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Scattering, loss, and gain of surface plasmons

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Scattering, loss, and gain of surface plasmons

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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

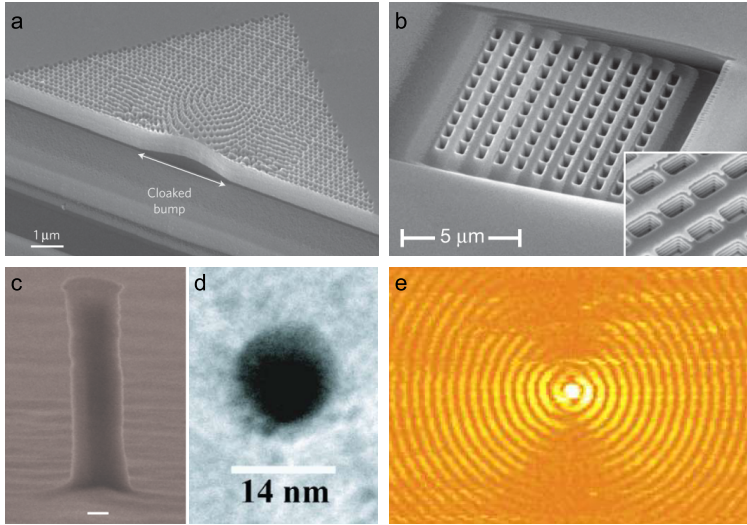


Figure 1.1: A selection of recent experimental results. **a**, An invisibility cloak. **b**, A negative index metamaterial. **c**, A metal coated nano pillar laser. The laser light will be confined within the pillar shown (scale bar is 100 nm). **d**, An image of a nanosphere laser. The black part is 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].

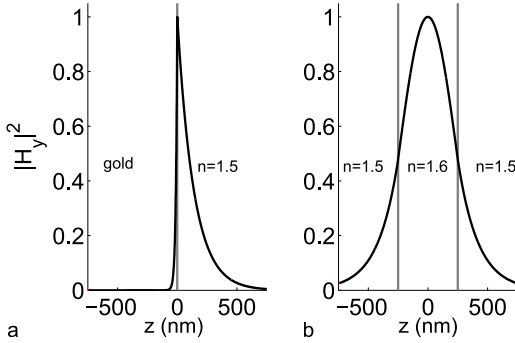


Figure 1.2: A comparison of the mode profile of a surface plasmon (a) and a waveguide mode (b) at a free space wavelength of 800 nm. The surface plasmon intensity is more strongly confined and is maximal at the interface. These advantages are at the cost of absorption losses.

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.

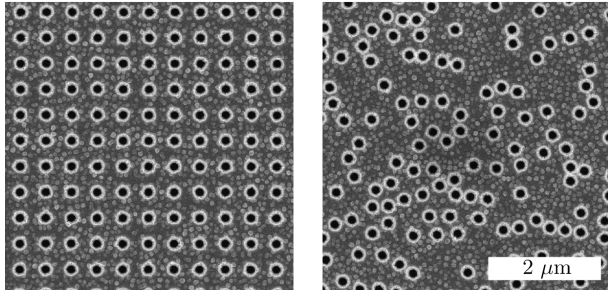
These differences are exploited in different applications. The field maximum on the interface makes surface plasmons very sensitive to the presence of any disturbance placed there. This advantage is exploited in different kind of biosensors [29]. The small surface plasmon mode size allows strong confinement 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.

Quasi-cylindrical wave contribution in experiments on extraordinary optical transmission

A metal film perforated by a regular array of subwavelength holes exhibits an unexpectedly large transmission at particular wavelengths, named the extraordinary optical transmission (EOT) of metal hole arrays [28]. EOT was first attributed to surface plasmon polaritons (SPP), reawakening a large interest in plasmonics [39–41] and subwavelength metallic surfaces [30, 42, 43]. Experiments soon revealed that the field diffracted at a hole or slit is not a SPP mode alone [34]. Further theoretical analysis [36] predicted that the extra contribution, the quasi-cylindrical wave (CW) [35, 44–46], impacts the EOT phenomenon too. In this chapter, we experimentally demonstrate the relative importance of the SPP and CW in EOT by considering hole arrays of different hole densities. From the measured spectra we extract microscopic scattering parameters, which allow us to evidence that the CW only impacts the EOT for high densities, when the hole spacing is roughly one wavelength. Besides providing a deeper understanding of the EOT and the related Wood’s anomaly, the extraction of microscopic scattering parameters from the measurement of macroscopic optical properties paves the way towards novel design strategies.

F. van Beijnum, C. R  tif, C. B. Smiet, H. T. Liu, P. Lalanne, and M. P. van Exter, *Nature* **492**, 411 (2012).

2.1 Introduction

Understanding the EOT quantitatively has been a major challenge in the past decade (see ref. [43] for a review). An important contribution to EOT may be the quasi-cylindrical wave (CW), which is the field on the metal surface diffracted at a subwavelength indentation, in addition to the surface plasmon contribution [35, 44, 46]. The justification to separate these two field contributions lies in the fact that the CW also exists in the absence of surface plasmons, namely in the case of a perfect electric conductor interface [35, 45, 47], and for a dielectric interface [48]. For the perfect electric conductor, the CW is nothing else than a cylindrical wave in free space with a $x^{-1/2}$ decay rate. For metals of finite conductivity at optical frequencies, the cylindrical behavior is only seen in the vicinity of the indentation, at a few wavelengths distance. At larger distances, the CW decay rate keeps on increasing gradually until it reaches an asymptotic algebraic value ($x^{-3/2}$) at large distances. Because of the periodicity of hole arrays, all these decay rates are simultaneously present in the EOT phenomenon.

