3.2 (c). Now, the plane of incidence cut across this azimuthal angle is perpendicular to the projected polarization of light. Thus, by selecting $\psi = 90^o$ in the ensemble of collected data, we obtain information about the optical response that corresponds to s-polarized light, even though the initially incoming light is p-polarized.

(b)

Reciprocal space

FIGURE 3.3: Angular-resolve Fourier map from a calibration sample (Thorlabs GH13-24V), obtained at the wavelength of ($\lambda = 530$ nm).

The projections of p'- and s'-polarization can be observed by the purple and yellow shadowed areas in figure 3.3 (a). Therefore, the optical responses for s- and p-polarized light can be extracted simultaneously in one single experiment. We performed two sets of experiments, depending on whether the samples were exposed to air or to immersion oil. To do so, the measurements were done using two different objective lenses: one an oil immersion lens 63x with 1.4 N.A., what allows the projection of high angles of incidence ranging from ($\Delta\theta_{inc} \approx \pm 54^o$), while in air we used a 50x with 0.9 N.A., allowing an smaller range of angles of incidence ranging from ($\Delta\theta_{inc} \approx \pm 43^o$).

As described above, angular-resolved reflectance spectroscopy scans map the optical responses in the Fourier plane for the reflected s'- and p'-polarization modes as indicated by figure 3.3. To obtain the spectroscopic dependence, the maps shown in this figure are measured at every wavelength. As described in section 2.2.3 and next sections, the collections for all angular-resolved reflectance spectroscopy scans for all wavelengths allow the reconstruction of the photonic band structure.

3.3.3 Magnetoplasmonic photonic band structure

The relationship between freely propagating light and SPPs in a metal/dielectric interface is given by:

$$k_{SPP} = k_x \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}}, \qquad (3.1)$$

where $k_x = k_{in} \sin \theta$ is defined as the in-plane component of the light wave-vector (k_{in}) impinging into a surface with angle θ, as shown in figure 3.4 and, ε_d and ε_m are, respectively, the permittivity of dielectric and metallic environment.

FIGURE 3.4: In (a) the sketch of a commercial optical disk grating, in which the grating geometry is described by the parameters.

As previously mentioned in section 2.2.3, the diffracted modes of a periodic crystal can be calculated by the Bragg equation:

$$k_{in} = k_{dif} \pm m \frac{2\pi}{\Delta}, \qquad (3.2)$$

where, k_{in}, k_{dif}, are the incident and diffracted wave-vector, Δ the grating periodicity, and m the diffraction order mode, where m assumes integer values, i.e. $0, \pm 1, \pm 2, ...,$. However the Bragg equation can be rewritten, in terms of propagating SPPs within a periodic one-dimensional photonic crystal, thus, providing the diffraction of different plasmonic modes, which are given by the following relationship:

FIGURE 3.5: Light and plasmonic diffraction modes calculated from equations 3.2 and 3.3, for two different dielectric media: (a) microscopy oil with refractive index n_{oil} = 1.5, and (b) air with n_{air} = 1.0.

$$k_{SPP} = k_x \pm \eta_{SPP} \frac{2\pi}{\Delta}, \tag{3.3}$$

where $\eta_{SPP} = \pm 1, \pm 2, \pm 3, ...$ corresponds to the SPP excitation order.

The angle of emission of the diffracted propagating SPP modes can be inferred allowing the calculation of the plasmonic resonances. Using the aforementioned equations 3.2 and 3.3, allows the calculation of the excepted light- and SPPs- diffraction modes for a periodic photonic crystal. The results are shown in figure 3.5 where the analytically calculated diffraction- and SPPs- band diagram are presented for a 1D periodic crystals Δ = 740 nm, corresponding to the DVD disk periodicity. The diffracted modes calculated through 3.2 are shown in green, while the forward- and backwards propagating SPPs modes obtained from 3.3 are shown in red and blue. Such dispersion relations are calculated for two different dielectric environments: one corresponding to the microscopy oil (a) and the other for air (b).

The complex band diagram shown in figure 3.5, reflects the spectral and

angular dependence of plasmonic modes. This phenomena was initially observed by Wood [46, 91], who detected absorptive anomalies in the reflected light upcoming of a grating, being named after him as Wood's anomalies. In this work we exploited Wood's anomalies to enhance the magneto-optic activity in magnetoplasmonic photonic crystals.

The effects of the magnetoplasmonic band structure on the transverse magneto optical Kerr effect (TMOKE) response were studied. For that purpose, the TMOKE amplitude is defined as the average intensity at the positive $I(H^+)$ and negative $I(H^-)$ saturated magnetic states, shown respectively by the red and blue shadowed area in figure 3.6. The TMOKE amplitude can be computed by:

$$\tau = \frac{I(H^+) - I(H^-)}{I(H^{avg})}, \qquad (3.4)$$

where $I(H^{avg})$ consists of average intensity of light obtained during alternated current cycles of the magnetic field.

FIGURE 3.6: A conventional hysteresis magnetic cycle is shown, from which the positive $I(H^+)$ and negative $I(H^-)$ average intensity are extracted for the field intervals demarcated by red and blue shadowed regions.

3.4 Results and Discussion

3.4.1 Plasmonic enhancement

FIGURE 3.7: Panel (a) presents the angle-resolved maps of ∇_r (defined by equation 3.5) of a coated non-corrugated polycarbonate film, measured around normal incidence. While in (b) a corrugated photonic crystal with similar coating displays magneto-optic activity.

Figure 3.7 presents angle-resolved reflectance maps of the magnetoplasmonic multilayer of composition Cr (4 nm) / Au (16 nm) / [Co (14 nm) / Au (16 nm)] x4 / Co (14 nm) / Au (7 nm), grown on top of a continuous layer of polycarbonate shown in 3.7 (a) and a CD grating with periodicity of $\Delta = 1200$ nm shown in (b).

Figure 3.7 shows, the difference of the intensity of the reflected p-polarized light measured at positive and negative saturation magnetic fields for all wavelengths and accessible angles of incidence:

$$\nabla r = I(H^+) - I(H^-), \tag{3.5}$$

displaying the different between emitted photons under a magnetic field switch, in which no normalization process is applied. A simple inspection of figure 3.7 reveals immediately that ∇r is virtually null for the unpatterned multilayer throughout the entire range of analyzed wavelengths and angles of incidence (panel (a)). In contrast, the map measured in the patterned magnetoplasmonic structure reveals substantial changes of the reflectance for particular spectral regions (panel (b)).

As shown in figure 3.5 SPP resonances are expected to exist for the patterned surfaces in the range of angles of incidence and wavelengths studied in the angular-resolved reflectance maps shown in figure 3.7. This suggests

and obvious link between the observed enhanced magnetoreflectance and
plasmons. This leaves the question open about whether localized or prop-
agating plasmons are responsible for the increased magneto-optical signal.
To decide on this issue, a proper experiment is to analyze the reflectance for
different polarizations of light, as propagating SPPs can be excited only by
p-polarized light.

To answer this question, we measured a Cr (4 nm) / Au (16 nm) / [Co (14
nm) / Au (16 nm)] x4 / Co (14 nm) / Au (7 nm) multilayer grown on a DVD
optical disk, where the grating has a periodicity of $\Delta = 740$ nm. These mea-
surements were done with the sample exposed to air, without immersion
oil. Figure 3.8 shows the angular-resolved reflectance and TMOKE maps
measured with p-polarized light (panels (a) and (c)) and s-polarized light
((b) an (d)). Figure 3.8 (a) reveals absorptive dispersion curves that can be
attributed to the excitation of propagating SPPs. Indeed, the observed de-
pendence on wavelength and angle of incidence matches well with the SPPs
band diagram calculated with equation 3.3 shown by the dotted gray lines.
Moving to figure 3.8 (b), we observe that the reflectance plot correspond-
ing to s-polarization does not reveal any clear absorption line, hinting at the
absence of any plasmon excitation. The enhanced absorption observed at
shorter wavelengths may be attributed to the intrinsic optical response of
Au, which is known to absorb light at those frequencies.

The TMOKE response matches well with the plasmon band diagram. In
particular, a remarkable TMOKE response is observed along of the disper-
sion lines calculated for propagating SPPs, along which high TMOKE am-
plitudes in the the order of 0.5% are obtained. However, such high TMOKE
amplitudes disappear whenever the light is switched to s-polarization, as
shown by figure 3.8 (d). Figure 3.8 (e) shows a profile of the TMOKE signal
extracted from panel (c), and corresponding to the wavelength of $\lambda = 700$
nm, revealing a derivative-like lineshape near the plasmonic resonances,
which are indicated by the blue and red dots. We will see in the next sec-
tion that this lineshape originates from the displacement of forward and
backward- propagating SPP resonances induced by magnetic fields. We
conclude, therefore, that propagating SPP resonances are at the origin of
the observed TMOKE in these magnetoplasmonic crystals.

FIGURE 3.8: Angular-resolved reflectance spectroscopy and TMOKE maps are shown for the magnetoplasmonic Cr (4 nm) / Au (16 nm) / [Co (14 nm) / Au (16 nm)] x4 / Co (14 nm) / Au (7 nm) multilayers measured with p-polarized light (panels (a) and (c)) and s-polarized light ((b) and (d)). (e) Profile of the TMOKE signal measured at $\lambda = 700$ nm acquired from dashed line shown in (c).

3.4.2 Plasmon resonances under magnetic field.

In the previous subsection, SPPs were identified as the origin of the enhancement of TMOKE, hence a proper analysis is required for an understanding of the effects of SPPs on the magneto-optic properties. The first to note is that the TMOKE response as a function of the angle of incidence shows a sign reversal in the vicinity of the SPP resonances (figure 3.8 (e)). This is a well-known result in the literature, where the observed differential lineshape is caused by a shift in the SPPs frequency induced by the magnetization in the grating [18, 19, 51, 52].

A schematic representation of such phenomenon is shown in figure 3.9, where the shift in the plasmon resonance frequency is shown for a forward- and backward-propagating SPPs respectively in (a),(c) and (b),(d). Whenever a magnetoplasmonic system is set to a magnetized state H^- or H^+ its resonance conditions will be shifted, changing the frequency distribution as shown in figure 3.9 (a) for backward propagation, where the initial ω corresponding to a non-magnetic or demagnetized state is then shift towards ω^+ or ω^-. A similar effect happens for forward propagation of plasmons (figure 3.9 panel (b)). Figure 3.9 (c) and (d) represent

FIGURE 3.9: A schematic representation of the the shift in the plasmon resonance frequency at positive and negative saturation ω^+ and ω^- is shown in the panels (a) and (b) for forward and backwards propagating SPPs. In (c) and (d) the differential plots originated by the subtraction of ω^+ and ω^- are presented.

the angular shifts that occur for opposite magnetization states for backward and forward propagation of plasmons, respectively. The subtraction of the ω^+ and ω^- curves is proportional to the TMOKE signal, which gives rise to the characteristic derivate-like lineshape observed near the frequencies of plasmon resonances. Hence, performing a differential plot between the

plasmon resonances as represented in figure 3.9 (c) and (d) shows a non-reciprocal propagation between forward and backwards propagating SPPs.

A non-reciprocal behavior under the influence of magnetism can be described qualitatively in terms of the Lorentz force acting on the collective motion of electrons, that can be written as:

$$\vec{F}_L = q(\vec{E} + \vec{v_{SPP}} \times \vec{B}), \tag{3.6}$$

where q, $\vec{E}$, $\vec{v}_{SPP}$ and $\vec{B}$ are respectively the electron charge, electric field, plasmon velocity and the magnetic field. In the absence of magnetism, we have:

$$\vec{F}_E = q\vec{E}. \tag{3.7}$$

While, the pure magnetic component of Lorentz force 3.6 can be reformulated as:

$$\vec{F_M} = q\mu_o\vec{v}_{SPP} \times \vec{M}, \tag{3.8}$$

that describes the force exerted by a magnetic field $\vec{H}$ on the propagating surface plasmons. The geometry of a 1D photonic crystal constrains the direction of magnetization and SPP propagation. If we consider that M and v_{SPP} are along $\hat{y}$ and $\hat{x}$, the equation 3.8 can be rewritten as:

$$F_M\hat{z} = q\mu_o v_{SPP}\hat{x} \times \vec{M}\hat{y}. \tag{3.9}$$

Fixing a magnetization state, reduces the expression to:
$F_M\hat{z} = \pm q\mu_o v_{SPP}M\hat{z}$. By defining $\vec{F} = \vec{F}_E + F_M\hat{z}$ we obtain:

$$\vec{F}_L(M^+) = q\vec{E} \pm q\mu_o v_{SPP}M\hat{z} \quad \text{and} \tag{3.10}$$

$$\vec{F}_L(M^-) = q\vec{E} \mp q\mu_o v_{SPP}M\hat{z}, \tag{3.11}$$

corresponding to the Lorentz Force for the positive and negative magnetization states $\vec{F}_L(M^\pm)$. The derived expressions 3.10 and 3.11 provide a qualitative understanding of the nonreciprocal effects on surface plasmons

at a given magnetic state, originated from the magnetic component of the
Lorentz force.

