Scattering, loss, and gain of surface plasmons

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Chapter 1

Introduction

The purpose of this introduction is to put this thesis in a broader perspective, namely the opportunities offered by nanoscale structuring of materials to obtain novel optical properties. To this aim we will sketch some developments in the last decade in the field of optics which inspired us. Simultaneously some general concepts that are often used throughout this thesis are outlined. The introduction concludes with a brief overview of the chapters in this thesis and the relation between these chapters is explained.

1.1 Possibilities of structuring materials on the nanoscale

In its most general sense, this thesis is about structuring materials on the nanoscale to obtain new optical properties or new optical instruments. Structuring means for example: perforation of a slab with holes; growing layers of different materials on each other; or creating little spheres, rods or bow ties. These structures range in size from a few tens of nanometers to a few hundred nanometer, i.e. much smaller or comparable to the wavelength of visible light. Using this nanoscale fabrication, scientists wish to expand the traditional spectrum of optical components: lenses, mirrors, filters, gratings, LED's and lasers.

Structuring materials on the nanoscale is already used commercially, for example in a simple mirror. Often metals are used as a mirror, but these are not ideal. In the case of a gold mirror this is exemplified by its yellowish color, which corresponds to the absorption of the blue part of the light spectrum. By stacking a number of subwavelength layers of two different transparent media an ideal mirror can be made for a given wavelength range. In these so-called Bragg reflectors, interference of light is cleverly used to obtain this ideal mirror.

Another famous example is a photonic crystal, which is the optical analogue of a semiconductor for electrons. It can consist, for example, of a three dimensional matrix of nanoscale spheres. These crystals can, theoretically, prevent an excited atom within the crystal to radiate and thus the atom remains excited [1, 2]. The reason is that, for a particular range of frequencies the only solution of Maxwell's equations inside the photonic crystal is to have no light at all.

Another fascinating example is the invisibility cloak, which is subject of current research. Such a cloak directs light around an object, in such a way that it seems as if the object is not there at all [3]. Some experimental demonstrations are recently reported [4, 5], albeit for limited sizes of the cloaked object. For an example see Fig. 1.1a, where a bump in a metallic mirror is made invisible [4], that is the mirror seems flat when observed through the cloak. This cloak is achieved by making a cleverly chosen hole pattern in the material that was placed in front of the mirror.

1.1.1 Added value of metals

The photonic crystal and the invisibility cloak can already be made using transparent media like semiconductors or polymers. For other applications the special optical properties of metals are explored. The main advantage of a metal is that it interacts strongly with light. This is exemplified by the high reflection of a metal mirror, whereas a single glass interface reflects only a few

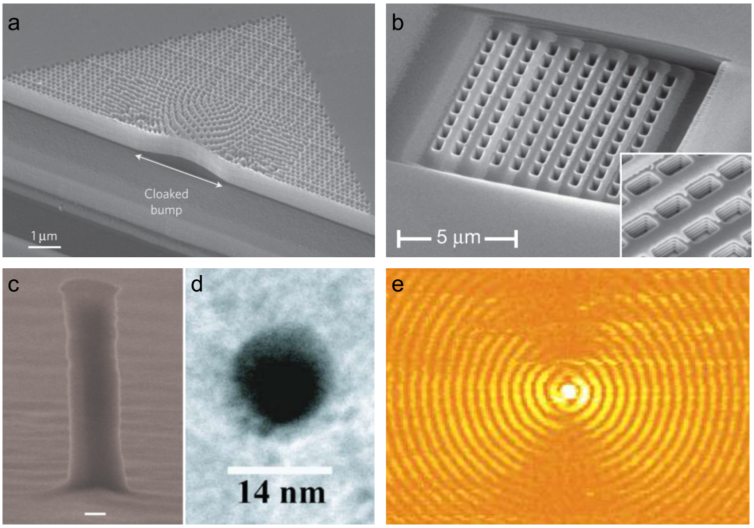

Figure 1.1: A selection of recent experimental results. **a,** An invisibility cloak. **b,** A negative index metamaterial. **c,** A metal coated nano pillar laser. The laser light will be confined within the pillar shown (scale bar is 100 nm). **d,** An image of a nanosphere laser. The black part is a gold nanosphere, and the gain medium is placed around this sphere. **e,** A near field image of a surface plasmon at a metal-air interface, excited at a subwavelength hole [10]. Figure reproduced with permission from: **a,** ref. [4], © 2009 NPG; **b,** ref. [8], © 2008 NPG; **c,** ref. [11], © 2007 NPG; **d,** ref. [12], © 2009 NPG; **e,** ref. [10] © 2005 ACS

percent of the light (at small angles).

