Chiral Optics in Corrugated Metasurfaces Interacting with Atomic-width Semiconductors

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A mis padres,
Fernando y Lola.

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Contents

Resumen

La óptica quiral es una subárea de la óptica que estudia la interacción de la luz con materiales quirales. En esta tesis, nos centramos en una estructura prometedora: metasuperficies corrugadas interactuando con semiconductores de anchura atómica, analizando teóricamente sus propiedades ópticas quirales.

En este trabajo, revisamos el marco teórico mínimo necesario para estudiar metasuperficies plasmónicas compuestas por matrices de agujeros rectangulares en láminas metálicas. Utilizando el método de modos acoplados, desarrollamos un nuevo formalismo que permite obtener directamente los coeficientes de reflexión en el campo lejano. Los acoplamientos geométricos son los elementos clave de este procedimiento, ya que establecen las conexiones entre los distintos órdenes de Bragg en el espacio de polarización.

Derivamos las ecuaciones que rigen las metasuperficies e identificamos tanto los términos responsables del *spin-momentum locking*, que establecen reglas de selección entre la polarización y el momento de los órdenes de difracción, como los términos que rompen este comportamiento al mezclar componentes de espín. Esta ruptura se atribuye a la discrepancia entre la definición de luz circularmente polarizada, relativa a la dirección de propagación de los órdenes de difracción correspondientes, y las simetrías del sistema, definidas con respecto a la dirección z de la metasuperficie.

Demostramos que, aunque esta ruptura puede ser despreciable en algunos casos, particularmente cuando los vectores de onda en el plano son pequeños, se vuelve fundamental cuando se excitan plasmones de superficie. En este último caso, pueden establecerse unas reglas de selección modificadas para el *spin-momentum locking*, interpretándose como un proceso de dispersión en dos pasos que involucra la excitación y desexcitación secuencial de un plasmón.

Estos resultados se obtienen también a partir de un formalismo complementario basado en la resolución de las amplitudes del campo eléctrico en las aperturas, en lugar de mediante el nuevo enfoque que involucra directamente los coeficientes de reflexión. Este procedimiento alternativo nos permite encontrar

expresiones analíticas para las amplitudes del campo cercano y esclarecer el papel de las resonancias plasmónicas en las propiedades quirales de las superficies corrugadas.

Asimismo, exploramos la interacción entre metasuperficies quirales y materiales bidimensionales, observando dicroísmo circular en las monocapas debido a la quiralidad inducida por la metasuperficie. Cuando el material 2D se coloca sobre un separador dieléctrico, demostramos que pueden excitarse resonantemente modos de guía de onda de polarización transversal-eléctrica inducidos por el separador, los cuales también producen dicroísmo circular en el material 2D. Este sistema obedece otro conjunto de reglas de selección modificadas para el *spin-momentum locking*, en las cuales el carácter transversal-eléctrico de estos modos sustituye al carácter transversal-magnético de los plasmones. Aplicamos el formalismo desarrollado para (i) estudiar una capa idealizada de moléculas quirales y (ii) analizar separadamente la absorción en los valles K/K' de una monocapa de WS_2. Además, extendemos nuestro análisis a nanoestructuras más sofisticadas, en particular, cavidades quirales, derivando las ecuaciones del sistema compuesto por una matriz de agujeros sobre una matriz de hendiduras.

Finalmente, exploramos el concepto de topología en sistemas fuertemente acoplados. Proponemos un modelo mínimo que presenta la ruptura de la correspondencia borde-volumen observada en estudios previos de sistemas complejos, cuyo origen aún no se ha explicado completamente. Nuestro modelo mínimo consiste en una cadena de primeros vecinos en un modelo *tight-binding* acoplada a una cadena Su-Schrieffer-Heeger. Nuestro trabajo descarta algunos mecanismos posibles de esta ruptura y presenta nueva fenomenología asociada a la ruptura de la correspondencia borde-volumen.