3.4.3 Measurements in air and oil

We discuss the plasmonic response of a magnetoplasmonic system on a
DVD grating. The magnetoplasmonic response was measured in two dif-
ferent dielectric environments, corresponding, respectively, to the sample
immersed in oil or exposed to air. We start the analysis with magnetoplas-
monic DVD grating exposed to microscopy oil immersion. In this case, the
objective lens allowed to collect light for angles of incidence in the range of
($\theta < 54^o$). Fourier angular-resolved spectroscopy maps are shown in figure
3.10. Panel (a) shows a monochromatic Fourier angular-resolved TMOKE
response for the wavelength of $\lambda = 800$ nm, where a set of plasmonic res-
onances modes can be identified with orders $\eta = \pm 1, \pm 2$. Panel (b) shows
the reflectance map where the first-order diffracted plasmons can be iden-
tified in the absorption pattern, in good agreement with the corresponding
calculated dispersion lines (obtained from equation 3.3).

Figure 3.10 (c), presents the Fourier angular-resolved TMOKE mapped
as a function of wavelength and angle of incidence. The most remarkable
result arises in the vicinity of the crossings between plasmon lines with
diffraction orders $\eta = \pm 1$ and $\eta = \mp 2$. These regions reveal a correlation
between the SPPs propagating modes and the TMOKE response. Such an
interplay between TMOKE and the plasmon modes result in large magneto-
optic TMOKE signals up to about 0.4%. Such large values are comparable
with the TMOKE enhancement found in experiments made in transmission
instead of reflection [18, 50, 92]. Note that such values are larger than the
intrinsic TMOKE response of unpatterned magnetic metals which, even at
large angles of incidence (where the signal is expected to be the largest) typ-
ically are not larger than 0.03%. What is remarkable is that such TMOKE
signals are measured at relative moderated values of the angle of incidence
(e.g., TMOKE is 0.4% at $\theta = 10^o$). This observation is relevant for techno-
logical applications, since obtaining large signals at low angles of incidence

FIGURE 3.10: Magnetoplasmonic crystal based on DVD exposed to immersion oil. (a) A TMOKE space map is shown for the wavelength of $\lambda = 800$ nm, represented by the dashed line in panel (c). In panel (b) we present the reflectance map together with the plasmonic dispersion lines calculated using equation 3.3. Panel (c) presents a TMOKE angle-resolved map with forward- and backwards- plasmon propagating modes represented by the blue and red dotted lines.

may be of interest, especially in conditions of quasi-normal angles of incidence. This is typically the configuration that was used, e.g., for the readout of optic storage disks [93, 94]. In the next paragraphs we discuss how commercial digital disks can be used to achieve large signals at virtually zero-angle of incidence. The approach that we used was to change the dielectric environment by replacing microscopy oil ($n = 1.5$) with air ($n = 1.0$).

We then embarked on the investigation of TMOKE response with the photonic crystal exposed to air, with attention to quasi-normal incidence. And here it is precisely where lays our most interesting outcome, since we demonstrate that unexpectedly large values of TMOKE can be measured at quasi-normal incidence ($\theta = \pm 1.3^{o}$), as identified in figure 3.11. By changing the environmental permittivity, the dispersion lines of the different plasmonic orders are changed. This is illustrated in figure 3.11 (a), which shows

a monochromatic Fourier angular-resolved TMOKE response, for the wavelength of $\lambda = 800$ nm, from which the dashed arrows represent the projection of light p-polarization. Panel (a) reveals the clear influence of plasmonics modes into the magnetic optical response. In 3.10 (b) the reflectance plot is shown, revealing strong absorption near to the calculated plasmonic modes shown by the gray dotted lines.

The analysis of the TMOKE response mapped for wavelengths and angles of incidence (figure 3.11 (c)), shows that the TMOKE amplitude for the lowest angles of incidence ($|\theta| < 5$), is unexpectedly large. The enhancement occurs for values of the wavelength and angles of incidence near the region where plasmon lines of orders $\eta = \pm 1$ cross each other. More specifically, we extracted the enhanced signals generated by the plasmonic resonances at almost normal incidence from the data shown figure 3.11 (c) (vertical dotted lines), and shown by a cross-sectional plot in figure 3.11 (d). In this figure we see that the spectral dependence of TMOKE amplitudes for angles of incidence ($\theta = \pm 1.3^o$) result in a remarkable amplification of the magneto-optic activity, that peaks 0.5%, near the cross of the plasmonic bands $\eta = \pm 1$. This is a remarkable outcome, as it shows that large TMOKE responses can be obtained in magnetoplasmonic crystals at quasi-normal incidence which, as aforementioned, may be of interest for some applications. Note that the essential ingredient to obtain these remarkable responses is the fact that first order plasmon lines run up to vanishingly small angles of incidence at some wavelengths (as shown figure 3.11). This is an important guideline, as first-order plasmon orders are expected to be more effective to enhance the optical signals than lower orders.

We finish this section by discussing briefly the TMOKE maps for large angles of incidence ($-20 < \theta$ and $\theta > 20$). We see that a noticeable TMOKE activity is only observed at short wavelengths. Near this region the plasmonic modes corresponding $\eta = \pm 2$ are found, but whether the enhanced signal is due to second-order plasmons or it is primarily the effect of the intrinsic signal at large angles of incidence is not obvious.

As a conclusion, we may say that our results provide general rules for magneto-optic enhancement and, in particular, show the potential of standard commercial disks as platforms for enhanced magneto-optic devices.

FIGURE 3.11: Magnetoplasmonic crystal based on DVD exposed to air. (a) Monochromatic TMOKE Fourier map shown for $\lambda = 800$ nm (corresponding to the dashed line in panel (c). (b) Reflectance map with the corresponding plasmonic band diagram superposed to it in gray dotted lines. (c) TMOKE map showing the effects on the magneto-optic signal of forward- and backward- propagating plasmon modes of different orders represented by blue and red dotted lines, respectively. (d) Profiles of the TMOKE amplitude at quasi-normal incidence.

3.4.4 TMOKE hysteretic loops

In the previous sections we have discussed the effects of SPPs on TMOKE by drawing the attention to the magnetic saturated states. Here we discuss the analysis of TMOKE signals for values of the magnetization all over the whole hysteresis cycle. To analyze the hysteretic dependence, we narrowed down the study to specific values of wavelength and angles of incidence

(3.12 (a)) measured in the magnetoplasmonic crystal exposed to immersion oil. By tracking down the values of TMOKE in this narrow region we can reconstruct the hysteresis loops for different angles of incidence, as shown in figures 3.12 (b)-(d).

FIGURE 3.12: The data shown in this figure was obtained from experiments done in a magnetoplasmonic crystal exposed to immersion oil. (a) Angle-resolved TMOKE map. The squared areas were chosen for the analysis of the magnetic field dependence of TMOKE. (b-d) Hysteresis loops acquired by SQUID (dashed lines) and the TMOKE (squares in panel (a), corresponding to different angles of incidence).

Hysteresis loops are shown in figure 3.12 (b)-(d) for different angles of incidence. Panels (b) and (d) show TMOKE loops compared with SQUID measurements presented as black-dashed lines. The reversal in sign of the hysteresis loop is evident and its origin arises from the sign of the plasmonic order $\eta = \pm 2$. Panel (c) displays the loop measured at zero angle of incidence. Note that for the wavelengths analyzed in figure 3.12 -corresponding to measurements in the sample exposed to immersion oil- there is no plasmon resonances at zero angle of incidence, thus explaining the absence of signal.

3.5   Conclusions

The investigation of the transverse magneto-optical Kerr effect has been presented for metallic multilayers deposited on top of the grating structure of commercial disks. Wood's anomalies, related to the excitation of surface plasmon polaritons, are distinguishable in angle-spectral-resolve reflectance maps. We demonstrated that the TMOKE signal is enhanced near the dispersion relations predicted for propagating plasmons of different orders. An outstanding result arises for samples exposed to air instead of oil, where plasmons drive a remarkable enhancement of TMOKE at quasi-normal incidence. These results demonstrate the potential of a proper engineering of magnetoplasmonic crystals on commercial digital disks has a large potential for nanodevices exploiting TMOKE responses.

Chapter 4

Non-Reciprocal Diffraction in Magnetoplasmonic Gratings

4.1   Abstract

Phase-matching conditions used to bridge the wave vector mismatch between light and surface plasmon polaritons (SPPs) have been exploited recently to enable nonreciprocal optical propagation and enhanced magneto-optic responses in magnetoplasmonic systems. Here we show that using diffraction in conjunction with plasmon excitations leads to a photonic system with a more versatile and flexible response. As a testbed we analyzed diffracted magneto-optical effects in magnetoplasmonic gratings, where broken time-reversal symmetry induces frequency shifts in the energy and angular spectra of plasmon resonances that result in exceptionally large responses in the diffracted magneto-optical effect. The concepts we present here can be exploited for the development of non-reciprocal optical devices that exploit diffraction to achieve tailored electromagnetic responses.

4.1.1   Chapter highlights

- Use of angle-resolved Fourier reflectance spectroscopy to analyze diffractive modes of light and plasmons.

- Report on the interplay between magnetoplasmonic modes and diffraction.

- The amplification of the transverse magneto-optical Kerr effect at plasmonic resonances on the diffracted light.

- The results described here open new perspectives on diffraction-based magneto-optical isolators.

4.2 Introduction

Surface plasmon resonances confine light into subwavelength dimensions which results in a corresponding increase in electric field intensities near plasmonic nanostructures [95]. This enables the use of surface plasmon resonances to enhance light matter interactions [96] and has prompted research into e.g. plasmon-assisted sensing [97, 98, 99, 100], strong coupling [101, 102, 103] and lasing [104, 105, 106]. Beyond these properties, novel device concepts offer expanded functionalities by incorporating materials responsive to external stimuli. Particularly, magnetism is considered to have potential technological impact for, e.g., labelfree detection using magnetoplasmonic sensors [77, 107] or ultracompact nonreciprocal nanophotonic devices [108]. These new developments benefit from enhanced light-matter interaction, realized by exciting surface plasmon resonances in or in immediate vicinity of magnetic materials [109]. A wide range of nanophotonic devices that combine plasmonics with magneto-optics have been demonstrated, ranging from very simple, such as nanoparticles made of ferromagnetic metals [82, 110, 111], to more complicated, e.g. structures that incorporate noble metal nanoparticles with magneto-optically active materials in dimers or far-field coupled arrays [20, 21].

Metallic diffraction gratings are one of the most common configurations to study surface plasmon polaritons (SPPs). They are also known as grating couplers due to their ability to couple light propagating in free space into SPPs and vice versa [44, 45, 46]. Nonreciprocal plasmon propagation and enhanced magneto-optical effects have been demonstrated in magnetoplasmonic gratings, fabricated of magnetic metals [18] or noble metals in contact with magneto-optical garnet materials [19, 35, 36]. Using such gratings, control over SPP excitation conditions by an external transverse magnetic fields

was demonstrated [18, 19]. Yet, all these previous studies focused on the excitation of plasmons and its consequences on the optical properties, but did not pay attention to a central property of a periodic grating: diffraction. Diffraction in magneto-optically active systems is an important phenomenon that is used, e.g., in diffracted magneto-optical effect (DMOKE) to probe magnetization reversal dynamics of submicron-sized magnetic patterns [38, 39, 40, 41, 42].