Another important result in the understanding of EOT is the development of a semi-analytic SPP model, that quantifies the SPP contribution to EOT. This microscopic model relies on scattering parameters that describe the interaction between light and the holes. The microscopic model correctly predicts the EOT lineshape, but underestimates the magnitude of the transmission by roughly a factor two for visible frequencies. These results suggest that the SPP is only responsible for roughly half of the EOT phenomenon [36], whereas the other half is due to the CW. So far, this interpretation is purely theoretical and remains conceptual and we are not aware of any experimental confirmation yet. The reason is probably that many scattering parameters in the model are dispersive and had to be calculated using complicated simulations [36]. Measuring all these parameters with sufficient accuracy is a tremendous experimental issue that we challenge here.

In this chapter, we provide a direct evidence for the respective role of SPP and CW in the EOT, by exploiting the fact that the SPP and CW have different characteristic damping lengths. This is achieved by measuring the transmission spectra of a series of hole arrays with varying hole density, designed to resonate at the same near-infrared frequency (≈ 750 nm). We find that the normalized transmission peaks of all the low-density arrays are virtually identical. The largest density array, which corresponds to the classical hole array in [28], has a hole spacing that corresponds to the CW damping length of one wavelength. This array shows a sudden boost of the normalized transmission compared to the transmission of the other arrays. The observa-

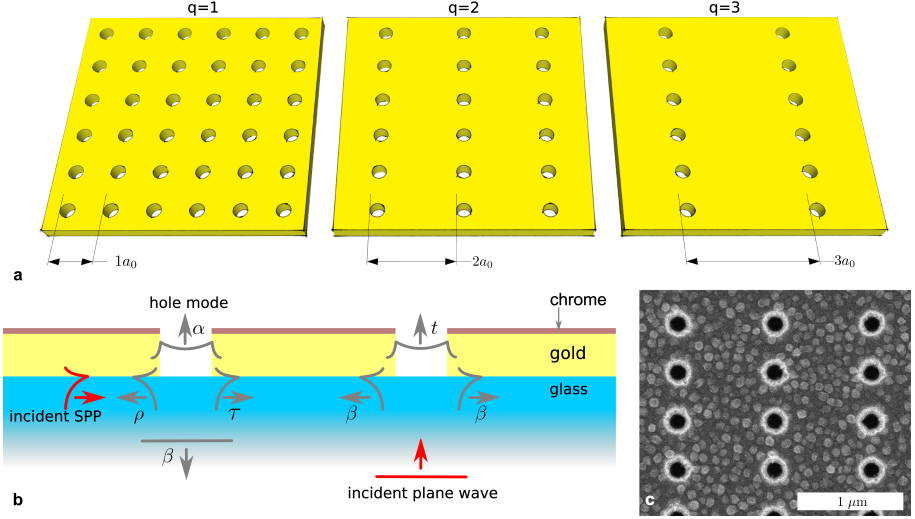


Figure 2.1: An illustration of our sample design. To demonstrate a quasi-cylindrical wave contribution to EOT, we compare the transmission of a series of hole arrays with different densities that all resonate at the same wavelength. **a**, To decrease the hole density, the distance between so-called hole chains is increased. **b**, The relevant scattering processes are sketched. At the left hole there is a surface plasmon incident (red arrow) that is either transmitted (τ), reflected (ρ), coupled into the hole (α) or to free space (β). At the right hole a free space mode is incident (red arrow), which scatters to a surface plasmon (β) or couples into the hole (t). **c**, A scanning electron microscope picture of the $q = 2$ hole array.

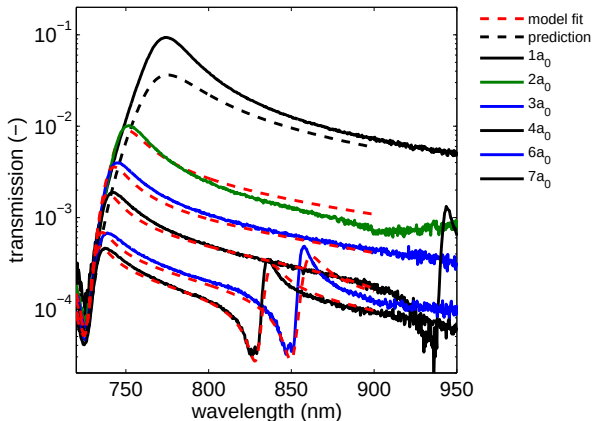
tion of this atypical behavior for the largest density array constitutes a direct proof of the embodiment of the CW in the EOT, or more generally of the CW in phenomena related to subwavelength metallic gratings.

2.2 Experiments

Figure 2.1 illustrates how we vary the hole density: we increase the distance between so-called hole chains, while keeping the properties of these chains fixed. The holes within the chains are spaced by $a_y = a_0 = 450$ nm. The distance between the chains is chosen to be $a_x = qa_0$, where $q=1, 2, 3, 4, 6$, and 7 . The holes perforate two metal layers: a 150 nm gold layer that is deposited on a glass substrate, and a 20 nm chromium layer that damps the SPP at the air interface. We measured the zeroth order transmission spectra of each array using x -polarized light, hence SPP propagating along the y -axis are negligible [10, 36, 49, 50].