Abstract

Chiral optics is a subfield of optics that explores the interaction of light with chiral materials. In this thesis, we focus on a promising structure: corrugated metasurfaces interacting with atomic-width semiconductors, and study theoretically its chiral optical properties.

In this work, we review the minimal theoretical framework to study plasmonic metasurfaces comprised of arrays of rectangular holes in metallic slabs. Using the coupled-mode method, we develop a novel formalism that directly obtains the reflection coefficients for the far-field. The geometric couplings are the key ingredients of this new approach because they establish the connections between different Bragg orders in polarisation space.

We derive the governing equations of the bare metasurfaces and identify both the terms responsible for the spin-momentum locking, which establish selection rules between the polarisation and the momentum of the diffraction orders, and the terms that spoil this behaviour by mixing the spin components. These breakdown terms are ascribed to the mismatch between the definition of circularly polarised light, relative to the propagation direction of the corresponding diffraction orders, and the system symmetries, defined with respect to the z-direction of the metasurface.

We demonstrate that even though this breakdown can be neglected in some cases, particularly for small in-plane wavevectors, it becomes critical when surface plasmon polaritons are excited. In the latter case, a modified set of spin-momentum locking selection rules can be established, being interpreted as a two-step scattering process involving the sequential excitation and de-excitation of a plasmon.

We also derive these results from a complementary formalism based on solving the amplitudes of the electric field at the apertures rather than from the novel approach involving the reflection coefficients directly. This alternate procedure allows us to find analytical expressions for the near-field amplitudes and elucidate the role of plasmonic resonances in the chiral properties of

corrugated surfaces.

We explore the interaction between chiral metasurfaces and two-dimensional materials, observing circular dichroism in the monolayers due to the chirality induced by the metasurface. When the 2D material is placed on top of a dielectric spacer, we demonstrate that transverse-electric spacer-induced waveguide modes can be resonantly excited, also inducing circular dichroism in the 2D material. This system satisfies another set of modified spin-momentum locking selection rules, including the transverse-electric character of these modes instead of the transverse-magnetic character of the plasmons. We apply the developed formalism to (i) study an idealised layer of chiral molecules and (ii) perform separate absorption analyses in both K/K' valleys of a WS_2 monolayer. We extend our analysis to more sophisticated nanostructures, namely chiral cavities, deriving the equations of the system composed of a hole array placed over a dimple array.

Finally, we explore the concept of topology in strongly coupled systems. We propose a minimal model that presents the bulk-edge correspondence breakdown observed in previous works in complex systems, which is still unexplained. Our minimal model consists of a first-nearest-neighbours tight-binding chain coupled to a Su-Schrieffer-Heeger chain. Our work rules out some possible mechanisms for the breakdown and presents new bulk-edge correspondence breakdown phenomenology.

Chapter 1

Introduction

Journey before destination.

– Brandon Sanderson [1].

1.1 Optics: Background and Evolution

Optics is the branch of physics dedicated to studying the behaviour and properties of light, including its interactions with matter and the construction of related instruments. Optics usually focuses on visible, ultraviolet, and infrared light, all of which are types of electromagnetic (EM) radiation. Other forms of EM radiation, such as X-rays, microwaves, and radio waves, exhibit similar properties due to their electromagnetic nature, although they are not typically categorised as "light".

The term "optics" originates from the Greek word *optikē*, which means "appearance" or "look", and whose root *optikos* pertains to vision or sight [2]. The history of optics dates back to this epoch, with early studies primarily conducted by ancient Greek philosophers. Around 300 BC, Euclid wrote "Optics", which founded geometrical optics by connecting geometry and vision [3]. In the 2nd century AD, Ptolemy contributed to the understanding of refraction and reflection processes [4].

During the Islamic Golden Age, significant advancements were made by scholars such as Alhazen (Ibn Al-Haytham), who wrote the seminal work "Book of Optics" around 1016 AD [5]. Based on observation and experiments, he proposed a new approach to explaining vision and light. He is often regarded as the father of modern optics.

