Preface

As everybody has experienced by looking at a mirror, light is almost completely reflected by metals. But they also exhibit an amazing property that is not so widely known: under some circumstances light can “flow” on a metallic surface as if it were “glued” to it. These “surface” waves are called surface plasmon polaritons (SPPs) and they were discovered by Rufus Ritchie in the middle of the past century. Roughly speaking, SPP modes generate typically from the coupling between conduction electrons in metals and electromagnetic fields. Free electrons loose their energy as heat, which is the reason why SPP waves are completely absorbed (in the visible range after a few tens microns). These modes decay through so short lengths that they were considered a drawback, until a few years ago. Nowadays that situation has completely turned. Nano-technology now opens the door for using SPP-based devices for their potential in subwavelength optics, light generation, data storage, microscopy and bio-technology.

There is a lot of research done on those phenomena where SPPs are involved, however there is still a lot of work to do in order to fully understand the properties of these modes, and exploit them. Precisely, throughout this thesis the reader will find a part of the efforts done by our collaborators and ourselves to understand the compelling questions arising when light “plays” with metals at the nanoscale. The outline of the thesis is:

i. Chapter 1: *Introduction*
   First, the fundamentals of SPPs are introduced. In fact, SPPs will be one of the most important ingredients in order to explain the physical phenomena investigated in this thesis.
   Our contributions, from a technical standpoint, have been carried out with the help of two different well known theoretical methods: the finite-difference time-domain (FDTD) and the coupled mode method (CMM). In this chapter, we summarize the most relevant aspects of these two techniques, looking for a better comprehension of the discussions raised along the remaining chapters.

Concerning the rest of experimental and theoretical techniques used, it is out of the scope of this thesis to rigorously describe all of them. Nevertheless, most of those methods, which will not be presented in the introductory chapter, will be briefly explained when mentioned.

ii. Chapter 2: *Extraordinary Optical Transmission*

Imagine someone telling you that a soccer ball can go through an engagement ring. At first, you could think that he or she has got completely mad. A situation like that could have been lived by the researchers who first reported on the extraordinary optical transmission (EOT) phenomenon. Thomas Ebbesen and coworkers found something like a "big" ball passing through a hole several times smaller than it, although there, the role of the ball was played by light. Before Ebbesen's discovery light was not been thought of being substantially transmitted through subwavelength holes. Until 1998, a theory elaborated by Hans Bethe, on the transmission through a single circular hole in a infinitesimally thin perfect conducting screen, had "screened" out any interest in investigating what occurs for holes of subwavelength dimensions. Bethe's theory demonstrated that transmission through a single hole, in the system described above, is proportional to $(r/\lambda)^4$where λ is the wavelength of the incoming light, and r is the radius of the hole. The proportionally constant depends on hole shape, but it is a small number (~ 0.24 for circular holes). It is clear that whenever $\lambda \gg r$ transmission is negligible. Nevertheless, Ebbesen and coworkers experimentally found that light might pass through subwavelength holes if they were periodically arranged on a metal surface. More importantly, in some cases even the light directly impinging into the metal surface, and not onto the holes, is transmitted. The SPP modes were pointed to be responsible of EOT.

It is not strange that such a breakthrough sparked a lot of attention in the scientific community. Furthermore, the EOT discovery is not only interesting from the fundamental physics point of view, but from the technological side as well.

The EOT phenomenon strongly depends on both geometrical parameters and material properties. Moreover, EOT does not only occur in two dimensional hole arrays (2DHAs), so other systems have been investigated in the last years. In this way, this thesis is partly devoted to study different aspects of EOT:

(a) We begin by investigating the influence of the chosen metal on EOT using the FDTD method. We analyze transmission spectra through hole arrays drilled in several optically thick metal films (viz. Ag, Au, Cu, Al, Ni, Cr and W) for several periods and hole diameters proportional to the period.

(b) We also study the optical transmission through optically thin films, where the transmission of the electromagnetic field may occur through both the holes and the metal layer, conversely to the "canonical" configuration where the metal film is optically thick, and the coupling between metal sides can only be through the holes.

(c) On the other hand, since the first experimental and theoretical papers some controversy arose over the mechanisms responsible to enhance optical transmission through an array of holes. Two mechanisms lead to enhanced transmission of light in 2DHAs: excitation of SPPs and localized resonances, which are also present in single holes. In this chapter we analyze theoretically how these two mechanisms evolve when the period of the array is varied.