Figure 2.2 shows the measured spectra on a semilog scale. Each array has a transmission peak between 735 and 775 nm, which shifts to shorter wavelengths as the hole spacing increases. The transmission minima of these resonances are at 724 ± 4 nm, which is very close to the expected value of $\text{Re } n_{\text{eff}} a_0 = 729$ nm, where n_{eff} is the ratio between the SPP wave vector and

Figure 2.2: Measured transmission spectra (solid curves) and the fitted surface plasmon model (red dashed curves). Each dashed curve has the same parameters except for the q -value. The parameters are based on a fit of Eq. (2.3) to the data of arrays with $q \geq 2$. The black dashed curve is a model prediction, plotting Eq. (2.3) with the same parameters as the other curves but with $q = 1$. The model accurately predicts all maxima and minima of the measured data for $q \geq 2$, showing that the transmission of these arrays is dominated by surface plasmons.



the free space wave vector [51, 52]. The magnitude of the transmission minima ranges between 0.6×10^{-4} and 2×10^{-4} . The resonance peaks are the result of a surface wave propagating in the x -direction, with the number of oscillations per lattice period equal to the q -value of the hole array. For the 4, 6, and $7a_0$ arrays additional resonances are also visible between 800 and 1000 nm. These resonances correspond to a surface wave propagating in the x -direction with $q-1$ oscillations per period.

As evidenced by the log-scale plots, the zeroth order transmission dramatically decreases as the hole density decreases. As will be justified hereafter, it essentially scales as $1/q^2$. Thus in Fig. 2.3, we plot $q^2 \times \text{transmission}$ on a linear scale, for $q = 1, 2, 4$, and 6. The results for $q = 3$ and $q = 7$ have been removed for clarity of the figure. By plotting the data on a linear scale we see that the scaled transmission is almost constant when going from $q = 2$ to $q = 6$. However for $q = 1$, the transmission peak is markedly larger, more than two times larger than all the other peaks. The fact that all the arrays except $q = 1$ virtually exhibit identical extraordinary transmission peaks is not fortuitous and, as will be interpreted hereafter, it reveals the resonant transmission due to SPPs only. With this respect, the sudden and marked increase of the peak transmission for the largest density ($q = 1$) constitutes an important result of the present work: it is a direct signature of the additional short-range contribution of the CW to EOT.

The previous conclusions are inferred qualitatively on the basis of the difference in damping lengths between SPP and CW. Helped by the microscopic SPP model in ref. [36], we will now aim at providing a fully quantitative analysis of the data in Figs. 2.2 and 2.3 to explicitly support our conclusions. The major drawback of the microscopic approach is that it requires the knowledge

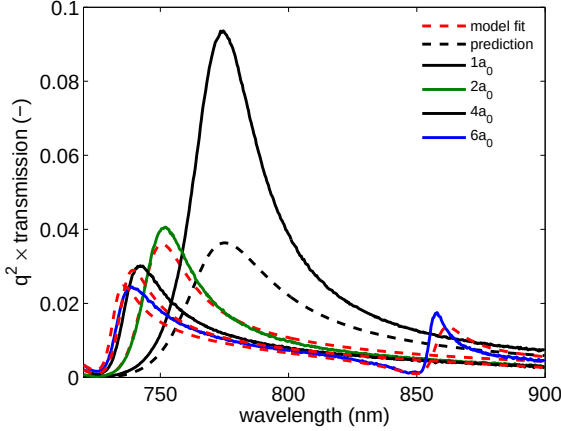


Figure 2.3: By plotting $q^2 \times \text{transmission}$, it is shown that there is a sudden increase of the transmission when comparing the $q = 1$ array to the other arrays. By plotting the SPP model too, we show that this behavior is not predicted by considering surface plasmons alone.

of many dispersive complex-valued parameters, which need to be calculated using full-vectorial simulations at every wavelength. This precludes its direct application to analyze experimental data. Hereafter, we revisit the model in depth and show that an analytical expression involving only five independent real non-dispersive fitted parameters may accurately reproduce the whole set of experimental spectra in Fig. 2.2 for $q \geq 2$, but produces wrong predictions for the case $q = 1$.

2.3 Simplifying the microscopic model

The microscopic model, as originally formulated in [36], considers one-dimensional hole chains (along the y -direction) as the elementary scatterers of the hole arrays and assumes that the electromagnetic interaction between the hole chains is mediated only by the SPP modes on unperturbed interfaces, the CW being totally neglected. The microscopic model couples three modes: the SPP mode at the gold-glass interface that consists of SPP propagating in both directions along the x -axis, the fundamental mode within the holes, and the free space modes.

The modes are coupled by the following scattering parameters, illustrated in Fig. 2.1b. An incident surface plasmon can: couple into the holes of the hole chain (α); couple to free space (β); or be transmitted (τ) or reflected (ρ). An incident plane wave can couple to a surface plasmon (β) or directly into the holes of the hole chain (t). The propagation of the surface plasmon from one hole chain to another is described by $e^{-ik_{\text{spp}}a_x}$, where $k_{\text{spp}} = 2\pi n_{\text{eff}}/\lambda$ is the wave vector of the SPP on a flat gold-glass interface and a_x is the distance between the hole chains. Because we kept the hole size constant for each array, the scattering parameters (α, β, ρ , and τ) should be the same for each array.