The experimentation with lenses in subsequent centuries led to the invention of the first compound microscope and the first refracting telescope between the late 16th and early 17th centuries [6]. However, in the 17th century, the scientific revolution experienced a major leap in optics. J. Kepler expanded on Alhazen's work and others' [7], while R. Descartes' "La Dioptrique" introduced a mechanical explanation of refraction [8]. Despite this, it was I. Newton and C. Huygens who profoundly shaped the field. Newton's "Opticks" [8], published in 1704, proposed the particle theory of light, while Huygens' wave theory laid the groundwork for understanding light as a wave phenomenon [8].

All these works established the foundations for what we know now as classical or geometrical optics, which deals with the approximation that light travels in straight lines (rays). As mentioned, this period saw the development of lenses and mirrors, as well as the understanding of reflection and refraction, which led to the invention of optical instruments such as telescopes and microscopes.

In the 19th century, the wave theory of light gained eminence with T. Young's double-slit experiment demonstrating interference [9], and A.-J. Fresnel's work on diffraction [9]. On top of that, J. C. Maxwell's equations unified electricity, magnetism and optics, creating the theory of electromagnetism and describing light as EM waves [10].

The 20th century marked the birth of quantum optics. In 1900, M. Planck modelled blackbody radiation using–unwittingly–the concept of energy quanta [11]. This revolutionary idea remained unnoticed for five years [11], until A. Einstein's theory of the photoelectric effect proclaimed and confirmed light's quantization [12]. N. Bohr, in 1913, demonstrated that atoms emit discrete energy amounts, explaining emission and absorption spectra lines [13]. These developments culminated in the theory of quantum electrodynamics, which describes optics and EM processes through photon exchanges. Finally, quantum optics, which studies the quantum mechanical properties of light, became relevant with the inventions of the maser in 1953 and the laser in 1960.

The polarisation of the light plays a key role in this thesis; thus, its historical path is worth mentioning. The first steps in this respect were taken by R. Bartholin, who in 1669 discovered birefringence (or double refraction) in calcite crystals (specifically, Iceland spar) [14]. In the early 1800s, É.-L. Malus discovered that light reflected at certain angles becomes polarised (which is the working principle of polarising sunglasses). His work laid the foundation for Malus's Law, which describes the intensity of polarised light [15]. As mentioned above, in the 19th century, T. Young and A.-J. Fresnel established that polarisation could only be explained if light was a transverse wave. J. C. Maxwell's EM theory of light described polarisation as the orientation of the oscillating electric and magnetic fields. Finally, the development of practical tools, like

the Nicol prism, facilitated the study of polarised light in various fields such as mineralogy and chemistry [16].

Applications of optics are vast and varied. In classical optics, lenses are used in eyeglasses, cameras, microscopes, and telescopes. Besides, reflection and refraction principles are employed in designing periscopes, binoculars, and various optical instruments. On the other hand, modern optics applications include lasers in surgical procedures, telecommunications and data storage. Optical fibres revolutionised telecommunications by enabling high-speed data transmission over long distances. Additionally, holography finds uses in data storage, art, security, and imaging.

The advancement of technology has propelled optics into the nano-optics or nanophotonics era. This cutting-edge field explores light-matter interactions on the nanometer scale, paving the way for innovations in areas such as quantum computing, ultra-sensitive sensors, and high-resolution imaging [17].

1.2   Nano-Optics and Nanophotonics

Nanophotonics, also known as nano-optics, studies the behaviour of light on the nanometer scale, as well as the interaction of nano-objects with light. Therefore, the main objective of this research area is to manipulate and control light at dimensions smaller than its wavelength. This often involves the use of dielectric structures like nanoantennas, or metallic components capable of transporting and focusing light through surface plasmon polaritons (SPPs) [17] or other bound modes. Besides, nanophotonics is an interdisciplinary field, integrating principles from optics, nanotechnology, quantum mechanics, electromagnetism, and materials science.