All things considered, we can say that up till now the mutual interplay of three important phenomena, i.e., plasmonics, magneto-optics and diffraction, has not been investigated in a single system. We show here that combining all the aforementioned properties provides new paths for flexible design of functional nanophotonic systems. To corroborate this point, we study the interplay of plasmon resonances and magneto-optics and their influence on diffraction properties of a simple diffraction grating. Despite its simplicity, it supports surface plasmon polaritons propagating both into backward and forward directions, enabling us to easily access how the nonreciprocal magnetic modulation of SPPs influences the diffracted beams.

4.3   Experiment description

4.3.1   Sample preparation

FIGURE 4.1:   Diffraction grating sketch: In (a) the grating parameters are shown where Δ, w, b and d are the periodicity, width, base and depth of the grating. The panel (b) shows a SEM image of the deposit magneto-plasmonic layer.

A multilayer being composed of Cr (4 nm) / Au (16 nm) / [Co (14 nm) / Au (16 nm)] x4 / Co (14 nm) / Au (7 nm), is deposited over a SrTiO$_3$ substrate by electron beam evaporation. Afterwards the sample is coated by an electron-sensitive polymer, and subsequently electron-beam lithography is performed to pattern one-dimensional photonic crystals. The latter are structured through ion milling, where the desired grating depth d can be controlled (a more detailed sample preparation protocol can be found in section 2.1.4). The resulting pattern can be observed in figure 4.1 (a), from which the grating parameters can be identified. Such parameters can be easily tuned through electron beam lithography, which allows a precise control of the periodicity Δ, width w and base b. This, in combination with the ion-milling technique, allowed a full control of the grating geometry. During the first part of this chapter only one grating is discussed, whose parameters are listed below. A SEM microscopy image from such structure is shown at 4.1 (b).

- Periodicity - $\Delta = 1000$ nm

- Width - $w = 800$ nm

- Base - $b = 200$ nm

- Depth - $d = 120$ nm

These grating parameters were selected aiming to maximize magneto-optical effects and to increase the fraction of diffracted light. It is known that the details of the grating morphology, such as the width and base of the gratings, have a noticeable influence on the propagation of plasmons and the efficiency of diffracted light (discussed in section 5.2). In order to deal with this complexity and to have guidelines for the design of the photonic crystals, we resorted to FDTD simulations using Lumerical (not shown) [112].

4.3.2 Diffraction measurements

We have exploited angle-resolved spectroscopy (described in chapter 2.2.4) to image the diffracting magnetoplasmonic crystals both in real and reciprocal space. A critical point is that we could decrease the spot size of the incoming light at the back aperture of the objective lens, as shown in figure 4.2 (a). Using this capability, only a narrow range of angles of incidence is projected to the sample. This procedure was fundamental to identify different diffraction modes of different orders $m = 0, \pm 1, \pm 2, ...$, in which light is radiated at different angles of emission.

FIGURE 4.2: Depiction of the rear aperture of an objective lenses. Panel (a) presents an incident light beam that is project to the sample. Panel (b) depicts the light diffracted from the one-dimensional photonic crystal, with diffracted orders corresponding to $m = 0, \pm 1, \pm 2$.

It is important to stress that in the experiments discussed in this chapter, the diffraction patterns were analyzed for incoming light at quasi-normal incidence, which defined a light cone with angles between about -5° to 5°,

resulting in a beam spot of around 500 μm of radius over the backside of the objective lens. As a consequence of its interaction with the grating, the diffracted modes were radiated at different angles of emission. From the detected intensity at the CCD camera, we could select the light that was emitted at specific angles, so that each diffracted mode ($m = \pm 1, \pm 2$) could be analyzed individually, including the specular reflection characterized by the diffraction mode $m = 0$, as seen in figure 4.2 (b). As an illustration, we show in figure 4.3, an experimental snapshot taken in the aforementioned grating at the wavelength $\lambda = 785$ nm, where diffracted modes of different orders could be analyzed individually.

FIGURE 4.3: A diffraction image taken from a photonic crystal with periodicity $\Delta = 1000$ nm at the wavelength of $\lambda = 785$ nm.

The signatures of SPP excitation can be observed in angular-resolved reflectance scans shown in figure 4.4 (a), which display the reflectance for the specular reflection and diffracted modes. For clarity, the plasmonic band modes, calculated using equation 3.3 (defined in chapter 3), are indicated in this figure by the blue and red dotted lines along with the diffracted modes shown in green. For convenience, in figure 4.4 the specular reflection mode $m = 0$ was multiplied by 0.2, thus scaling it to an amplitude comparable to the diffracted modes $m = \pm 1$. The specular mode reveals a clear trace of absorption in the wavelength range of $\lambda = 730$ to 850 nm, therefore matching with the expected plasmonic band diagram. In the same specular region the diffraction orders $m = \pm 1$ display a more evident plasmonic absorption, that is located at the crossing of the forward- and backward-propagating SPP modes $\eta = +1, -3$ and $\eta = -1, +3$.

A precise observation of SPP resonances can be seen in figure 4.4 (b), that displays cross-sectional plots extracted from 4.4 (a), for the wavelengths

FIGURE 4.4: Panel (a) shows an angular resolved reflectance map where the diffracted light intensity is displayed. The calculated band diagram for propagating surface plasmons indicated by the blue and red dotted lines are shown in superposition to the diffracted data. (b) Cross-sectional plots extracted from the horizontal dashed lines in panel (a), which correspond to the wavelengths of $\lambda = 745$, 785 and 825 nm. In addition, the location and direction of the propagation of surface plasmons is indicated by blue and red arrows.

marked by the white dashed lines. Hence figure 4.4 (b) shows the angular resolved normalized diffracted intensity for three different wavelengths $\lambda = 745$, 785 and 825 nm, corresponding to the curves shown in blue, red

and green respectively. In addition 4.4 (b) displays the accurate location of the forward- and backward- propagating SPP, being represented by red and blue arrows. The absorption trend can be observed at the crossing with the plasmonic modes, where the most absorptive region can be found at the wavelength of $\lambda = 785$ nm, which is located at the crossing of two plasmonic modes.

The experimental data displayed in figure 4.4 offers a unique approach to observe specular reflection and diffraction modes simultaneously, providing a precise way to identify plasmonic resonances and enabling the quantification of their interplay with the reflected and diffracted light. In the remaining of the chapter we discuss how magnetism can play a role in these relationships.

4.3.3 Diffracted Magneto-Optical Measurements

FIGURE 4.5: In (a) a light beam having wavevector k_0 impinges over a magnetoplasmonic multilayer system, which supports backward- and forward- propagating SPP modes shown in blue and red waves. The TMOKE response arising from the diffracted beams $m = \pm 1$ shown as circular diffracted beams reveals a unusual magneto-optic behavior that arises from the interaction of plasmons with the magnetized medium. The TMOKE amplitudes obtained on the circular spots at (a) are translated as changes in the magneto-optic sign, resulting in a sign reversal of the hysteresis loops, as shown by (b).

Figure 4.5 (a), depicts schematically the interplay between diffraction and plasmons in a magnetoplasmonic crystal. In the figure, backward- and forward- propagating SPPs are shown in blue and red respectively. After the interaction with the photonic crystal, the outgoing light is decomposed into the specular and four observable diffraction modes with orders $m = 0, \pm 1, \pm 2$, from

which its corresponding TMOKE response is calculated through the expression: $\tau = \frac{I(H^+) - I(H^-)}{I(H^{avg})}$ (defined previously in equation 3.4). The magneto-optic activity on the diffracted order can be observed in the circular plots in figure 4.5 (a), corresponding to the spacial distribution of TMOKE effects into a diffracted beam. Such response is obtained through the application of the previously mentioned expression for τ into the data obtained by Fourier maps measured at different magnetic fields. The corresponding magneto-optic sign and amplitude shown in figure 4.5 (a), are associated with the orientation and amplitude of the hysteresis loops shown in figure 4.5 (b). Besides that, an extraordinary response arising from the diffracted modes $m = \pm 1$ is observed in the circular plots, presenting a sequence of changes in the TMOKE sign and amplitude.

The results schematized in figure 4.5 can be understood in terms of light interacting with the photonic crystal in two processes, depicted in 4.6. In the first place, the incident light with wavevector k_0 can be diffracted and emitted at different angles for different orders of diffraction (figure 4.6 (a)). On the other hand, light can excite SPPs of opposite directions (figure 4.6 (b)), which can be also diffracted by the crystal, with different orders emitted at different angles (figure 4.6 (c)). We will see that the diffracted plasmonic modes are influenced strongly by the magnetization, in comparison with the diffracted light. In particular, the SPP wave-vector k_{SPP} is modified by the magnetization according to the following expression [18, 51, 52, 92]:

$$k_{SPP}(M) = \pm k_{SPP}\left(1 \mp \frac{M\varepsilon_{xy}}{\beta}\right),\qquad(4.1)$$

where $\beta = \sqrt{\varepsilon_m \varepsilon_d}(1 - \varepsilon_m^2/\varepsilon_d^2)$, ε_{xy} is the off-diagonal component of the dielectric function tensor and ε_m, ε_d are the dielectric and metallic layer dielectric functions, and M is the magnetization. Thus, whenever a SPP decays back into a photon, its original wave is changed, being shift from k_0 to $k_\pm$ depending on the external magnetic field and SPP propagation direction. As a result, the angle of emission of photons generated by the diffracting plasmons depends on the direction of the plasmon propagation, giving way to an angular displacement of the two possible SPP propagation states, as shown in 4.6 (c) (discussed previously at section 3.4.2).

FIGURE 4.6: (a) The incident light is diffracted by a magnetoplasmonic photonic crystal. (b) Part of the incident light can be converted into forward- and backward- propagating surface plasmons, which are strongly interacting with the magnetized layer. (c) Under a magnetized sample, the angle of emission of plasmons scattered back to light are different for opposite directions of propagation.

The response for the diffracted and specular reflections is shown in figure 4.7 (a), where the TMOKE amplitude is plotted over a wide range of angles of incidence and wavelengths. In this figure, the forward and backward propagations of SPP modes are represented by the blue and red colored dotted lines. The observation of this experimental data reveals immediately that the magneto-optic response appears near a plasmonic mode, being vanishingly small elsewhere, except for the spectral regions of short wavelength in the diffracted modes.

To properly depict the exposed information let us take a look on the diffracted orders individually, starting with the specular reflection, i.e. $m = 0$. At almost normal incidence the TMOKE response peaks to 1% near $\lambda = 785$ nm that corresponds to the crossing of two plasmonic propagating modes $\eta = \pm 2$. Moving towards the diffracted modes $m = \pm 1$, a nonreciprocal behavior can be observed directly from the TMOKE response. As expected, we observe that the sign of the TMOKE response is reversed for diffracted modes of opposite signs $m = \pm 1$, in agreement with the expected sign of the ordinary TMOKE, which is given by $\vec{\sigma} = \vec{k}_{dif} \times \vec{M}$, where k_{dif} is the wavevector of the diffracted light. In addition, the direction of propagation of SPPs determines the sign of TMOKE sign which is given by $\vec{\sigma} = \vec{k}_{SPP} \times \vec{M}$, where k_{SPP} is the wave-vector of the surface plasmons.

FIGURE 4.7: Panel (a) presents an angle-resolved spectrometry plot of TMOKE, where specular and diffraction modes can be also observed. In the plot, plasmon bands of different orders -calculated using equation 3.3 -are also indicated. (b) TMOKE cross-sectional plots were extracted from the horizontal dashed lines shown in panel (a), corresponding to the wavelengths of $\lambda = 745$, 785 and 825 nm. For clarity, the angular TMOKE corresponding to $\lambda = 745$ and 825 nm was vertically displaced.

The most remarkable observation is the increase of the TMOKE amplitude at the diffracted modes $m = \pm 1$, where the maximum TMOKE amplitude is found at $\lambda = 785$ nm peaking up to 3%. These TMOKE values are two orders of magnitude higher than the ones obtained from conventional magnetic materials, being in the order of 0.03% [113]. Our samples

present TMOKE values comparable to the highest values obtained in the literature in structures containing magnetic complex oxides [19, 35]. We stress that these previous results were reported for relatively thick materials, with thickness in the order of the micron. In contrast, our large TMOKE values are obtained by reflecting light on nanometric thin films. Beyond that, the TMOKE reported here is obtained from simple (polycrystalline Au and Co deposited at room temperature). Both simplicity in materials and heterostructures with nanometric thickness make our approach appealing for their use in technological applications.