Only the hole spacing a_x depends on the parameter q , namely $a_x = qa_0$.

In line with the coupled mode approach proposed in [36] we derived (see appendix) the transmission coefficient t_f of a single hole chain, modified by the SPP excited at the surrounding hole chains:

$$t_f = t + \frac{2\alpha\beta}{e^{-ik_{\text{spp}}a_x} - (\rho + \tau)}. \quad (2.1)$$

The assumption that the hole chains only excite a plane SPP wave in the x -direction is valid if the incident polarization is along the x -axis. Moreover, the holes within the hole chain have to be less than a SPP wavelength apart [36]. The model neglects the influence of the metal-air interface. In the appendix we show this assumption holds if the surface plasmons at that interface are sufficiently damped by the chromium layer.

The optical transmission measured at a particular angle is a sum of the field contributions from all illuminated hole chains. If we define N_0 as the number of illuminated hole chains for the $q = 1$ array, $N = N_0/q$ is the number of hole chains as a function of q . In the zeroth diffraction order, the contributions from all hole chains are in phase, hence the transmitted intensity is [53]

$$T \propto 1/q^2 |t_f|^2, \quad (2.2)$$

where the proportionality factor is independent of q , as further discussed in the appendix.

Equations (2.1) and (2.2) are not yet simple enough to be used for modeling experimental data. Its six parameters are complex, so we have twelve real-valued parameters. To reduce the number of free parameters we combine both $\alpha\beta$ and $\rho + \tau$ into one complex parameter each. Furthermore, we need only the phase difference between t and $\alpha\beta$. For the complex variable k_{spp} we combine linearly interpolated literature values for the gold properties [54] with a measured value of the index of refraction of the substrate (1.51). This leaves us with a model of five free parameters.

Another crucial step towards fitting the model to the data is the wavelength dependence of the parameters. The following approximations are used: 1) $t \propto \lambda^{-2}$, as indicated by several studies [43, 55–57]; 2) $\alpha\beta \propto \lambda^{-4}$ in rough agreement with a factor 4 reduction for a wavelength increase of 38%, as reported in ref. [36]; 3) we keep $\rho + \tau$ constant, as calculations show that this sum has limited dispersion; 4) the phase of $\rho + \tau$ and the phase difference between $\alpha\beta$ and t are also kept constant with wavelength. So finally, we have the following fit equation:

$$T = q^{-2} \left| \frac{\lambda_0^2}{\lambda^2} p_1 + \frac{\lambda_0^4}{\lambda^4} \frac{p_2 e^{ip_3}}{e^{-ik_{\text{spp}}qa_0} - p_4 e^{ip_5}} \right|^2. \quad (2.3)$$

Parameter	Fit value	Error
p_1	0.0740	0.0006
p_2	0.0243	0.0006
p_3	0.681π	0.013π
p_4	0.857	0.009
p_5	0.132π	0.006π
$ e^{-ik_{\text{spp}}a_0} $	1.021	N.A.

Table 2.1: The values of the fitting parameters p_1 to p_5 . For completeness we also give the absolute value of $e^{-ik_{\text{spp}}a_0}$, evaluated at 800 nm. Also we give an estimate of the error in the fit values.

with $\lambda_0 = 800$ nm and p_1 to p_5 the five real-valued parameters in the model. The physical significance of parameters p_1 and p_4 is discussed in the appendix.

2.4 Fitting the microscopic model to the transmission spectra

The fitted curves obtained with the model are shown with the red dashed curves in Fig. 2.2. For the fit we only use the data of arrays $q = 2$ to $q = 7$, as we expect the transmission of these arrays to be dominated by SPP. To make sure that the minima are fitted well, we fitted $^{10}\log(\text{data})$ to $^{10}\log(\text{model})$. The result is plotted in Fig. 2.2 on a logarithmic scale, and the fit values are given in Table 2.1. For the arrays with $q=2, 3, 4, 6, 7$ the model describes the spectral positions of all minima and maxima very accurately, including the $(q-1, 0)$ resonances. Also the transmission magnitude is well modeled.

Remarkably too, Eq. (2.3) used with $q = 1$ (black dashed curve in Figs. 2.2 and 2.3) fails at predicting the experimental data for $q = 1$. In particular it underestimates the transmission peak by a factor 2.5. The latter is consistent with the theoretical predictions in [36], which conclude that SPP account for only half of the total transmitted energy at peak transmittance for a resonance wavelength of 700nm. In the Appendix, we expand the SPP microscopic model to incorporate the CW (see Eq. (2.7) therein), this extended model accurately describes the experimental data for $q = 1$ (see Fig. 2.6). Let us emphasize that this result is achieved using the exact same scattering parameters and does not require any additional fitting.

The observation that the transmission spectra of these six different hole arrays (including the particular $q = 1$ case) can be described with only five parameters is a big success of the microscopic theory. Moreover, it is of importance that these parameters are directly related to elementary scattering processes. Apparently the resonance of perforated metal surface can be modeled with a combination of a SPP mode and a CW on an unperturbed surface and a few extra scattering parameters, which can be inferred from simple transmission measurements.