Conventional optical elements, such as lenses, cannot typically resolve light at deep subwavelength scales due to Abbe's diffraction limit [18]. This defines the minimum distance (d) between two points that can be optically distinguished by a lens or microscope, expressed as:

$$d = \frac{\lambda}{2n \sin \theta},$$ (1.1)

where λ/n is the wavelength of light in a material with refractive index n, and θ is the half-angle of the light cone focusing onto the lens. However, techniques such as localised surface plasmons around nanoscale metal objects or scanning near-field optical microscopy (SNOM) allow light to be confined to nanometer-sized volumes. Using evanescent waves provides a wavevector in a specific direction larger than the free-space wavevector and, by Heisenberg's indeterminacy principle ($\Delta k \, \Delta x \sim 1$), this enlarged Δk allows a reduction in

the real space extension Δx. Yet, as evanescent waves decay exponentially away from the surface, the lateral confinement of light occurs only near the surfaces of the used elements. This confinement enhances light-matter interaction, leading to novel phenomena and practical applications discussed below.

Historically, nanophotonics originated from the mathematical analysis of near-field optics in the early 20th century, which sought to achieve deep-subwavelength resolution. The concept of manipulating light at the nanoscale was first imagined through the work of scientists such as Lord Rayleigh, who explored the scattering of the light through subwavelength particles [19]; or E. H. Synge, who proposed a subwavelength microscope [20].

However, experimental realisations had to wait for 20th-century technology because precise control over these confined EM fields was needed. The development of SNOM in the 1980s provided the experimental tools to visualise and manipulate light at the nanoscale, boosting experimental advancements in nanophotonics [21–24]. Furthermore, the progress in sophisticated nanofabrication techniques such as electron beam lithography [25] and focused ion beam [26] allowed rapid developments in nanophotonics.

As mentioned, confining light using evanescent waves requires a material platform capable of supporting modes with $\Delta k \gg 1$, which led to the research on SPPs. During the whole 20th century, mathematical descriptions and experimental observations of SPPs emerged across diverse domains [27–32]. SPPs are, thus, a pivotal phenomenon in nanophotonics and this thesis. They are collective oscillations of free electrons coupled with EM waves at a metal-dielectric interface, and their dispersion relation is given by:

$$k^{\mathrm{SPP}}(\omega) = \frac{\omega}{c}\sqrt{\frac{\epsilon_M \, \epsilon}{\epsilon_M + \epsilon}}, \tag{1.2}$$

where k^{SPP} is the SPP wavevector, ω is the energy/frequency, c is the speed of light in vacuum, and ϵ_M (ϵ) is the relative permittivity of the metal (dielectric).

Unlike free-space light, SPPs possess a wavevector greater than that of the corresponding light wave in vacuum, resulting in a confined mode that decays exponentially perpendicular to the interface. Because SPPs lie outside the light cone, they cannot be excited by propagating light and their excitation requires special techniques. In this thesis, we will inspect the use of photons and a periodic grating to satisfy the momentum matching condition [33]. However, other procedures can also be undertaken to fulfil this momentum-matching requirement via photons: employing a prism (Kretschmann-Raether and Otto configurations [34, 35]) or a defect on the metal surface [33].

Nanophotonics, in general, and SPPs, in particular, offer a wide range of both potential and well-established applications. Detecting biological and chemical

species can be addressed with unprecedented sensitivity using surface-plasmon-resonance-based sensors [36]. Also, sensitive spectroscopy measurements of even single molecules can be achieved using surface-enhanced Raman scattering (SERS) by creating hot spots of high EM intensity and small volume [37]. On the other hand, photovoltaic cells' efficiency is improved by incorporating plasmonic nanostructures, which enhance light absorption and reduce recombination losses. This indeed constitutes the field of plasmon-enhanced photovoltaics and contributes to the development of sustainable energy [38]. For optoelectronics and optical communication, the miniaturisation of optical elements is possible using spasers (plasmonic lasers) as wires for the light. These elements are desirable due to their subwavelength optical cavities, providing low threshold current (which improves power efficiency) and fast modulation (which increases data transmission) [39].