From figure 4.7 (a), cross-sectional plots have been extracted for wavelengths $\lambda = 745$, 785 and 825 nm marked by the black horizontal dashed lines, which are displayed in figure 4.7 (b). These cross-sections reveal evident magnetoplasmonic features, with the characteristic derivative-like lineshape that we have discussed previously (chapter 3). Figure 4.7 (b) also provide the accurate location of plasmon resonances, marked by the red and blue arrows. For instance, the most prominent derivative like curve is located around the wavelength $\lambda = 785$ nm, at the crossing of two plasmon lines of order $\eta = \pm 2$ resulting in a maximized TMOKE amplitude. Meanwhile, at the wavelengths $\lambda = 745$ and 825 nm the plasmon resonances are split, which results in a smaller TMOKE amplitude.

To complete the analysis, we split the angular response into positive and negative angles of incidence around the direction along the normal to the surface $(-5 < \theta < 0)$ and $(0 > \theta > 5)$, respectively. In the subsequent analysis, we consider the light specularly reflected or diffracted that comes exclusively from either positive or negative angles of incidence (see the dashed areas in 4.8). The diffracted beam is decomposed in two sections, one being generated by positive-, while the other by negative-angles of incidence, as indicated by a diagonal dashed line. Thus, the TMOKE amplitude can be calculated for photons arriving with positive and negative angles of incidence. This is done through the summation of all the pixels indicated by the area inside the dashed box. This operation is repeated for all wavelengths, for the diffracted modes corresponding to $m = 0, \pm 1$ as indicated by dashed lines in figure 4.7 (a). The result is shown in a spectral dependent TMOKE

plot shown in 4.9, which displays the average TMOKE amplitude as a function of wavelength for the different diffraction orders $m = 0, \pm 1$.

FIGURE 4.8: Schematics of the integration method. The diffracted light corresponding to negative and positive angles of incidence from a normal-incident beam is collected individually, for each wavelength, the average TMOKE amplitude is collected in the regions marked by the dashed squares.

Figure 4.9, which shows data collected exclusively from light incident at negative angles, reveals that the the sign of the TMOKE of specular and diffracted modes are inverted from each other. This observation can be understood from the fact that the TMOKE amplitude represents the changes in the intensity from a positive and negative magnetic states, $\tau_I = \frac{I(H^+) - I(H^-)}{I(H^{avg})}$. Therefore if TMOKE is positive for the specular mode $m = 0$ (e.g., in the spectral region of $\lambda \approx 750$ - 830 nm, see figure 4.9), this means that the intensity collected for H^+ is larger than for H^-, i.e., $I(H^+) > I(H^-)$. The latter

FIGURE 4.9: Spectral average TMOKE response collected exclusively for negative angles of incidence, the areas from which the TMOKE was calculated are marked by the dashed rectangles in figure 4.8.

condition implies that more light is specularly reflected for field H^+ than for H^-. One would expect, therefore, that less light can be emitted in diffracted modes for H^+ than for H^- in the vicinities of a forward-propagating SPP. As a result, for diffracted modes the intensity collected at H^- is expected to be larger than for H^+, i.e., for diffracted modes $I(H^+) < I(H^-)$. Consequently, a negative TMOKE is expected for diffracted modes in the spectral region of $\lambda \approx$ 750 - 830 nm, in agreement with the data shown in figure 4.9.

4.4 Conclusions

We have explored how magnetization can be used to modulate the plasmon excitation conditions so that, in combination with diffractive properties, they can be used as a powerful pathway towards flexible design of magnetoplasmonic systems. Our study makes obvious some consequences of magnetically induced shift of the SPP resonance. Firstly, we emphasize that the presence of magnetization not only shifts the SPP excitation conditions in the energy space, but also induces shifts in the angular space. They are explicitly demonstrated in derivative TMOKE line-shapes seen at SPP resonances in Figure 4.7 (b).

As a second observation, we call the attention to the fact that the presence of SPPs results in significant changes in the magneto-optical activity in the diffracted beam. With zero angle of incidence, a maximum of 3% intensity change between the opposite magnetization states in diffraction is observed. This can be considered a large magnitude for TMOKE. In conventional magneto-optical geometries, the intensity modulation in transverse magneto-optical Kerr effect is usually less than 0.1% while for magnetoplasmonic systems higher numbers have been reported, up to 2% in magnetoplasmonic gratings. In transmission geometry magnetoplasmonic systems have been shown to support even higher modulation, though accompanied with a large losses.

Our experiments were performed with a simple metallic grating in order to highlight the interaction between the three phenomena under investigation: plasmons, magneto-optics and diffraction. However, we anticipate

that our results can be generalized and used to design more complex diffractive elements, such as many plasmonic metasurfaces, where they could find use in designing non-reciprocal, isolating devices.

Chapter 5

Optimizing TMOKE in Plasmonic Gratings: Towards One-Way Plasmons

5.1 Abstract

Here we analyze two important aspects of grating couplers in relation to TMOKE signals. At first, maximizing the reflectance and TMOKE simultaneously is necessary for applications which seek to obtain the largest possible responses. We have therefore explored how optimal conditions depend on the photonic crystal geometrical parameters, from which a relation between reflectance and TMOKE responses is measured in specular and diffracted modes. We conclude that the reflectance and TMOKE are optimized using light at normal incidence and collecting the specular light along the same direction.

On the other hand, we have explored how excitation of SPPs with off-normal incident light can be exploited to achieve one-way propagation. In our study we demonstrate that one-way SPPs and associated large TMOKE signals can be achieved under these conditions. Interestingly, we found remarkably large TMOKE responses (above 4%) enhanced by one- way propagating plasmons. This observation opens up very interesting perspectives in nanophotonics, as it might be exploiting in on-chip grating couplers to external optical links, eventually enabling one-way propagation in integrated photonic circuits.

5.1.1 Chapter highlights

- Conditions for simultaneous maximization of reflectance and TMOKE.

- Excitation of individual forward- and backward- SPP propagating modes.

5.2 Geometry effects: maximizing the reflectance-TMOKE relation

In the previous chapter we have analysed how plasmon resonances can enhance the TMOKE response of magnetoplasmonic crystals. There we focused our attention to a specific grating of given width and periodicity. Yet, we know that the way light is coupled to plasmons and the associated near-field distributions can be strongly dependent on the geometry and morphology of the grating structure. This, in turn, is expected to be influential in the final TMOKE response of the magnetoplasmonic crystal. To address this issue, we analyzed the relation between grating geometry and the reflectance and TMOKE responses. We note that, for technological applications, a high reflectance combined with large TMOKE signal is desirable. In consequence, conditions are optimal when both parameters are maximized.

To make a quantitative analysis of these optimal conditions, we define the relevant parameters as follows. For that purpose, we focus our discussion on the intensity collected in the specular reflection of light incoming at normal incidence on a grating with parameters (periodicity - $\Delta = 400$ nm, width - $w = 320$ nm, base - $b = 80$ nm, depth - $d = 120$ nm). The angle-resolved reflectance map is shown in figure 5.1 (a), which reveals a clear absorption pattern, originated by plasmonic resonances centered at the wavelength of $\lambda = 690$ nm (signaled by white lines). As expected, TMOKE is largely enhanced in the vicinity of the SPP resonance (figure 5.1 (b)). From these maps, we extracted reflectance and TMOKE profiles at different wavelengths, showing their dependence on the angle of incidence, see figures 5.1 (c) and (d), respectively. In these profiles, we define TMOKE$_{max}$ as the maximum TMOKE amplitude, having a corresponding reflectance given by (R(TMOKE$_{max}$) as indicated in figures 5.1 (c) and (d)).

FIGURE 5.1: Panel (a) and (b) shows the reflectance and
the TMOKE angular resolved plot, in which the light beams
arise from the specular reflection and diffraction modes, cor-
responding to a grating with periodicity of $\Delta = 400$ nm.
In (c) and (d), reflectance and TMOKE cross-sectional plots
were extracted from the horizontal dashed lines shown in
panel (a) and (b) respectively, corresponding to the wave-
lengths of $\lambda = 630, 690$ and 750 nm.

Following the aforementioned interest for applications, we have analyzed the optimal conditions that maximize the values of TMOKE$_{max}$ at R(TMOKE$_{max}$) in a large ensemble of gratings of different geometric parameters (see the appendix B). In order to follow this discussion, we define the filling fraction Γ of a grating as the ratio of the grating width (w) and the periodicity (Δ). i.e. $\Gamma = \frac{w}{\Delta}$ (see sketch 4.1 (a)). In our study we have analyzed the optical properties of a large collection of samples consisting in [Cr (4 nm) / Au (16 nm) / [Co (14 nm) / Au (16 nm)] x4 / Co (14 nm) / Au (7 nm)] multilayers, with filling fractions in the range $0.20 < \Gamma < 0.90$ and periodicities 400 nm $< \Delta <$ 1000 nm. We first draw the attention to figure 5.2 (a), which plots the values of R(TMOKE$_{max}$) and TMOKE$_{max}$ measured in the first-order diffracted modes $m = \pm 1$. Figure 5.2 (a), reveals a general rule, by which the TMOKE amplitude increases as the reflectance decreases; however, two groups of samples – grouped according to their value of the filling factor Γ- can be easily identified in relation to their steeper or gentler slope in the R(TMOKE$_{max}$)-TMOKE relation (5.2 (a)). More specifically, the gratings with $\Gamma > 0.70$ exhibit large values of TMOKE (up to above 3% highlighted in blue), but at the expense of a rather low reflectance. On the contrary, gratings with $\Gamma < 0.60$ display lower values of TMOKE, but can exhibit relatively large values of reflectance (highlighted in orange).

The data discussed in the previous paragraph correspond to the intensities collected in the first-order diffraction modes. One could wonder what happens in the specular reflection ($m = 0$), where the reflectance (and, therefore, R(TMOKE$_{max}$) should be larger than in the diffracted modes. To answer this question, we plot in figure 5.2 (b) the values of R(TMOKE$_{max}$) and TMOKE$_{max}$ collected in the specular reflection for a large ensemble of gratings with different periodicities and filling factors $\Gamma > 0.70$. An important observation is revealed by this figure, since it shows that relatively large TMOKE values –up to about 2.5% – can be obtained with reflectance of about 0.25, which is a significant improvement with respect to the case of diffracted orders, where, for similar values of TMOKE, the reflectance was about 0.05. We should stress that the intrinsic TMOKE values are usually in the range of 0.01%, so that, despite the apparent moderate TMOKE signals obtained in the specular reflection (i.e., in the order of a few %), the

FIGURE 5.2: In (a) and (b) the reflectance at the maximum TMOKE amplitude ($R(\text{TMOKE}_{max})$ is shown as function of maximum TMOKE amplitude (TMOKE_{max}) for the diffracted and specular reflected light respectively. A set of gratings having a different filling fraction Γ is highlighted in orange and blue, corresponding to a $0.20 < \Gamma < 0.60$ and $\Gamma > 0.70$, respectively.

present results signify a remarkable enhancement with respect to the intrinsic TMOKE signal of about two orders of magnitude with respect to the intrinsic TMOKE signal. All in all, the data plotted in figure 5.2 provides useful guidelines for the design of magnetoplasmonic crystals with optimized reflectance and TMOKE responses and indicates that a trade-off exists between both parameters, so an optimized solution should seek a compromise between them. Additionally, we can conclude that optimal solutions can be sought in the specular reflection at normal incidence, where a reasonable compromise can be found between reflectance and TMOKE.