A fascinating example of the possibilities that metals have to offer is that a negative refractive index can be made using metals [6–8]. This means that light does not refract as always presented in textbooks, but it will refract in the opposite direction. An experimental demonstration of this effect is performed using the structure in Fig. 1.1b which is a small prism of negative index material. Also an artificially made refractive index of zero is demonstrated, using a combination of metals and dielectrics [9]. Hence, the phase velocity of light in this structure exceeds the speed of light in vacuum.

1.2 Losses and loss compensation

The advantages that metals have to offer, are at the cost of the losses that metals suffer from. In particular for visible light, metals have tremendous losses, that seem to hinder further application of metals in optics. In some systems the losses are partially caused by imperfections induced by the nanofabrication [13, 14]. Also cooling [15] and annealing [16] can improve the performance of a metal structured on the nanoscale. Alternatively, losses may also be reduced by cleverly engineering alloys [17]. Nonetheless, the ohmic losses seem to be inevitable and a strategy that can fully eliminate losses is the compensation of losses using a so-called gain medium.

A gain medium is a material that is supplied with energy from an external source, named a pump. This energy can be transferred to an optical field that propagates through the medium, via the process of stimulated emission. In many cases, gain can be considered as the opposite of loss, as the amplitude of a wave traveling through the gain medium increases exponentially instead of decaying exponentially. Of course energy needs to be conserved, and therefore there can not be more energy extracted from the gain medium than supplied by the pump.

1.2.1 Lasing as advantage and a nuisance

Another important aspect of a gain medium is that it can be used to create a laser [18–20], where the word laser is an acronym for light amplification by stimulated emission of radiation. Besides the light amplification, lasers also have an additional element: a resonator. This resonator provides feedback to the light emitted in the cavity. Using this feedback mechanism the properties of the stimulated emission can be controlled, for example: wavelength, polarization and the intensity profile of the laser beam.

Nowadays there is a need for making nanoscale lasers. These lasers may be integrated into computer systems or could be of value in display technology. For these nanoscale lasers metals are considered, because the metals allow one to scale down the volume of a laser dramatically. An example of a nanopillar laser [11] and a nanosphere laser [12] is shown in Figs. 1.1c and 1.1d respectively.

Lasing may also be a nuisance when trying to compensate the losses of a metal nano structure [21, 22]. The laser light may be much brighter than the signal of interest or the available gain may be limited by the laser action. Hence, the subject of loss compensation is not as simple as only adding the gain material.

1.3 Our experiments: surface plasmons and subwavelength holes

In this thesis we will study the issues of loss, loss compensation and lasing, using so-called surface plasmons. The reason for choosing surface plasmons is that they are relatively simple and well understood. Hence, they allow us to understand the influence of loss compensation and lasing better. Surface plasmons are first predicted in 1957 [23]. Because one can not couple to a surface plasmon by simply illuminating a metal surface with light, clever techniques are required to excite surface plasmons. These techniques involve, for example, surface roughness, prisms [24, 25], diffraction gratings [26, 27], and subwavelength holes in the metal film [28].

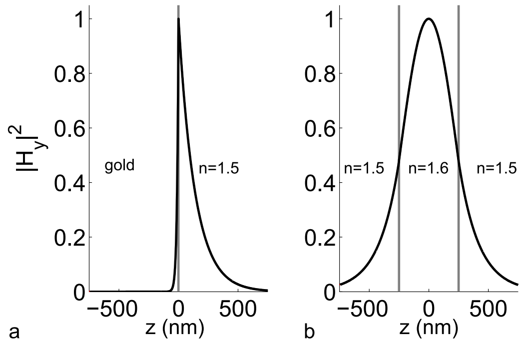

Figure 1.2: A comparison of the mode profile of a surface plasmon (**a**) and a waveguide mode (**b**) at a free space wavelength of 800 nm. The surface plasmon intensity is more strongly confined and is maximal at the interface. These advantages are at the cost of absorption losses.