In summary, nanophotonics is a rapidly evolving field with important implications for technology and society. Its ability to manipulate light at the nanoscale opens up a plethora of new possibilities, driving innovation across multiple domains and promising to transform the way we interact with and harness light.

1.3   Chiral Optics

Light polarisation may serve as a critical degree of freedom, enabling richer and more nuanced interactions between light and matter. For instance, one of its key uses is detecting chirality at the nanoscale. Chirality, derived from the Greek word *kheir* for "hand", refers to a geometric property where an object or system cannot be superposed onto its mirror image. This characteristic is shared across multiple disciplines, including physics, chemistry, and biology. In nature, chirality is observed in numerous cases, such as the helical structure of DNA, the left- and right-handed configurations of amino acids, and the asymmetric shapes of snail shells. These examples illustrate the fundamental and ubiquitous nature of chirality in the physical world.

Merging chirality with optics gives rise to chiral optics, a branch of optics that explores the interaction of light with chiral materials. Chiral materials, which lack mirror symmetry, can influence the propagation and behaviour of light, leading to phenomena such as optical rotation and circular dichroism [40]. L. Pasteur, renowned for his work in the wine industry treating bacterial contamination, deserves special mention as the first to observe optical rotation and demonstrate chirality in molecules [41]. These effects are exploited in various applications, from molecular detection to advanced optical devices [42].

The concept of chirality becomes blurry when it is not applied to stationary elements but to less evident systems such as objects influenced by EM fields [43–45]. The symmetry operations that interconvert distinguishable elements provide the keystone for the distinction between true and false chirality. A true chiral system coincides with its mirror image only by applying space inversion, while a system with false chirality can also be interconverted by time reversal combined with any proper spatial rotation. However, this is not truly relevant for us because we will not distinguish between these two types of chirality through this thesis.

By combining chiral light-matter interaction phenomena with nanoscale manipulation enabled by nanophotonics, chiral metasurfaces rise as a promising platform for advanced studies. They are engineered two-dimensional structures designed to interact differently with circularly polarised (CP) light depending on the handedness of the polarisation (left- or right-handed) and to manipulate light at subwavelength scales. These metasurfaces exhibit unique optical properties, such as asymmetric transmission (different transmission depending on the polarisation or spin of the incoming light) and polarisation conversion, making them valuable for developing photonic devices [46, 47].

Polarisation control in chiral metasurfaces can be conducted using the intrinsic properties of the material or its constituent elements (intrinsic chirality), or employing the geometry of the spatial arrangement of achiral elements (extrinsic chirality) [48, 49]. In the case of the latter, geometric-phase metasurfaces (GPMs), also known as Pancharatnam-Berry phase metasurfaces, exploit the concept of the Pancharatnam-Berry [50, 51] (or geometric) phase to control the phase of light in a spatially varying manner. This phase is acquired throughout a cyclic evolution and depends uniquely on the geometry of the path traversed. These GPMs manipulate the phase of light by varying the orientation of subwavelength structures across the surface, and are commonly used to control the phase front of light in applications such as holography and beam shaping [52–57].

Although GPMs are a subset of chiral metasurfaces and this thesis focuses only on GPMs, they are occasionally referred to as chiral metasurfaces due to their differential behaviour towards left- and right-handed CP light. However, they are strictly GPMs, as the light phase is manipulated by tuning the orientation of their subwavelength constituents (geometric configuration). Furthermore, GPMs are versatile and can operate with various polarisations, extending beyond just CP light.