Yet, an important aspect requires some explanation, regarding the distinctive trends observed for gratings in the first-order diffracted mode ($m = \pm 1$) of different filling factors $\Gamma > 0.20$ and $\Gamma < 0.60$, see figure 5.2 (a). In order to shed light on this issue, we performed FDTD calculations to obtain the near-field distributions for gratings of filling fractions $\Gamma = 0.80$ and $\Gamma = 0.20$, respectively, shown in figure 5.3. The simulations were performed by considering light at normal incidence, and the values of Γ were

chosen to be representative of the two distinct groups reported in figure 5.2. Panels (a) and (c) the spatial distributions of electromagnetic field densities, while panels (b) and (d) show the changes in intensity in the near-field distribution (ΔNF(M)) for distinct magnetization states $\Delta NF(M) = NF(M^+) - NF(M^-)$. The simulations reveal a big difference depending on the value of Γ. For $\Gamma = 0.80$, the field distribution is mostly located outside the cavity formed in the gratings (figure 5.3) (a), and the ΔNF(M) associated distribution is large and extended in space (figure 5.3) (b). On the contrary, for $\Gamma = 0.20$, there is a significant fraction of electromagnetic fields inside the cavity (figure 5.3) (c) while the ΔNF(M) distribution is much more confined and weaker (figure 5.3 (d)). The results of the FDTD simulations are

FIGURE 5.3: Image in the near-field obtained by FDTD simulation of a 1D photonic crystal illuminated at normal incidence with different filling fraction $\Gamma = 0.80$ ((a) and (b)) and $\Gamma = 0.20$ ((c) and (d)). Where the electric field and ΔNF(M) distribution are shown in panels (a) and (c), and (b) and (d), respectively.

in line with the experimental data displayed in figure 5.2, in which gratings with larger Γ exhibit larger ΔNF(M), i.e. magneto-optic response. The simulations for Γ = 0.20, seem to suggest that lower values of the filling fraction may, eventually, drive to localized plasmon resonances, which are less effective in yielding larger magneto-optic responses.

5.3 Excitation of SPPs with light at oblique incidence

In the previous section, we explored the TMOKE response enhanced by plasmons when light is incident along the normal to the surface. One important conclusion is that a compromise can be found to obtain optimized reflectance and TMOKE response in the specular reflection, where light reflected from the sample is collected along the same direction as the incident light. Here we explore an alternative geometry, by which we analyze the TMOKE response in diffracted modes when light is incident along an oblique direction. Using this geometry, we analyzed the properties of the same photonic crystal that was previously investigated in chapter 4.

- Periodicity - $\Delta = 1000$ nm

- Width - $w = 800$ nm

- Base - $b = 200$ nm

- Depth - $d = 120$ nm

The experiments were carried out with light incident at 30° from the normal. As usual, we measured the angle-resolve reflectance and TMOKE maps, which are shown, respectively, in figures 5.4 (a) and (b). Similarly, we plot in figures 5.4 (c) and (d) the reflectance and TMOKE profiles measured at different wavelengths as a function of the angle of incidence. The image of figure 5.4 (a) reveals an intense beam emerging at the emission angle -30°, which corresponds to the specular reflection of the off-normal angle excitation, together with the two diffraction modes corresponding to $m = +1, +2$. In the diffracted modes we can observe the cross-shaped reflectance dips characteristic of the SPP resonances. As expected, the TMOKE

response is clearly enhanced along the SPP dispersion lines (figure 5.4 (b)) with the characteristic derivate-like line-shape (see figure 5.4 (d)).

FIGURE 5.4: Panel (a) and (b) shows the reflectance and the TMOKE angular resolved plot from a off-angle excitation, in which the light beams arise from the specular reflection and diffraction modes, corresponding to a grating with periodicity of $\Delta = 1000$ nm. In (c) and (d), reflectance and TMOKE cross-sectional plots were extracted from the horizontal dashed lines shown in panel (a) and (b) respectively, corresponding to the wavelengths of $\lambda = 815$, 780 and 745 nm.

One particularly interesting observation is that the angle of emission of the $m = +1$ diffracted mode is nearly along the normal to the surface (figure 5.4). In these conditions, the TMOKE signal is enhanced to about 2% (figure 5.4 (d)), while the reflectance is in the order of $\approx 0.1 - 0.15$ (figure 5.4 (c)). In the previous section we have seen that, with light at normal incidence, the TMOKE in the specular reflection can reach values up to about 2.5% with a reflectance of about 0.25. Here we see that off-normal incident light at 30° gives TMOKE signals of about 2% in the first-order diffracted mode, with reflectance below 0.20. Comparing these results, we see that the reflectance and TMOKE are better optimized using light at normal incidence and collecting the specular light along the same direction.

5.4 Excitation of one-way propagation of SPPs.

Here we show that light coming at oblique incidence can be used to excite SPPs propagating along one way, i.e., to excite SPPs propagating only along either forward or backward directions. The principle of operation is sketched in figure 5.5. When light is coming at normal incidence, the coupling of diffracted modes to forward- and backward- propagating SPPs is symmetric. In contrast, at oblique incidence, this symmetry is broken, and the coupling of diffracted modes to SPPs depends on the direction of propagation. Eventually, at selected wavelengths, the propagation of SPPs is possible only along one direction.

We explored this possibility in the same grating discussed in the last section. In figures 5.6 (a) and (b) we show the angle-resolved reflectance maps measured in a spectral range centered on $\lambda = 835$ nm corresponding, respectively, to light incident at negative and positive oblique angles of incidence ($\pm 40^o$). The reflectance dips in the diffraction modes $m = -1, -2$ (panel (a)) and $m = 1, 2$ (panel (b)) reveal one-way SPPs, corresponding to forward- and backward-propagation, respectively.

We address now the study of the TMOKE signal associated with the excitation of the one-way plasmons. For that purpose, we present in 5.7 (a) and (b) the angle-resolved TMOKE maps for the forward and backward propagating SPPs, respectively, which, as expected, show how the signal increases

FIGURE 5.5: Schematics of a single plasmonic mode excitation. (a) Under particular conditions, the incident light beam can couple only to backward-propagating SPPs modes, as indicated by the green circles. The beam decomposition is shown in (b), indicating the corresponding diffraction modes ($m = -1, -2$ indicated by orange circles) and specular reflection ($m = 0$).

in the vicinity of the plasmon resonances. TMOKE profiles taken at $\lambda = 835$ nm show that the sign of the TMOKE amplitude is determined completely by the direction of the propagation of SPPs. More specifically, we see that the TMOKE amplitude is positive for forward propagating plasmons excited in the plasmonic mode $\eta = +3$, while the negative TMOKE amplitude is associated with backward propagating SPPs excited at the plasmonic mode $\eta = -3$ (see figure 5.7 (c)).

The data shown in figures 5.6 and 5.7 demonstrate that one-way SPPs and associated TMOKE signals can be achieved by illumination with light at off-normal angles of incidence. As mentioned in chapter 1, grating couplers are relevant devices that can link data transmitted via optical fibers to on-chip photonic circuits. The possibility to engineer the angle of incidence on the on-chip grating would enable to control the direction of the flux of optical signals in integrated circuits coupled to optical external links, opening interesting perspectives in integrated nanophotonics.

FIGURE 5.6: In (a) and (b) the reflectance is shown for a single mode excitation of forward- and backwards- propagating SPPs respectively. In (c) the cross-section at $\lambda 835$ nm presented for a single excitation is shown in red and blue.

FIGURE 5.7: In (a) and (b) the TMOKE amplitude is shown
for a single mode excitation of forward- and backwards-
propagating SPPs respectively. In (c) the cross-section at
$\lambda = 835$ nm presented for a single excitation is shown in
red and blue.

5.5 Numerical simulations of one-way plasmons.

Finally, we show that the one-way propagation of SPPs can be reproduced through numerical simulations applied to a magnetoplasmonic crystal that mimics the experimental one. As shown in figure 5.8, the reflectance map obtained by FDTD simulations presents the expected signatures of SPP excitations as cross-shaped reflectance dips. In this figure we indicate different resonant conditions labeled as (I)-(IV), corresponding to the wavelengths $\lambda = 685$ and 823 nm and angles of incidence $\theta = \pm 5.25^o$. Note that the excitation points (I),(IV) and

FIGURE 5.8: Reflectance map of a simulated photonic crystal, in which particular conditions of wavelength and angles of incidence are labeled (I)-(IV).

(II),(III) rest each over a single plasmonic excitation, corresponding to the modes $\eta = +2$ and $\eta = -2$, a forward- and backward- SPP propagating mode, respectively. In order to elucidate the relation between TMOKE amplitudes and unidirectional plasmons and compare with the experimental data (figure 5.7), we performed near-field simulations where the field distribution and the ΔNF(M) (defined in section 5.2) are shown in figure 5.9.

A first observation of figure 5.9 (a) and (b) (corresponding to $\lambda = 685$ nm) reveals an asymmetric distribution of electric fields in the near-field, which is especially evident inside the cavities of the grating. Note that in going from (I) to (II), the asymmetric distribution is inverted (figures 5.9 (a) and (b)), as expected from switching between forward- and backward-propagating SPPs ($+k_{SPP}$ and $-k_{SPP}$ with $\eta = +2, -2$, see 5.8). A similar trend is observed for the near-field distributions presented in figure 5.9 (c) and (d), corresponding to points (III) and (IV) of figure 5.8 (where $\lambda = 823$ nm). Now, if we look at the magnetic response of the near-field distributions quantified by ΔNF(M) (figure 5.9 (e)-(h)), we see that the spatial distribution

of ΔNF(M) also reverses sign when switching from forward- to backward-propagating SPPs. It is important to note that the near-field and ΔNF(M) distributions have their spacial distribution symmetry determined exclusively by the K_{SPP} direction, being independent of the angle of incidence, as discussed previously 5.4.

FIGURE 5.9: (a-d) A near-field distribution, and (e-h) the
magneto-optic behavior ΔNF(M) is shown for four distinct
plasmon excitation sites (I-IV depicted in figure 5.8).

Before closing this section, we have an important conclusion regarding the amplification capability of the TMOKE response by the diffracted modes. More specifically, we note that particularly large values of TMOKE (above ±4%) are found in the vicinity of plasmon orders $\eta = \pm1$, corresponding to the diffraction modes $m = \mp2$ (figure 5.7 (c)). These are among the highest TMOKE values we have measured in the ensemble of crystals studied in this Thesis. Curiously enough, the TMOKE signal measured in the diffracted mode $m = \pm1$ is smaller ($\approx \pm2.5\%$) than for $m = \pm2$ ($\approx \pm4.2\%$). One may wonder if the higher-order diffraction modes in combination with first-order plasmon resonances may pave the way to a more efficient way to enhance TMOKE.

5.6 Conclusions

We investigated the optimal conditions to achieve simultaneous maximization of reflectance and TMOKE responses. For that purpose, we compared the optical properties at normal and off-normal incidence of a large ensemble of magnetoplasmonic crystals of different periodicities and metallic filling ratios. From our extensive study we conclude that the best compromise is found by using light at normal incidence and collecting the specular light along the same direction.

We then moved to the study off-angle SPP excitations, to demonstrate that by selecting a proper angle of incidence, a one-way propagating surface plasmon can be excited. Thus, we could study selectively the influence of one-way plasmons on the TMOKE response. Interestingly. we found that particularly large TMOKE amplitudes -upwards of 4%- o 4.2% -for particular conditions of one-way plasmon excitation. FDTD simulations confirm these experimental results and corroborate that the sign of the TMOKE enhancement is exclusively dependent on SPP wavevector K_{SPP} and does not depends on the angle of incidence.

Chapter 6

Electrically Tunable Magnetoplasmonic Gratings

6.1 Abstract

Here we investigate the effects of magnetism and ferroelectricity on the propagation of surface plasmons polaritons (SPPs). With this in mind, we studied plasmonic crystals composed of ferromagnetic and ferroelectric materials. We analyzed the magnetoelectricplasmonic response using angle-resolved Fourier spectroscopy with in-situ applied magnetic and electric fields. Remarkably, our experiments reveal that the plasmonic band structure can be modulated by electric fields. As a result, the amplitude of the transverse magneto-optic Kerr effect (TMOKE) can be modulated and even its sign can be reversed by appropriate electric field protocols. The present results pave a new path to the control of the TMOKE response through a proper control of the dielectric environment.

6.1.1 Chapter highlights

- An advanced lithography design was developed and implemented, which allowed the optical access and in-situ ferroelectric and magnetic measurements.

- We analyzed the effects of the ferroelectric polarization on the transversal magneto-optic Kerr effect (TMOKE) response: under certain conditions, the sign of TMOKE can be reversed by electric fields.

6.2   Introduction

As discussed in chapters 3, 4, the incorporation of ferromagnetic layers in magnetoplasmonic crystals adds the possibility of nonreciprocal effects induced by the breaking of time-reversal symmetry [18, 19]. Along similar lines, an extra functionality can be implemented through the replacement of a conventional centrosymmetric dielectric layer by a noncentrosymmetric material, which breaks space inversion symmetry [114, 115]. We have followed this approach by inserting a ferroelectric material, $BaTiO_3$ in our case, which is known to have large ferroelectric polarization [116, 117, 118] and particularly large electro-optic coefficients [5, 64, 65]. The latter property is especially relevant for the propagation of plasmons [119, 120], as it enables the modulation of the refractive index by electric fields [121, 122]. The exploration of the electric modulatation of the optical properties of magnetoplasmonic crystals required the development of advanced lithography to enable the in-situ characterization of ferroelectric and magnetic effects on the propagation of SPPs. For that purpose, we used angular-resolved reflectance spectroscopy to study the effects of applied magnetic and electric fields on the transverse magneto-optic Kerr effect (TMOKE) response.