(d) There are systems displaying EOT different from holey metallic films. One of them is built by monolayers of close-packed silica or polystyrene microspheres on a quartz support and covered with different thin metal films (Ag, Au and Ni). We show that the optical response from this system shows remarkable differences as compared with the "classical" 2DHA configuration.

iii. Chapter 3: *Theory of NRI Response of Double-Fishnet Structures*
Veselago demonstrated that the existence of an isotropic, homogeneous and lineal (i.h.l) medium characterized by negative values of both the permittivity (ε) and the permeability (μ) would not contradict any fundamental law of physics. A substance like that is usually called left-handed material or alternatively, it is said to posses negative refraction index (NRI), and it behaves in a completely different fashion from conventional materials. At the interface between a NRI material and a conventional dielectric medium interesting things would happen. For instance, the current transmitted into a NRI medium would flow through an "unexpected" direction, forced by the Maxwell's equation boundary conditions. Unluckily, no natural material is known to posses a negative value of its refractive index. To date, the only way to achieve NRI materials is by geometrical means. Nevertheless the optical properties of the constituting materials are still important. For instance, as the dielectric constant of metals is "intrinsically" negative, NRI researchers explore how to induce negative permeability on them by designing their geometry in particular ways. This is the reason why these kind of materials are usually called "meta-materials" because their optical response may be different than the optical response of its bulk components. In this chapter we investigate the optical response of one of these metamaterials presenting NRI, a two-dimensional array of holes penetrating completely through a metal-dielectric-metal film stack (double-fishnet structure).

iv. Chapter 4: *Plasmonic Devices*
The special properties of SPPs are being considered for potential uses in circuits. Namely, the possibility of building optical circuits aimed by SPPs has sparked a great interest in the scientific community. As SPPs on a flat surface propagate close to the speed of light, an hypothetical optical SPP-device would be faster than its electronic counterpart. Moreover, different frequencies do not interact, thus several channels would be available for

sending information. A last advantage, SPP-based technology would be compatible to electronic technology since both share the same supporting medium. Transporting optical signals and/or electric ones would be then possible, depending on the characteristics of a specific instrument.

On the contrary, two disadvantages in the use of SPPs instead of electrons arise: **(i)** SPPs are much more difficult to control than electrons on metallic structures (e.g. surfaces), being efficiently scattered by defects present on them, and **(ii)** the finite propagation length of SPP modes. Note that the latter would not be an actual inconvenient in the case of highly miniaturized circuits. Although the SPP modes are well positioned candidates, as we say, they are strongly scattered by any relief on the surface and, due to the mismatch between freely propagating waves and SPPs, they are difficult to be properly excited. A lot of theoretical and experimental works have been devoted on how to guide and generate SPPs.

Regarding the coupling mechanism of light with SPPs, note SPPs can not be excited by an incident plane-wave, because of their evanescent character. There are various coupling schemes that allow light and SPPs to be coupled: prism coupling, grating coupling and near-field coupling. These setups for exciting SPPs are not always useful for certain applications. In Chap. 4 we discuss the advantages and disadvantages of those methods, and we demonstrate a device that enables to create a source for SPPs with remarkable advantages with respect to the other proposals.

In the same chapter we explore different ways for guiding SPP-like modes. Devices for guiding SPPs by means of metallic bumps or holes drilled on a metal surface have been suggested. Another possibility is to guide electromagnetic waves by either a channel cut into a planar surface or a metallic wedge created on it. These structures support plasmonic modes called channel plasmon polarions (CPPs) and wedge plasmon polarions (WPPs) respectively. The surface could be either a metal or a polar dielectric, characterized by negative dielectric constant values. We investigate both CPPs and WPPs by means of rigorous simulations, aimed to elucidate their characteristics, especially, at telecom wavelengths.

We use that information for suggesting a $SPP \leftrightarrow WPP$ conversion device. Lastly we study how gradually tapering a channel carved into a metal surface enables enhanced electromagnetic fields close to the channel apex.

v. Chapter 5: *Optical Field Enhancement on Arrays of Gold Nano-Particles*
Light scattering by arrays of metal nanoparticles gives rise to nanostructured optical fields exhibiting strong and spatially localized field intensity enhancements that play a major role in various surface enhanced phenomena. In general, local field enhancement effects are of high interest for fundamental optics and electrodynamics, and for various applied research areas, such as surface enhanced Raman spectroscopy and microscopy, including optical characterization of individual molecules. Furthermore, the highly concentrated EM fields around metallic nanoparticles are thought to enhance, in turn, non-linear effects, which can pave the way for active plasmonic-

based technologies. Also biotechnology can take advantage of such high intensified optical fields. It is well known that individual metal particles can exhibit optical resonances associated with resonant collective electron oscillations known as localized surface plasmons (LSPs). Excitation of LSPs results in the occurrence of pronounced bands in extinction and reflection spectra and in local field enhancement effects. Such nanoparticles periodically arranged, may cause additional interesting effects. Besides, if nanoparticles are deposited on a metal surface, the emergence of a new channel for light being excited (SPPs) may lead to new phenomena. In this chapter we investigate the optical response of arrays of gold nanoparticles on both dielectric and metal substrates. By means of the FDTD method we analyze the experimental results consisting on: reflection and extinction spectra measurements along with the non-lineal response known as two-photon excited (photo) luminescence (TPL) generated by inter-band transitions of d-band electrons into the conduction band.

http://www.springer.com/978-3-642-23084-4

Optical Properties of Nanostructured Metallic Systems
Studied with the Finite-Difference Time-Domain Method
Rodrigo, S.G.
2012, XX, 163 p. 95 illus., 9 illus. in color., Hardcover
ISBN: 978-3-642-23084-4