2.5 Conclusion

We have presented the first experiments that quantitatively show the respective contributions of surface plasmons polaritons (SPP) and the quasi-cylindrical wave to extraordinary optical transmission (EOT). This in-depth analysis has been made possible by deriving the elementary scattering parameters defined in [36] from classical transmission measurements performed on a series of hole arrays. The possibility of making these microscopic scattering parameters experimentally accessible may be paramount for understanding and designing complex periodic or aperiodic metallic structures. Hence, variations to this modeling approach may have important implications in applying plasmonic structures to sensors, photovoltaics, LEDs or lasers.

2.6 Methods

The sample is fabricated as follows. Using e-beam lithography we created pillars on a glass substrate. Then we deposited 150 nm of gold and subsequently deposited the 20 nm chromium. Finally, we etched away the pillars. The function of the chromium layer is to heavily damp surface plasmons on the gold-air interface. We analyzed scanning electron microscope pictures of each array to conclude that the average hole radius is 81 ± 4 nm. By studying the position of the diffraction orders using a 635 nm laser diode, we concluded that the lattice parameters a_0 of all arrays are equal within 2 nm. Each array covers an area of $400 \mu\text{m} \times 400 \mu\text{m}$.

The transmission spectrum of each array is measured using the following setup (not shown). The light of a halogen lamp is filtered (longpass, 600 nm) and coupled into a $200 \mu\text{m}$ diameter fiber. The light coupled out of the fiber is polarized along the x -axis. The end facet of the fiber is imaged onto the sample with a magnification of 1.5. The $300 \mu\text{m}$ diameter spot is centered on the $400 \mu\text{m} \times 400 \mu\text{m}$ arrays. The zeroth order transmission is imaged ($M=2/3$) onto a $365 \mu\text{m}$ diameter fiber that leads to an Ocean Optics 2000+ USB spectrometer. To measure the reference spectrum, we move the sample out of the beam using a stage.

An accurate measurement of the transmission spectrum requires sufficient spatial and temporal coherence of the illuminating and detected light. The spatial coherence at the sample is ensured by the limited numerical aperture ($\text{NA} \approx (6 \pm 2) \times 10^{-3}$) of the illumination, which corresponds to a coherence length of a few tens of micrometers on the sample [53], being much larger than an SPP propagation length. The temporal coherence is ensured by the spectral resolution of the detecting spectrometer. The 1 nm resolution corresponds to coherence times of hundreds of femtoseconds [53] and is an order of magnitude better than strictly needed.

Appendix

In this appendix, we provide the equations of the SPP model [36] and of the SPP+CW model [45] that are used for fitting the experimental data. We also explain the fitting process in more detail.

For the 2D hole arrays considered in the experiment, the transmission coefficient t_F of the zeroth-order plane wave can be expressed with a Fabry-Perot equation [36],

$$t_F = \frac{t_A t'_A e^{ik_0 n_{FM} d}}{1 - r_A r'_A e^{i2k_0 n_{FM} d}} \quad (2.4)$$

for which we assume that the transmission of light is mediated by the least-attenuated fundamental mode of subwavelength holes. In Eq. (2.4), t_A and r_A (resp. t'_A and r'_A) are the transmission and reflection coefficients of the fundamental mode at the gold-glass (respectively chromium-air) interface, as defined in Fig. 2.4, n_{FM} is the complex effective index of the fundamental mode, d is the thickness of gold film and $k_0 = 2\pi/\lambda$. Then the zeroth-order power transmittance is $T = (n_g/n_a) |t_F|^2$, where $n_g = 1.5$ and $n_a = 1$ are the refractive indices of glass and air, respectively. In the absence of surface-wave-mediated resonance at the air interface, due to the damping chromium layer, t'_A and r'_A depend only weakly on the wavelength in the spectral range of interest (essentially fixed by the resonance of the glass interface), and $|r'_A| \ll 1$ [36, 58]. Thus Eq. (2.4) can be simplified as:

$$t_F = t_A t'_A e^{ik_0 n_{FM} d} \quad (2.5)$$

in which the sole resonance term is t_A . Within the scope of the pure-SPP model, which assumes that only SPPs are carrying energy between the hole

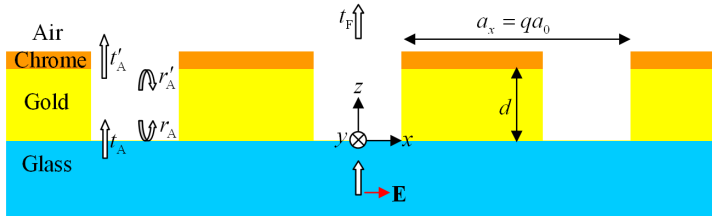


Figure 2.4: Scattering parameters t_F , t_A , t'_A , r_A and r'_A defined for a 2D hole array. The x -polarized plane wave (sketched by the vertical arrow in glass region) is normally incident from the glass side in the experiment. The array periods in x - and y -directions are qa_0 and a_0 respectively, and the gold film thickness is d . t_F is the transmission coefficient of the zeroth-order plane wave, t_A (resp. t'_A) is the transmission coefficient from the incident plane wave to the transmitted fundamental mode of the hole array at the gold-glass interface (resp. from the fundamental mode to the transmitted zeroth-order plane wave at the gold-chromium-air interface), and r_A (resp. r'_A) is the reflection coefficient of the fundamental mode at the gold-glass (resp. gold-chromium-air) interface.