6.3 Experiment description

6.3.1 Sample preparation

A multiferroic heterostructure being composed of ferroelectric and ferromagnetic materials in association with a plasmonic material was constructed and tailored as a base of a magneto- and electric-active photonic crystal. The ferroelectric structure is composed of a BaTiO$_3$ (BTO) layer, having thickness of 200 nm, being grown over a bottom electrode of composition La$_{0.7}$Sr$_{0.3}$MnO$_3$ (10 nm), a well-known collosal magnetoresistance perovskite with metallic behaviour [123, 124]. The whole heterostructure was deposited on top of a SrTiO$_3$ (STO) commercial substrate.

After the whole multilayered structures is grown, a complex lithography process -described in chapter 2.1.4- is developed, in which the electric- and magnetic-active contacts are coated with a magnetoplasmonic multilayer, being composed of Cr (4 nm) / Au (16 nm) / [Co (14 nm) / Au (16 nm)] x4 / Co (14 nm) / Au (7 nm).

FIGURE 6.1: Schematics of the magneto-electric-plasmonic crystal: In (a) a structure side view displays the grating parameters where Δ, w, and d are the periodicity, width and depth of the grating. Panel (b) reveals the top view of the grating structure, where l, h_c and l_c are the grating length, contact width and length, respectively.

As mentioned in section 2.1.4, the electric-active contact cannot exceed a certain area (typically < 10000 μm^2), otherwise the polarization of BTO may not be switched due to the presence of leakage currents. To minimize the effects of leakage currents and increase the capability of switching the ferroelectric polarization, a thick passivation (Al$_2$O$_3$ in our case) was deposited all over the surface, including the active layers where electric fields must be

applied. Because of this latter observation, a lithographic step is required to open a window exclusively over the active layers, which is done through a chemical etching process of the passivation layer (see section 2.1.3), enabling electric contact over that area. Finally, the photonic structure is patterned through e-beam lithography followed by an ion-milling resulting in the structure shown schematically in figure 6.1.

The full side view of the magneto-electric-plasmonic crystal can be seen in figure 6.2, where the long-range contacts are seen over the passivation layer, thus providing electric to the system. For the presented study, we focused on crystals with parameters, $\Delta = 740$ nm, $w = 560$ nm, $d = 175$ nm, $l = 40$ μm, $h_c = 80$ μm and $l_c = 130$ μm, corresponding to the grating periodicity, width, depth and wire length, while h_c and l_c are the active contact width and length, respectively (real device images are presented in appendix C).

FIGURE 6.2: A cross-sectional view or the magnetoelectric-plasmonic crystal, revealing the full layered structure. The difference in (a) and (b) is the different material used for passivation, i.e., photoresist and Al$_2$O$_3$, were used, respectively.

6.3.2 Control of the magnetization and ferroelectric polarization

Before we advance into the experimental results, we establish the allowed set of configurations between magnetization and polarization (figure 6.3). More specifically, the magnetization is confined along the grating longer

axis, which corresponds to the $\hat{y}$ direction, and assumes positive and negative values $M^+\hat{y}$ and $M^-\hat{y}$. On the other hand, figure 6.3 (b) shows the microscopic ferroelectric domain configuration for the electric saturated states, where the polarization is fully aligned along the $\hat{z}$ direction, taking positive and negative values $P_+\hat{z}$ and $P_-\hat{z}$. Figures 6.3 (c)-(d) show schematically different possible domain configurations of a ferroelectric depolarized state P_{depol}. The case depicted in figure 6.3 (c) corresponds to ferroelectric domains pointing along in-plane and out-of-plane directions. Alternatively, the ferroelectric depolarized state can be caused by domains aligned in an anti-parallel fashion along the out-of plane direction. For instance, the cases depicted in figures 6.3 (d) and (e) correspond to different domain sizes, giving way to different domain wall densities. The different domain configurations displayed in figures 6.3 (c)-(e) will be discussed later in this chapter in relation to the origin of the modulation of TMOKE by electric fields.

FIGURE 6.3: Schematics of the orientation of ferromagnetic and ferroelectric domains. (a) The magnetization is confined along the grating major axis, defined as $\hat{y}$ direction. (b) The two possible ferroelectric saturated polarization states, lying along the $\hat{z}$ axis. (c) Ferroelectric domain configuration in the depolarized state, in which some domains lay in the $\hat{x}\hat{y}$ plane, while other are oriented along the out-of-plan $\hat{z}$ direction. (d) and (e) show a ferroelectric depolarized state with domains along the out-of-plan $\hat{z}$ direction, having different domain size and domain wall density.

During the next sections we will explore the influence of magnetism and ferroelectricity on the propagation of plasmons and the transverse magneto-optic Kerr effect (TMOKE). Our study was done under the saturated magnetic (M^+, M^-) and ferroelectric (P_+, P_-) states, as well in ferroelectrically depolarized states (P_{depol}), where the ferroelectric domain polarizations are oriented along different directions, giving a macroscopic state with null polarization.

6.3.3   Electric modulation of SPP propagation and TMOKE

The analysis of the SPP propagation and TMOKE responses was done in three different states of the macroscopic ferroelectric polarization. In the depolarized state (P_{depol}), the ferroelectric layer is depolarized through an ac electric field "degaussing" protocol [72], where a polarizing voltage is initially applied and decreased continuously to zero by reversing the sign of the applied field between successive states. Alternatively, measurements were also done by saturating the ferroelectric polarization along two opposite directions along the out-of-plane orientation (*c*-axis). The TMOKE amplitude, measured from angular-resolved reflectance and TMOKE maps, was obtained through the expression: $\tau = \frac{I(H^+) - I(H^-)}{I(H^{avg})}$ for the three polarization states $\tau(P_{depol})$, $\tau(P_+)$ and $\tau(P_-)$.

Figure 6.4 (a) shows the full reflectance map measured with a saturated positive magnetization and depolarized state $R(P_{depol})$. This figure includes the dispersion lines of SPPs calculated from the expression (3.3), where ε_{BTO} and ε_m are the dielectric function of the bulk BTO and the magnetoplasmonic multilayer, used to calculate k_{SPP}, defined as: $k_{SPP} = k_x \sqrt{\frac{\varepsilon_{BTO}\varepsilon_m}{\varepsilon_{BTO}+\varepsilon_m}}$. A clear evidence of the effects of the ferroelectric state can be observed in the reflectance profiles measured for the polarization states $R(P_{depol})$, $R(P_+)$ and $R(P_-)$, as shown in figure 6.4 (b)-(d). These reflectance profiles are extracted from the wavelengths corresponding to $\lambda = 620$, 710 and 780 nm highlighted by the horizontal white dashed lines in 6.4 (a). In particular, the most abrupt changes in reflectance happen near the wavelengths of $\lambda = 710$ nm, as displayed in figure 6.4 (c), where the polarization state changes substantially the angular distribution of the light emitted from the crystal.

FIGURE 6.4: (a) Reflectance map for a depolarized state $R(P_{depol})$. Propagating SPP dispersion lines are shown in white. The panels (b)-(d) show the cross-sectional reflectance plots, that were extracted from the horizontal dashed lines in (a), corresponding to the wavelengths of $\lambda = 780, 710, 620$ nm. The different curves in each panel display the profiles that correspond to the different ferroelectric polarization states, i.e. $R(P_{depol})$, $R(P_+)$ and $R(P_-)$ displayed in red, green and blue, respectively.

An important observation can be drawn from inspection of the data shown in figure 6.4. More specifically, we observe that the angular distributions of the emitted light for the two saturated polarized states $R(P_+)$ and $R(P_-)$, are very similar. This simple observation implies that linear

electro-optic effects are not the main mechanism for the observed changes in the reflectance patterns, as they would imply different optical properties for the two ferrolectric saturated states. On the contrary, photoelastic effects, in which changes of refractive indices are driven by mechanical stress [53, 54], could account for the fact that, while reflectance patterns are very similar for the saturated states, a clear change is observed with respect to the depolarized states. This argument is developed later in this chapter. Finally, we note that the modulation with electric fields is strongly dependent on the wavelength; the angular distribution is modified largely at the wavelength $\lambda = 710$ nm, but the changes are minor for the other wavelengths shown in 6.4 ($\lambda = 620, 780$ nm).

The polarization states not only affect the reflectance pattern as aforementioned, they also modify the TMOKE response. To visualize this magnetoelectric effect we calculate the TMOKE signal for each polarization individually. For instance, the TMOKE response for a fixed polarization P_+ state is given by:

$$\tau(P_+) = \frac{I(H^+, P_+) - I(H^-, P_+)}{I(H^{avg}, P_+)}, \tag{6.1}$$

where, H^+ and H^-, are the magnetic field states, being set in positive and negative magnetic saturation, while H^{avg} is collected from an averaged ac magnetic field. A similar expression can be defined for the P_{depol} and P_- states.

The angle-resolved TMOKE maps shown in figure 6.5 correspond to the P_{depol} and P_+ states. Similarly to reflectance maps, the TMOKE maps for the P_+ and P_- states are very similar and, thus, figure 6.5 (b) only shows the map corresponding to P_+. In the maps of figure 6.5 we have also plotted the SPPs band diagram for backward- and forward- propagating SPPs, calculated from equation 3.3, shown in section 3.3.3. At first sight, it may seem from figure 6.5 (a) that TMOKE is roughly the same for positive and negative angles of incidence. However, a more detailed inspection reveals that under certain conditions there are important asymmetries in the TMOKE response. This is the case, e.g., of the narrow spectral region around the

wavelength $\lambda = 700$ nm, where a noticeable suppression of the TMOKE effect is observed for the forward propagating SPP resonance orders $\eta_{SPP} = -3, +2$. This observation reveals an asymmetric optical response induced by changes in the ferroelectric polarized state, whereby the plasmonic dispersion lines are clearly asymmetric with respect to the angle of emission near the wavelength $\lambda = 700$ nm. It is remarkable to note that the angle-resolved TMOKE maps measured in the $\tau(P_+)$ state -shown in figure 6.5 (b), also shows a suppression in the TMOKE response at the same wavelength, but now the suppressed TMOKE signal is observed for the backward- instead of the forward- propagating SPP mode $\eta_{SPP} = -2, +3$. As discussed in the following, the fact that different polarization states affect differently the forward- and backward- SPP modes gives way to the unprecedented observation of a reversal of the TMOKE sign with electric fields.

FIGURE 6.5: In (a) and (b) the spectral dependence of TMOKE amplitude is shown for two distinct polarization state P_{depol} and P_+. The dispersion lines of forward- and backward- propagating SPPs are indicated in the figures.

To go deeper into this question, we analyzed TMOKE profiles measured as a function of the angle of emission at different wavelengths within a narrow range around $\lambda = 700$ nm. The profiles, shown in figure 6.6, demonstrate that the asymmetric optical response induced by the electric fields is strongly dependent on wavelength. The most remarkable observation is

the reversal of the TMOKE sign as a function of the ferroelectric polariza-
tion state, particularly noticeable at wavelength $\lambda = 710$ nm (figure 6.6 (b)),
where the TMOKE sign is inverted from the polarized to the depolarized
state. This is a remarkable result, as it enables reversing the sign of the
amplitude of a relatively large TMOKE signal with electric fields ($\pm 0.4\%$ at
$\lambda = 710$ nm). We stress that this electric field-modulation is particularly rel-
evant, as the intrinsic TMOKE response of magnetic materials is well below
the modulated value, see discussion in section 4.3.3).

FIGURE 6.6: Panels (a)-(c) display cross-sectional TMOKE
plots extracted from the previous figure, corresponding to
the wavelengths $\lambda = 700, 710, 720$ nm, where a narrow an-
gular region is shown. Panels (d)-(f) reveal a much broader
angular spectrum corresponding to the wavelengths $\lambda =$
620, 760, 800 nm.