In these systems, spin-momentum locking (SML) is a phenomenon in which the spin of photons is linked to their propagation direction, being a manifestation of spin-orbit interactions of light. This effect is particularly pronounced in

certain chiral structures, leading to robust and unidirectional light propagation depending on the handedness of the incident light [58, 59]. Chiral plasmons, on the other hand, refer to collective oscillations of free electrons in chiral metallic nanostructures or in GPMs, exhibiting strong chiral optical responses that enable new ways to control and manipulate light at the nanoscale [60].

The field of chiral nano-optics and chiral materials has experienced significant growth in recent years. Researchers have developed highly efficient chiral metasurfaces for polarisation control and multifunctional optical devices [61, 62]. Advances in chiral plasmonics have led to the creation of nanoscale sensors with enhanced sensitivity and selectivity [60, 63], which allow the detection and discrimination of chiral molecules or enantiomers. Furthermore, GPMs have facilitated the development of compact optical components crucial for imaging systems, augmented reality devices, and quantum information processing [62, 64, 65].

1.4   Integration with 2D Materials

Two-dimensional (2D) materials have revolutionised the field of materials science and condensed matter physics. The journey began with the discovery of graphene in 2004 by A. Geim and K. Novoselov, who mechanically exfoliated a single layer of graphite using ordinary scotch tape [66] and measured the electrical conductivity of this one, earning them the Nobel Prize in Physics six years later. Graphene, a single layer of carbon atoms arranged in a hexagonal lattice, exhibits remarkable electrical, optical, and mechanical properties [67, 68]. Its exceptional characteristics inspired a wave of extensive research and development in atomically thin materials [69–72], with potential applications across domains such as electronics, photonics, and energy storage [73–77].

This plethora of 2D materials includes transition metal dichalcogenides (TMDs), hexagonal boron nitride (h-BN), or black phosphorus (BP). These materials exhibit diverse properties –ranging from metallic to semimetallic, semiconducting to insulating, with direct and indirect band gaps spanning from the ultraviolet to infrared spectrum– thereby opening up new possibilities in fields such as nanophotonics, valleytronics, spintronics, sensing, and nonlinear optics, among others [70, 71]. TMDs, for instance, are explored for applications in transistors, sensors, photodetectors, and flexible electronics due to their semiconducting properties [78, 79]. h-BN, often referred to as "white graphene", serves as an excellent dielectric material for encapsulating other 2D materials, and as an ideal substrate for high-quality graphene-based devices [80, 81]. With its tunable bandgap, BP shows promise in field-effect transistors, optoelectronics and photodetectors [82, 83].

In the field of nanophotonics, 2D materials hold particular significance due to their strong light-matter interactions and tunable electronic properties [74]. Graphene is a conductor that exhibits broadband absorption and exceptional carrier mobility, which has paved the way for ultrafast photodetectors and modulators [84–86]. Moreover, graphene supports plasmons that are 100 times more confined than SPPs in metals and are tunable using an external static electric field (in the infrared and THz regimes) [87–89]. Notably, 2D semiconductors can support excitons –bound electron-hole pairs– at room temperature, making them ideal candidates for photonic and optoelectronic applications [74]. Their atomic thinness and unique properties have established them as an indispensable research platform, also for studying fundamental physical phenomena [70, 79]. Moreover, their compatibility with existing semiconductor technology facilitates the development of innovative devices [70, 73, 78]. Among these materials, TMDs stand out for their strong photoluminescence and valleytronic properties [90], which hold promise for optoelectronic applications such as light-emitting diodes (LEDs) [91] or valley-polarised LEDs [92, 93].

1.5   Hybrid Nanostructures

In recent years, coupling plasmonic metasurfaces with 2D materials has attracted significant attention in nanophotonics and materials science fields. As mentioned, plasmonic metasurfaces, composed of subwavelength metallic structures, can manipulate EM waves at the nanoscale, leading to precise control over light-matter interactions [17, 46, 94]. When integrated with 2D materials, these metasurfaces form hybrid systems exhibiting novel functionalities. Recent advancements have demonstrated that such coupled systems can improve the efficiency of photodetectors [95], sensors [96, 97], and light-emitting devices [98], among other applications [99]. Furthermore, these developments are significant not only for their technological implications but also for advancing the fundamental understanding of light-matter interactions at the nanoscale [100].