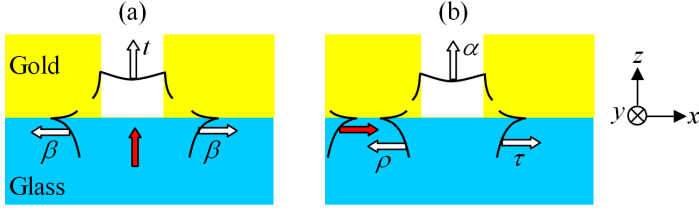


Figure 2.5: SPP-scattering parameters t, α, β, ρ and τ defined for a single y -periodic chain of holes. **a**, For an incident plane wave (sketched by the red arrow in glass region), t is the transmission coefficient from the plane wave to the fundamental hole-chain mode, and β is the SPP launching coefficient (sketched by the horizontal arrows). **b**, For an incident SPP (red arrow), ρ and τ are the reflection and transmission SPP coefficients, and α is the coupling coefficient to the fundamental hole-chain mode.

chains at the glass interface [36], t_A under normal incidence can be expressed as

$$t_A = t + \frac{2\alpha\beta}{u^{-1} - (\rho + \tau)}. \quad (2.6)$$

where $u = e^{ik_{\text{spp}}qa_0}$ is the phase shift experienced by the SPP in one period qa_0 ; t, α, β, ρ and τ are SPP-scattering parameters associated to the scattering of SPPs by individual hole chains (see Fig. 2.5 for a definition and [36] for more details), t corresponds to the direct transmission, β denotes the SPP launching coefficient, α describes the coupling from the excited SPP to the fundamental chain mode, and ρ and τ characterize the in-plane reflection and transmission of the SPP. The pure-SPP model and the associated Eq. (2.6) are used for fitting the transmission spectra and for obtaining the red dash-dotted curve in Fig. 2.2.

Recently, two of the authors have extended the pure-SPP coupled-wave model by incorporating the contribution of the CW to the transmission. Importantly, it is found that the scattering coefficients associated to the CW are (in the limit of small apertures) strictly identical to the SPP scattering coefficients α, β, ρ and τ , and that a coupled-wave model considering both the SPP and quasi-cylindrical wave can thus be straightforwardly deduced from the pure SPP model. Details concerning the derivation can be found in [45]. In the generalized SPP+CW (Hybrid Wave, or HW) model, which is considering the total field on the interface and which is therefore much more accurate, the refined "equivalent" of Eq. (2.6) is

$$t_A = t + \frac{2\alpha\beta}{(1/\sum H_{HW} + 1) - (\rho + \tau)}. \quad (2.7)$$

where $\sum H_{HW} = \sum H_{SPP} + H_{CW}$ combines a SPP part, $\sum H_{SPP} = 1/(u^{-1} -$

1), and a CW part, $\sum H_{CW}$, which represents a lattice summation of CW field at multiples of period qa_0 ($\sum H_{CW}$ is known analytically [45]).

As explained in the main text, with the SPP model the zeroth-order power transmittance T for our samples with periods qa_0 and a_0 can be expressed with a fitting-ready form deduced from Eq. (2.6),

$$T(q) = q^{-2} \left| t_{fit} + \frac{2\alpha_{fit}}{u(q)^{-1} - (\rho_{fit})} \right|^2. \quad (2.8)$$

where $t_{fit} = tt'_{A,fit}$, $\alpha_{fit} = 2\alpha\beta t'_{A,fit}$, $t'_{A,fit} = e^{ik_0 n_{FM} d} \sqrt{n_g/n_a} t'_A$. Equation (2.8) exhibits an analytical dependence of T with q , in which the complex fitted parameters t_{fit} , α_{fit} and ρ_{fit} are all independent of q , and $u^{-1}(q) = e^{ik_{spp}qa_0}$ is known. This enables a unified theoretical framework for analyzing samples with the same metal thickness, hole size and periodicity a_0 along the y -direction but with different periods in the x -direction (as done in Fig. 2.2 in the main text for $q=2-7$).

As indicated in the main text, the wavelength dependence of t_{fit} , α_{fit} and ρ_{fit} are adopted to be $t_{fit} = p_1(\lambda_0/\lambda)^2$, $\alpha_{fit} = p_2 e^{ip_3}(\lambda_0/\lambda)^4$ and $\rho_{fit} = p_4 e^{ip_5}$, where the real quantities p_1 to p_5 are independent of the wavelength. Thus on overall, the whole series of experimental spectra obtained for various q 's (this represents a large quantity of data) are fitted with only five real parameters. The drastic reduction in the parameter space not only evidences that the microscopic model captures the essence of the physical mechanism of the EOT, but also allows for a very robust fitting procedure.