A surface plasmon is a solution of Maxwell's equations in which light is confined to a metal-dielectric interface. This is illustrated in Fig. 1.2, where we show the absolute square of the magnetic field of a typical surface plasmon on a metal-dielectric interface. We compare the surface plasmon to a typical waveguide in which the light propagates at the same phase velocity (see right). There are three important differences that are unique to surface plasmons: the mode size is smaller, only a single interface is needed for wave guiding, and the field maximum is at the interface instead of inside the waveguide.

These differences are exploited in different applications. The field maximum on the interface makes surface plasmons very sensitive to the presence of any disturbance placed there. This advantage is exploited in different kind of biosensors [29]. The small surface plasmon mode size allows strong confinement of optical fields [30–32], a property that is meritorious in nanophotonics.

These advantages are at the cost of the absorption losses that surface plasmons experience, which is a consequence of the field penetration into the absorbing metal. These losses increase with increasing confinement of the surface plasmon, because a larger fraction of the light is inside the metal. As an illustration of these losses, the typical propagation length of a surface plasmon is of the order of 10 μm, whereas light in optical fibers can be used to transmit data across the Atlantic Ocean.

In our experiments, we use subwavelength holes in a thin metal film to excite surface plasmons. These holes scatter the incident light in almost all directions and therefore part of the light is also coupled to surface plasmons. In Fig. 1.1e, the surface plasmon field excited at a subwavelength hole is made visible in an experiment [10].

In 1998 it has been shown that if these hole are placed in an array, see Fig. 1.3a, with the holes placed roughly a wavelength apart, the metal film exhibits an unexpectedly large transmission at certain resonant wavelengths [28]. This so-called extraordinary optical transmission was interpreted in terms of surface

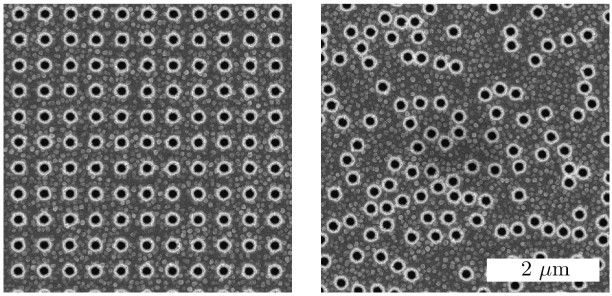

Figure 1.3: Scanning electron microscope images of two patterns that are used throughout the thesis: left an array of subwavelength holes and right a random pattern of the same holes.

plasmons that are excited at one hole and coupled out at other holes. This interpretation has been subject of intense debate [33–37]. The periodicity of the array allows for a resonant excitation of the surface plasmons, which makes the process more efficient [38] but it also limits the phenomenon to particular wavelengths and angles.

1.4 Overview of this thesis

In Chapters 2 to 5 we study the interaction between the surface plasmons and the subwavelength holes. We use many of these insights in Chapters 6 and 7 to study the effect of gain on the surface plasmons.

In Chapter 2 we show experimentally that not only surface plasmons contribute to the extraordinary optical transmission of metal hole arrays. Chapter 3 presents a study of random patterns of subwavelength holes, shown in Fig. 1.3b, where we focus particularly on the question whether the transmission of these patterns is dominated by surface plasmons or by light that is transmitted through the holes directly.

We continue our work on random patterns in Chapters 4 and 5, by applying the concept of speckle correlation functions to surface plasmons. Chapter 4 shows that this technique allows us to quantify the radiative and non-radiative surface plasmon losses. We extend this work in Chapter 5, showing that losses induced by the holes on the surface plasmon can be understood using the well-known Rayleigh scattering.

In Chapter 6 we demonstrate loss compensation of surface plasmons on a metal hole array. We measure the transmission of a hole array with a gain layer in its close proximity and show that the transmission increases up to a factor 31 as a result of the gain. In Chapter 7 we show that in these same hole arrays we also observe surface plasmon lasing. This intriguing result proofs that we have fully compensated the absorption loss of surface plasmons.