Diverse plasmonic platforms have been investigated in this respect, including metallic nanowires [101, 102], metallic nanoparticles [95, 103, 104], and anisotropic plasmonic arrays [105]. 2D materials combined with metasurfaces have also been engineered to exploit the unidirectional propagation of surface plasmons and enhance chiral interactions [106]. Naturally, numerous 2D materials have also been studied for their interaction with plasmonic structures: graphene [96, 103], h-BN [80, 81], among others. Especially relevant to this thesis are the TMDs, which are investigated for their strong light-matter interactions and valley-dependent phenomena [106–108].

1.6   Motivation and Objectives of the Thesis

Although the study of hybrid nanostructures has blossomed over the last decade, the coupling of GPMs and 2D materials with chiral properties remains underdeveloped. The 2D materials we are most interested in are the TMDs because they are semiconductors with remarkable properties in nanophotonics. They also feature a honeycomb lattice structure, and because of this, their chirality emerges from their valley-selective behaviour: depending on the spin of the circularly polarised light, different emission and absorption features are observed at the K and K' valleys of TMDs. At the same time, GPMs are considered to provide a chiral platform to selectively excite SPPs depending on the spin and wavevector of the incident light. Both of these components are exceptional for enabling precise control and manipulation over the polarisation and directionality of light at the nanoscale [109]. Therefore, combining them could lead to progress in valleytronics, as well as the emergence of novel and synergistic phenomena, with potential advancements in fields such as chiral sensing, imaging, and quantum information processing [106, 110, 111].

Especially relevant for this thesis was the work [105] by Prof. Ebbesen's research group in 2018. They presented the chiral coupling of valley excitons from a WS_2 monolayer with spin-momentum-locked plasmons generated by a GPM, coining the resulting polaritons as chiralitons. They pursued the achievement of strong coupling between both plasmonic and excitonic modes. A visual abstract of their work is presented in Figure 1.1(a), a SEM image of the GPM in Figure 1.1(b), and their main results in Figures 1.1(c-f).

Figures 1.1(c, d) show the absorptance of the system for incident spin $+$ and spin $-$ light, respectively. The interplay between the exciton of the WS_2 monolayer with the spin-momentum-locked plasmon from the GPM is observed, noting that the excited plasmon depends on the spin of the incident light. On the other hand, Figure 1.1(e) shows the difference in reflectance for incident spin $-$ and spin $+$ light, proving again the spin-selective excitation of SPPs. Finally, Figure 1.1(f) also represents the difference in reflectance but for incident p-polarised and s-polarised light instead. In this case, the equal contribution of TM and TE light into spins $\pm$ is observed, resulting in equal reflectances for both polarisations in the central spectral region.