For the SPP+CW model, Eq. (2.8) becomes,

$$T(q) = q^{-2} \left| t_{fit} + \frac{2\alpha_{fit}}{1/\sum H_{HW}(q) - (\rho_{fit})} \right|^2. \quad (2.9)$$

where $H_{HW}(q)$ depends on q in a known analytical way [45]. Therefore once the unknown quantities t_{fit} , α_{fit} and ρ_{fit} are determined by the previous fitting procedure, the prediction of the SPP+CW model is immediately computed with the fitted quantities; no additional fitting or numerical calculations are required. In Fig. 2.6, we show the results of the SPP+CW model for $q = 1$ (see the red sold curves). On the logarithmic scale the deviations between the model and the data are similar to that of the other samples for $q = 2 - 7$. On the linear scale it is clearly seen that the CW boosts the transmission by more than a factor two. The small deviations between experiment and the SPP model for $q = 2 - 7$ or the SPP+CW model for $q = 1$ are probably due to the many approximations we had to make to derive our fit expression.

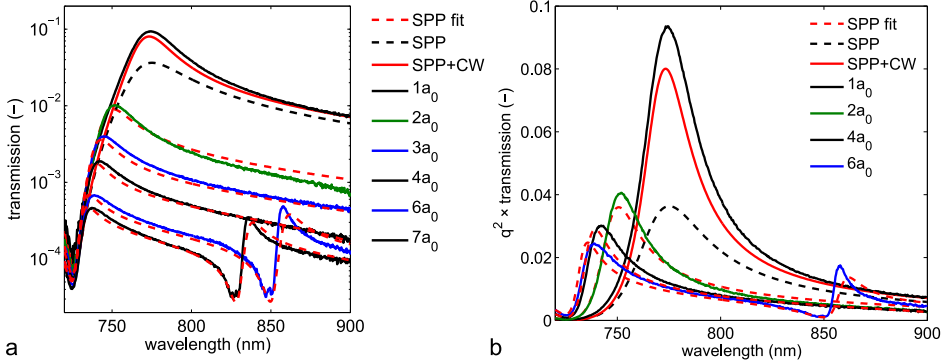


Figure 2.6: Results of the SPP model and the SPP+CW model using the fit parameters from the SPP model. **a**, On a logarithmic scale it is seen that the SPP+CW model fits the $q = 1$ data for well over three orders of magnitude. **b**, By plotting $q^2 \times \text{transmission}$ on a linear scale, we show that the predicted transmission for the SPP+CW model is more than two times larger than for the SPP model. This shows that the CW explains the discrepancy between the SPP model and the experimental data.

Also, we fit the data on a logarithmic scale, and therefore the fit routine allows some deviations at the maximum transmission, in favor for a better fit of the minima. We believe that the remarkable agreement obtained for $q = 1$ in Fig. 2.6 between the experimental data and the SPP+CW model and not for the pure-SPP model constitutes a critical proof of the embodiment of the quasi-cylindrical wave in the EOT.

Finally, we would like to discuss the physical significance of two of our fit parameters. The parameter p_1 allows us to calculate the transmission enhancement and suppression, as this parameter characterizes the light that is transmitted without surface wave resonance. Given the fitted $p_1^2 = 5.5 \times 10^{-3}$, the maximum transmission of 0.093 for $q = 1$ is 17 times larger than this value, while the minimum of 8.5×10^{-5} is 65 times smaller. For the $q = 6$ sample, the enhancement and suppression are reduced because of SPP absorption and the virtual absence of the CW. The enhancement is now 4.4, while suppression is reduced to a factor 3.3.

A second interesting fit parameter is $p_4 = |\rho + \tau|$. This parameter quantifies the scattering losses of the SPP wave at the hole chains. Its fit value of $p_4 = 0.857$ should be compared with the modulus of $e^{ik_{\text{spp}}a_0} = 0.98$ around the resonance wavelength. This comparison shows that the SPP scattering losses are roughly 7 times larger than the absorption losses for our densest ($q = 1$) sample, while they are approximately equal for the $q = 7$ low-density sample.

Transmission processes in random patterns of subwavelength holes

The optical transmission of random patterns of holes is believed to depend on the transmission of the independent holes only. By comparing the transmission spectra of random patterns with different hole densities, we show that the quasi-cylindrical wave plays an important role in the transmission of samples with a large hole densities. Furthermore we report on a speckle pattern seen in the transmission of these hole patterns. By studying the degree of depolarization in this speckle pattern, as a function of hole density, we are able to quantify the role of surface plasmons to the transmission.

F. van Beijnum, C. R  tif, C. B. Smiet, and M. P. van Exter, *Opt. Lett.* **36**, 3666 (2011).

3.1 Introduction

Research on the optical properties of holes in metal films has long been subject of fundamental and applied research. In 1944 Bethe calculated the transmission of a circular hole in a thin perfectly conducting film [55]. In 1998, the discovery of the extraordinary optical transmission of metal hole arrays [28] revived the interest in the optical properties of subwavelength holes and plasmonics. The ability of holes to couple light from free space to surface plasmons (SPP), makes them important for future applications [59, 60].

Calculating the transmission properties of subwavelength holes in metal films has proven to be a challenge [43]. The field at the metal surface turns out to be more complex than anticipated: not only SPP are excited at holes and slits, but also the quasi-cylindrical wave (CW) at a short distance from the hole, and the Norton wave at large distances [44–46].

So far, the transmission of random patterns of subwavelength holes were believed to accurately represent the transmission of a single hole [56, 61]. In this chapter we will argue that both the CW and SPP also contribute to the transmission of random patterns of subwavelength holes. To reveal these transmission processes we compare random patterns of different hole densities.