In addition to the sign reversal, the profiles shown in figures 6.6 (a)-(c)
show also changes within the angular distribution of the emitted light as
a function of the polarization state. To illustrate this point, in panel (a) a
blue vertical dashed line is drawn at the maximum of TMOKE for the $P_\pm$
polarized states, which is clearly shifted from the angular position of the
TMOKE peak for the P_{depol} state. The angular displacement of the TMOKE

peak signals between the different polarized states is also noticed in the following panels (b) and (c), in which the gray and black dashed lines illustrate the evolution of the maximum TMOKE amplitude as a function of the wavelength. As shown in figures 6.6 (d)-(f), the asymmetric TMOKE response with respect to the angle of emission for different ferroelectric polarization states is also noticeable for other spectral regions, for which particular conditions of wavelength and angle of emission can be found in which the sign of the TMOKE response can be reversed. Consequently, conditions can be found in which a strong magnetoelectric effect can be observed in the magneto-optic response for a wide range of wavelengths. This is an important observation, which extends the concept of magnetoelectric coupling in multiferroic structures [125], which seek to reverse the magnetization with electric fields, to the field of optics. Thus, instead of magnetization, our experiments reveal that magneto-optic signals can be reversed by using electric fields.

Finally, we explored the reproducibility of the polarization effects on the TMOKE response by measuring the signal between the states (P_{depol}, P_+, P_-) over two complete cycles measured at three different wavelengths $\lambda = 700, 710, 720$ nm. The data, which is plotted in figure 6.7, shows that the changes of the TMOKE response -including the sign reversal- can be reversibly switched for the polarization states P_{depol}, P_+ and P_-. Figure 6.7 shows the reproducibility of the polarization effects on the TMOKE response, where the polarization was cycled between the three states (P_{depol}, P_- and P_+) over two cycles.

Summing up, our results show that the optical response of magneto-electric-plasmonic crystals can be modulated by electric fields. In the experiments, the positive and negative ferroelectric saturated states have very similar optical properties, whereas the macroscopically depolarized state exhibits distinctive properties. In particular, we observe that, under proper conditions, the sign of TMOKE can be reversed between saturated states and depolarized states. The fact that the saturated states of opposite signs show similar optical responses indicates that linear Pockels electro-optic effects, although present, are not most relevant factor to the observed phenomenon. Alternatively, changes of the refractive index associated to changes in the

FIGURE 6.7: Panels (a)-(c) shows the TMOKE response as a function of the polarization state P_{depol}, P_- and P_+, corresponding to the wavelengths $\lambda = 700, 710, 720$ nm.

mechanical stress of the ferroelectric layer are compatible with the observations, since the opposite states of saturation would give the same changes of permittivity, while the depolarized could have a different value, provided that it sustains a different mechanical stress state. As described in section [53], the related phenomenon is usually referred as photoelastic effect and, unlike the electro-optic effect, is a universal property of materials, as photoelasticity is observed for all symmetry classes [53]. In this picture, in the saturated states the ferroelectric domains are fully aligned along opposite out-of-plane *c*-axes directions which would give identical strain states. In contrast, we surmise that in the depolarized case the BTO thin film may sustain substantial changes in the domain configuration that would yield a strain state different from the saturated states. This discussion is further elaborated in section 6.5.

6.4 Plasmon resonances under electro and magnetic fields

In section 3.4.2, we discussed the effects of magnetic fields on plasmonic resonances. Here we describe how the different polarization states of the ferroelectric can modulate the plasmon resonances, via changes of the refractive index by photo-elastic effects. Thus, globally, the plasmon resonances are affected by the magnetically shifted plasmon frequencies, (ω_+ and ω_-), together with the changes associated to the ferroelectric domain structure (changes in $n(P_{depol})$ and $n(P_\pm)$).

The superposition of these two effects is shown in figure 6.8. Panel (a) shows the effects of the magnetic field, by which the resonance frequency ω_+ and ω_- (shown in blue and red) are slightly displaced depending on the orientation of the magnetization M^+ and M^-. This effect induces a displacement in the frequency space as shown in panel (b), where the modes ω_+ and ω_- -visible as dips in the intensity of the reflected intensity- are shown in blue and red, respectively. The subtraction of the reflected intensities associated with the two magnetized states results in the derivative-like curve shown by the green dashed line, which is associated with the TMOKE response, as previously discussed in chapter 3.

Now we discuss the effects of the different ferroelectric domain configurations on top of the magnetic field-induced effects discussed in the previous paragraph. This can be seen in figure 6.8 (c), which shows magnetic field-induced shifts of the plasmon resonances for the depolarized P_{depol} and polarized $P_\pm$ ferroelectric states. We discuss the particular case of the intersection between two plasmonic modes of different orders i.e. $\eta = -2, +3$ and $\eta = 2, -3$, where the effects of the electric field-induced changes in the refractive index are shown schematically, resulting in additional shifts in the plasmon dispersion lines labelled as $\omega_\pm^{P_{depol}}$ and $\omega_\pm^{P_\pm}$. The effects on the intensity of reflected light are depicted in figure 6.8 (d) where the plasmonic modes $\omega_+^{P_\pm}$, $\omega_+^{P_{depol}}$ and $\omega_-^{P_\pm}$, $\omega_-^{P_{depol}}$ are displaced unevenly in frequency. Finally, the subtraction of the curves in panel (d) result in the derivative-like lineshapes for the polarization state $P_\pm$ and P_{depol} shown in 6.8 (e), representing the TMOKE signal for these different ferroelectric polarization states.

Interestingly, these results reproduce in a qualitative way the differential lineshape observed in the experimental results shown in figure 6.6 (b).

FIGURE 6.8: Frequency-splitting ω, induced by the magnetization state $M^\pm$, resulting in the displacement in the SPP band diagram show in (b), from which the plasmonic differential lineshape can be obtained. In a similar way the frequency-splitting is shown for two distinct ferroelectric states P_{depol} and $P_\pm$ in (c) respectively, resulting in two sets of SPP diagrams $\omega_\pm^{P_{depol}}$ and $\omega_\pm^{P_\pm}$ shown in (d). Finally the differential lineshape is obtained for P_{depol} and $P_\pm$ (e).

6.5 Relevance of the protocol for electric field cycling

In the following, we discuss in more detail how the different (magneto-) optical states can be accessed electrically. For that purpose, we considered two different ways to modulate the applied electric fields. In a first approach (I), we cycled the field without changing the direction of rotation around the hysteretic loop, as in a conventional ferroelectric loop (figure 6.9 (a)). The second method (II) consisted in changing the field discontinuously by reversing its sign between consecutive steps (figure 6.9 (b)). This approach

reproduces the ac-depolarization protocol that we have described in the beginning of this chapter. In both approaches, we measure the normalized changes in intensity amplitude around the wavelength $\lambda = 715$ nm and angle of emission $\phi = 10^o$. Figures 6.9 (c) and (d) also include a ferroelectric polarization electric field measured at the frequency 1 KHz using the top-bottom configuration. The P-E loops were measured by applying an electric bias on the active top-electrode and grounding with the LSMO bottom-electrode following the protocols described in the Ref, [126]. We define the normalized changes in intensity ($\chi(V)$) amplitude as:

$$\chi(V) = \frac{I(P_V) - I(P_{avg})}{I(P_{avg})}, \tag{6.2}$$

where $\chi(V)$, $I(P_V)$ and $I(P_{avg})$ are the normalized changes in intensity, the intensity at a given applied voltage V, and the average intensity, respectively. The results of these measurements are shown in figures 6.9 (c) and (d), which display the $\chi(V)$ amplitude $\chi(V)$ as a function of the applied voltage for the I and II loops. As seen in these figures, the two methods resulted in strikingly different optical responses. More specifically, cycling the electric field using the approach (I) did not result in any appreciable change of the $\chi(V)$ amplitude (figure 6.9 (c)). In contrast, by using the method (II), the $\chi(V)$ amplitude an sign could be change in a substantial way (figure 6.9 (d)). This is a very relevant result, since it indicates the protocol required to access the ferroelectric polarization states that enable a modulation of photonic properties of the crystal. At the same time, these experiments indicate that the microscopic domain configuration that can be reached in the depolarized state using method (II) is different from the domain configuration that can be reached in depolarized state (coercive field) using the approach (I).

We consider two possibles scenarios for the microscopic ferroelectric domain configuration of the depolarized state that are compatible with the experimental results displayed in figures 6.4-6.7. One central point in the following discussion is the fact that the depolarized state reached by cycling the electric field using the protocol (I) is different from the depolarized state attained following the ac-depolarizing protocol (II). More specifically,

we surmise that the depolarized state achieved after following the approach (II) is composed of anti-parallel domains oriented along the out-of-plane direction, with a relatively large domain size and relatively low density of domain walls (the case depicted in figure 6.3 (d)). On the contrary, two scenarios can be proposed for the depolarized state attained following the degaussing protocol (II). One possibility is that this state is composed of domains pointing along in-plane and out-plane directions (figure 6.3 (c)). In this case, the strain state would be different from that of the two opposite saturated states, which would give way to two different refractive indices by the photo-elastic effect. Alternatively, one can envisage that all domains are pointing along the out-of-plane direction, but with relatively small domain size and larger density of domain walls (figure 6.3 (e)). The significantly large density of domain walls -which are under a different strain state than the domain itself- would explain the different refractive index with respect to the two saturated states. Further experiments -e.g., X-ray diffraction under electric fields or piezoresponsive atomic force microscopy- are required to fully discriminate between these two possibilities.

It's noteworthy to stress that the large modulation of normalized changes in intensity ($\chi(V)$) by electric fields using the discontinuous method (II) (figure 6.9 (d)) can be indeed observed in the images taken in the Fourier plane, as shown in figure 6.10. Panels (a) and (b) show the $\chi(V)$ response collected in the Fourier plane for two data points -represented in blue and red, respectively in figure 6.9 (d)- that give $\chi(V)$ amplitudes with opposite signs. Additionally, figure 6.10 (c) shows the Fourier image corresponding to a point in the saturated ferroelectric state (shown in green figure 6.9 (d)). While in the former (panels (a) and (b)) the $\chi(V)$ the enhancement is clearly visible following the plasmon dispersion lines, in the latter (panel (c)) they are barely visible.

To end up, we note that the computation of the normalized changes in intensity $\chi(V)$ induced by the ferroelectric polarization is done through the average of the intensity collected by the pixels located over a plasmonic mode, indicated by the orange square in figure 6.10 (a) and (c). Therefore,

FIGURE 6.9: In (a) and (b) the applied voltage path is shown as a function of time, while in (c) and (d) reveals the optical response depending on polarization induced state during the different electric field path. In superposition a BTO ferroelectric hysteresis loop obtained at 1 kHz are shown in panels (c) and (d) in orange. The shadowed areas are described later in figure 6.10

the average intensity extracted from each frame at a different electric potential is extracted and shown in figure 6.9 (b) and (d). Besides that, figure 6.9 (a) and (b) show a small optical response exclusively near the SPPs resonances, where the measured response is inverted between two states with different ferroelectric domain configuration (corresponding to red and blue data points in figure 6.9 (d)). As discussed above, such inversion of the sign of the normalized changes in intensity ($\chi(V)$) amplitude may be induced by the different ferroelectric domain patterns, tailored during the ac-depolarization process described above.

6.6 Conclusions

We have shown that the photonic band structure and TMOKE response can be modulated in multiferroic photonic crystals. Remarkably, the sign of

FIGURE 6.10: Panels (a)-(c) display the normalized average intensity $\chi(V)$ in the Fourier plane for three polarization states, corresponding, respectively, to the data highlighted in blue, red and green in figure 6.9 (d). Plasmon resonances are visible in (a) and (b), corresponding to the states blue and red (6.9 (d)). Panel (a) shows an orange square, from which the normalized average intensity presented in figure 6.9 is calculated. Panel (d) shows a zoom of the region where the plasmonic effects are extracted.

TMOKE can be even reversed by electric fields under specific conditions of wavelength and angles of incidence and appropriate protocols for electric field cycling. From the analysis of these results, we conclude that photo-elastic effects are primarily at the origin of the modulation of TMOKE by electric fields. These results demonstrate that a proper engineering of magnetoelectroplasmonic crystals can lead to the development of devices with advanced functionalities, which can be used to control and tune light-matter interaction through different external stimulus.