The analysis of those results was based on phenomenological models. The main motivation for this thesis was then to comprehend how the SML emerges in the GPMs, how the spin-momentum-locked plasmons are generated and their characteristics, and how these phenomena are merged with the valley excitons from the TMDs. A theoretical and numerical analysis of this interaction could yield new insights into the behaviour of combined 2D materials and chiral
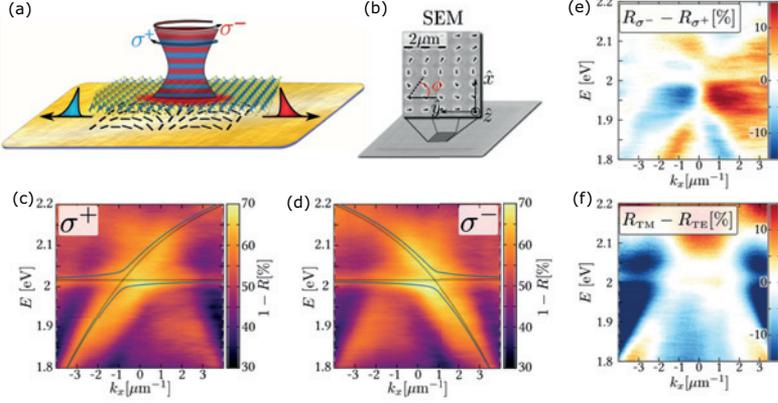
Figure 1.1: Selection of figures from [105]. (a) Visual abstract of the work. (b) SEM image and scheme of the plasmonic metasurface. (c, d) Absorptance of the system for incident spin + and spin − light, respectively. (e, f) Differences in reflectance for two sets of incident light: spin − and spin + in (e); p and s in (f). Reprinted (adapted) with permission from T. Chervy, S. Azzini, E. Lorchat, S. Wang, Y. Gorodetski, J. A. Hutchison, S. Berciaud, T. W. Ebbesen, and C. Genet, "Room temperature chiral coupling of valley excitons with spin-momentum locked surface plasmons", ACS Photonics 5, 4, 1281–1287 (2018) [105]. Copyright 2018 American Chemical Society.

metasurfaces. Furthermore, shortly before the beginning of the thesis, another topological phenomenon appeared in photonics: the breakdown of the celebrated bulk-edge correspondence in plasmonic chains within a cavity [112]. The study of a minimal model has been undertaken in this thesis to elucidate the origin of this breakdown.

The objectives of each chapter in this thesis are as follows:

- Chapter 2 aims to establish the theoretical framework for studying the GPMs proposed throughout this thesis. Using a minimal model for a hole array, we explain how to analyse this kind of structures via two different procedures: the standard way, which involves calculating the electric field amplitudes at the holes' apertures, and a novel formalism that computes the system's reflection coefficients directly. Finally, we extend this formalism to address the study of dimple arrays (our GPMs) in an opaque metal film.

- In Chapter 3, we focus on the study of the bare GPMs, developing this novel approach based on the reflection coefficients. Under this framework, we demonstrate the emergence of the SML and its breakdown in the equations, aiming to deepen our understanding of spin-momentum-locked plasmons. We examine different GPMs: step-wisely rotated dimples

along x-direction (as in [105]), Kagome lattice, hexagonal lattice, and graphene-like lattice; all of them composed of rectangular dimples, in order to find relations between system geometry and optical properties.

- Chapter 4 pursues similar goals as Chapter 3 but through the standard approach of the coupled-mode method by computing the electric field amplitudes at the apertures. We analyse the bare metasurface through their eigenvalues and eigenstates, allowing us to identify the plasmonic response of the GPM as the collective eigenmode of the metasurface. This detailed eigenanalysis also proves the emergence of the SML and its breakdown, as we did in Chapter 3.

- In Chapter 5, we aim to portray the interplay between excitons from 2D materials with the SPPs emerging from GPMs. We provide the theoretical description to characterise the system composed of a 2D material over a GPM. We study several types of 2D materials, although our main focus is on TMD monolayers (WS_2 monolayer in particular). We extend the limits of our formalism by examining idealised thin layers of molecules and trying to capture the K/K' valleys of TMDC monolayers.

- Chapter 6 extends the range of considered GPMs by taking into account chiral cavities as platforms for interacting with 2D materials. The novelty of these chiral cavities resides in placing a hole array over a dimple array, creating an in-region where a 2D material could be placed. We present the theoretical description of this new system, the quantities of interest, and the numerical simulations of the total absorptance and the electric fields inside and above the cavity.

- Chapter 7 gathers the study of several topological ladders to identify the origin of the bulk-edge correspondence (BEC) breakdown in this kind of strongly coupled systems. We review previous works on strong coupling in cavities and the SSH model to introduce the concept of BEC breakdown. We describe our approach for a minimal model capable of characterising the BEC breakdown.

- In Chapter 8, we summarise the main conclusions of each chapter. We comment on the relevance of the work, as well as the future directions.