Chapter 7

Outlook and Perspectives

In this work we have analysed extensively the interplay between surface plasmons, diffraction and magneto-optic effects, showing that the phenomenon of diffraction can be exploited to achieve a big enhancement of the TMOKE response. In particular, we have shown that under appropriate conditions of wavelength and angles of incidence, off-normal incident light can couple efficiently to one-way plasmons. Under such conditions, remarkably large values of TMOKE have been measured enhanced by these unidirectional plasmons, indeed the largest values that we have found in our extensive study of magnetoplasmonic crystals. Our observation that the TMOKE signal measured in the diffracted mode $m = 1$ is smaller (2.5%) than for $m = 2$ (4.2%) –see section 5.4- may imply that higher-order diffraction modes in combination with first-order plasmon resonances may pave the way to a more efficient way to enhance TMOKE. Consequently, this observation provides with interesting guidelines to engineer magnetoplasmonic crystals to test this possibility.

A further interesting perspective is to investigate the coupling of off-normal incident light to one-way plasmons in ferroelectric magnetoplasmonic systems, where the electric field could modulate the resonance conditions in such a way that the excitation of one-way plasmons is turned on- and off at will. This can have an interest in some prospective applications, as it would enable a control of unidirectional flow of plasmons in integrated photonic circuits.

Finally, our investigation in ferroelectric magnetoplasmonic crystals has shown that, for the particular analyzed systems, photoelastic effects can be

exploited to modulate the TMOKE response. In this case, it seems that electro-optic effects are not playing main role in the electric field modulation of the magnetoplasmonic response. Nevertheless, for other device architectures, electro-optic effects may do have a role. One particularly interesting situation is given in magnetoplasmonic crystals grown on BTO single crystals with in-plane ferroelectric domains. In this situation, the excited plasmons would travel perpendicular to the direction of the ferroelectric polarization, which is known to yield the largest Pockels effect in BTO ([5, 64, 127]). If successful, this strategy would enhance the functionality of the magnetoplasmonic crystals.

Chapter 8

List of publications and communications

List of scientific publications derived during this Thesis:

- R. Cichelero, M.V. Kataja, M.C. Quiles and G. Herranz. Non-Reciprocal Diffraction in Magnetoplasmonic Gratings., accepted OpEx 2018 (in production).

- R. Cichelero, M.A. Oskuei, M.V. Kataja, S.M Hadimi and G. Herranz. Angle-resolved spectroscopic maps of commercial disk-based magnetoplasmonic crystals., accepted JMMM 2018 (in production).

- L. Vistoli,W. Wang,Q. Zhu,B. Casals, R. Cichelero, A. Barthelemy, S. Fusil, G. Herranz, S. Valencia, R. Abrudan, E. Weschke, K. Nakazawa, H. Kohno, J. Santamaria, W. Wu, V. Garcia and M. Bibes. Giant topological Hall effect in correlated oxide thin films, Nature Physics 5 2018.

- H.B. Vasili, B. Casals, R. Cichelero, F. Macia. J. Geshev, P. Gargiani, M. Valvidares, J. Herrero-Martin, E. Pellegrin, J. Fontcuberta and G. Herranz, Direct observation of multivalent states and $4\,f \rightarrow 3\,d$ charge transfer in Ce-doped yttrium iron garnet thin films, Physical Review B 96 2017.

- B. Casals, R. Cichelero, P.G. Fernandez. J. Junquera, D. Pesquera, M. Campoy, I.C. Infante, F. Sanchez, J. Fontcuberta and G. Herranz, Giant Optical Polarization Rotation Induced by Spin-Orbit Coupling in Polarons, Physical Review Letters 117 2016.

- B. Casals, M. Espinola, R. Cichelero, S. Geprags, M. Opel, R. Gross and G. Herranz, Untangling the contributions of cerium and iron to the magnetism of Ce-doped yttrium iron garnet, Applied Physics Letters 108 2016.

List of conferences attended during this Thesis:

- Oral Presentation at Metamaterials 2018 Conference, Espoo - Finland, August 26, 2018

- Poster Presentation at Joint European Magnetic Symposia (JEMS2018) Conference, Mainz - Germani, September 3, 2018

- Poster Presentation at TO-BE cost action, spring meeting, Sant Feliu de Guixols - Spain, March 12, 2018

- Poster Presentation at International School of Oxide Electronics (ISOE2017), Corsica - France, April 11, 2017

- Poster Presentation at Spanish Nanophotonics Conference (CEN2016), Valencia - Spain, June 22, 2016

- Poster Presentation at International conference on Magnetism (ICM2015), Barcelona - Spain, July 5, 2015

Appendix A

Lithography protocols

The following appendix contains a detailed lithography protocol, containing the basic list of tasks that have been done to achieve the final samples. In the first two sections the methods for optical and electron beam lithography are described, followed by the evaporation protocol. The fourth section contains the full protocol done to achieve the samples built to achieve the present work.

A.1   Optical lithography protocols

Two different approaches were used in optical lithography, consisting in one layer or two layer of photo-resist (PR), the second method has the advantage of making the liftoff easier due the existence of an undercut induced during the preparation. The photo-resists used during the current study are Shipley S1813 and MicroChem SF5.

A.1.1   Standard optical lithography steps (SopL)

- Cleaning the substrate with acetone and isopropanol.

- Evaporation of fluids (substrates may retains water) at 150 °C

- Spin coating of S1813, with 4500 rpm and acceleration of 1.0s

- Soft baking at 95 °C.

- Optical exposition with 405nm laser light (i-line) with fluency superior to 150 mJ/cm^2

- Development time ranging from 30 to 60 seconds.

- Deposition

- Liftoff of S1813 with acetone

  If the sample is going to be exposed to any etching process, a hard baking is recommended before the film deposition:

- Hard baking at 110 °C, for 3 - 10 minutes.

A.1.2 Double layer photo-resist lithography steps (DLopL)

- Cleaning the substrate with acetone and isopropanol.

- Evaporation of fluids (substrates may retains water) at 150 °C

- Spin coating of SF5, with 4500 rpm and acceleration of 1.0s

- Soft baking the SF5 at 150 °C.

- Spin coating of S1813, with 4500 rpm and acceleration of 1.0s

- Soft baking at 95 °C.

- Optical exposition with 405nm laser light (i-line) with fluency superior to 150 mJ/cm^2

- Development for 30 seconds.

- Hard baking the S1813 at 110 °C.

- Development for 60 seconds.

- Deposition

- Liftoff of S1813 with acetone

- Liftoff of SF5 with EBR-PG

  If the sample is going to be exposed to any etching process, a hard baking is recommended before the film deposition:

- Hard baking at 110 °C, for 3 - 10 minutes.

A.2   Electron beam lithography protocols

Similarly to the optical case the e-beam lithography, two different recipes can be used depending on the purpose of the lithography, for hard nano-patterning a double layer e-beam resist is recommended, meanwhile for simple patterning development a single layer can do the job.

A.2.1   Standard electron beam lithography steps (SebL)

- Cleaning the substrate with acetone and isopropanol.

- Evaporation of fluids (substrates may retains water) at 150 °C

- Spin coating of PMMA, with 4500 rpm and acceleration of 1.0s

- Soft baking at 150 °C.

- E-beam exposition, dosis ranging from $120 - 200 \mu C/cm^2$

- Development in MIBK (1:3), with development time ranging from 30 to 60 seconds.

- Deposition

- Liftoff of PMMA with acetone

  If the sample is going to be exposed to any etching process, a hard baking is recommended before the film deposition:

- Hard baking at 150 °C, for 3 - 10 minutes.

A.2.2   Double layer electron beam lithography steps (DLebL)

- Cleaning the substrate with acetone and isopropanol.

- Evaporation of fluids (substrates may retains water) at 150 °C

- Spin coating of 495 PMMA A6, with 4500 rpm and acceleration of 1.0s

- Soft baking at 150 °C.

- Spin coating of 950 PMMA A7, with 3000 rpm and acceleration of 1.0s

- Soft baking at 160 °C.

- E-beam exposition, dosis ranging from $120 - 200\mu C/cm^2$

- Development in MIBK (1:3), with development time ranging from 30 to 60 seconds.

- Deposition

- Liftoff of PMMA with acetone

  If the sample is going to be exposed to any etching process, a hard baking is recommended before the film deposition:

- Hard baking at 150 °C, for 3 - 10 minutes.

A.3   Electron beam evaporation

The protocol for the electron beam evaporation is given below:

- N_2 Cleaning.

- Vacuum below 10^{-6} mbar.

- Evaporation at rates below 0.1 nm/s.

Appendix B

Ensemble of gratings and its magneto-optic response

As mentioned in chapter 5, the construction of figure 5.2 is based in the following tables of values, where the grating periodicity, metallic fraction, TMOKE$_{MAX}$ and R(TMOKE$_{MAX}$) are listed. Tables B.1 and B.2 corresponds to the TMOKE response emitted from the diffracted modes ($m = \pm 1$), while B.3 is based on the specular reflection ($m = 0$) TMOKE response.

B.1   Metallic fraction $\Gamma > 0.7$, diffracted modes $m = \pm 1$

Periodicity Δ(nm)	Metallic fraction Γ	TMOKE$_{MAX}$	R(TMOKE$_{MAX}$)
1000	0.92	0.021	0.043
1000	0.88	0.022	0.07
1000	0.84	0.024	0.06
1000	0.80	0.028	0.04
800	0.90	0.021	0.115
800	0.85	0.027	0.05
800	0.80	0.033	0.04
800	0.75	0.015	0.14
740	0.89	0.021	0.11
740	0.84	0.025	0.06
740	0.79	0.024	0.08
740	0.74	0.009	0.22
740	0.69	0.005	0.26
600	0.87	0.015	0.20
600	0.80	0.021	0.03
600	0.73	0.018	0.11
600	0.66	0.008	0.32

B.2 Metallic fraction $\Gamma < 0.6$, diffracted modes $m = \pm 1$

Periodicity Δ(nm)	Metallic fraction Γ	TMOKE$_{MAX}$	R(TMOKE$_{MAX}$)
1000	0.50	0.012	0.47
1000	0.40	0.010	0.58
1000	0.30	0.008	0.75
1000	0.20	0.006	0.82
800	0.60	0.011	0.53
800	0.50	0.010	0.61
800	0.40	0.005	0.84
800	0.30	0.006	0.91
600	0.65	0.012	0.62
600	0.50	0.009	0.65
600	0.35	0.008	0.82
600	0.20	0.006	0.92

B.3   Metallic fraction $\Gamma > 0.7$, specular reflection $m = 0$

Periodicity Δ(nm)	Metallic fraction Γ	TMOKE$_{MAX}$	R(TMOKE$_{MAX}$)
1000	0.92	0.005	0.95
1000	0.88	0.007	0.90
1000	0.84	0.010	0.83
1000	0.80	0.009	0.85
800	0.90	0.004	0.88
800	0.85	0.007	0.78
800	0.80	0.008	0.76
800	0.75	0.008	0.78
740	0.89	0.012	0.60
740	0.84	0.010	0.63
740	0.79	0.014	0.41
600	0.87	0.007	0.62
600	0.80	0.006	0.63
600	0.73	0.005	0.68
600	0.66	0.007	0.64
400	0.75	0.024	0.22
400	0.63	0.018	0.24
400	0.50	0.017	0.27

Appendix C

Multiferroic heterostructure device

C.1   Structural images

In this appendix we provide SEM images (top-view) of the real ferro- electric and magnetic heterostructure. If figure C.1 the top contact (top electrode) structure can be observed, in which the long-range contacts are placed over a passivation layer, while the magnetoplasmonic active layer (direct contact with BTO) exists only in the regions market by purple squares. Subsequently the magnetoplasmonic active layer and simultaneously top electrode placed over the BTO thin film is shown in figure C.2, where the electric active areas are marked by the red rectangles. Finally the patterned photonic structure is shown in figure C.3 the grating metallic wires also work as ferroelectric top electrode.

C.1.1 Long range structure

FIGURE C.1: SEM image: which shows the long-range top electrodes places over the passivation layer.

C.1.2 Active layers and bridges

FIGURE C.2: SEM image: which shows the electric active areas (marked by the rectangle red) containing the photonic crystal.

C.1.3 Nano-scale, magnetoplasmonic crystals place over BTO

FIGURE C.3: SEM image: shows the grating structure responsible for the ferroelectric switching.

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