Figure 3.1a shows a scanning electron microscope (SEM) picture of a typical structure. We studied two sets of seven samples of which the inverse density (area per hole) is chosen qa_0^2 with $a_0 = 450$ nm and $q = 1, 2, 3, 4, 9, 16, 25$. One set has circular holes (average diameter: 120 ± 6 nm) and the other has square holes (average side length: 125 ± 5 nm). To avoid proximity effects in the fabrication the holes have a minimum side to side distance of 50 nm. Figure 3.1b illustrates the layer structure: a glass substrate with 150 nm gold, and 20 nm of chromium. The arrows illustrate direct (dashed) and indirect (dash-dotted) transmission processes that we study.

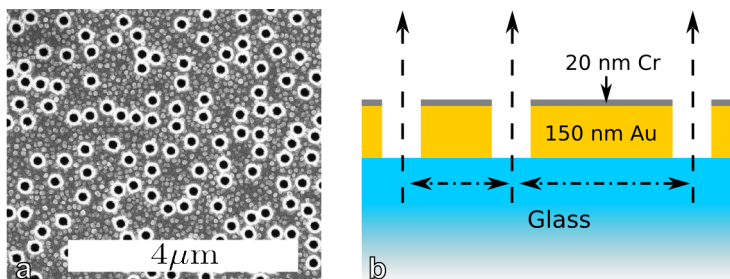


Figure 3.1: **a**, A SEM picture of one of the studied samples. The holes in a metal film are positioned randomly, samples with various hole densities are made. **b**, The layer structure of the metal film. The arrows illustrate direct (dashed) and indirect (dash-dotted) transmission processes that we study.

3.2 Modeling different transmission processes

Before presenting the experiments, we calculate the contributions of the transmission processes to the zeroth order transmission. We separate the transmission in a direct and an indirect contribution (see arrows Fig. 3.1b), and first include only SPP for the indirect transmission. We define $t_{d,i}$ to be the directly transmitted field for hole i , which is i -independent for identical holes. In the zeroth order the directly transmitted fields are all in phase, and therefore interfere constructively. The SPP field at hole i is a sum over the contributions from all neighboring holes j : $t_{s,i} = \sum_j t_{s,ij}$. As each indirect contribution $t_{s,ij}$ picks up a random phase, due to the random hole positioning, the amplitudes $t_{s,i}$ are uncorrelated. Hence the transmitted intensity T in the zeroth order diffraction is:

$$T = \left| \sum_{i=1}^N t_{d,i} + t_{s,i} \right|^2, \\ \lim_{k_{\text{spp}} \gg \alpha} \langle T \rangle = N^2 |t_d|^2 + N \langle |t_s|^2 \rangle, \quad (3.1)$$

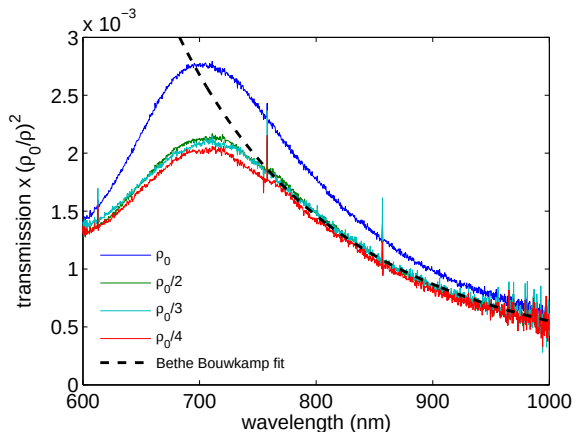
where $\langle \cdot \rangle$ denotes ensemble averaging, N is the number of holes, k_{spp} is the SPP wave vector and α is the inverse propagation length. In the limit $k_{\text{spp}} \gg \alpha$ there is effectively no correlation between amplitude and phase of $t_{s,i}$ and therefore the crossterms $t_{s,i} t_{s,j}^*$, $t_d t_{s,j}^*$, and $t_{s,i} t_d^*$ cancel out for $i \neq j$. In our experiments $N \gg 1$, making the SPP contribution negligible in the zeroth order.

In contrast to the SPP contribution just discussed, we expect the amplitude and phase of the CW to be correlated because this contribution decays rapidly, even within a wavelength. Hence the crossterms do not average out and an extra contribution proportional to N^2 is expected. However, this contribution will only be found if the average hole spacing is sufficiently small. Thus if we compare patterns with different hole density, the ratio $\langle T \rangle / N^2$ should change if the hole density is sufficiently large.

3.3 Recorded transmission spectra for different hole densities

We measure the zeroth order transmission of our samples using a standard white light transmission spectroscopy setup (not shown). The light from a halogen lamp is filtered (longpass, 600 nm) and coupled into a 200 μm fiber. The end facet of this fiber is imaged onto the sample with a magnification of 1.5. The transmitted light is imaged ($M=2/3$) onto a second fiber that leads to an Ocean Optics 2000+ USB spectrometer. The small NA ($\text{NA}=(6 \pm 2) \cdot 10^{-3}$) of the detection optics singles out the zeroth order transmission.

Figure 3.2: Intensity transmission of the four densest samples, normalized by $(\rho/\rho_0)^2$. The normalized transmission of the densest sample is enhanced as a result of the quasi-cylindrical wave.



To see an effect of the CW the transmission spectra are scaled with $(\rho_0/\rho)^2$, with $\rho_0 = 1/(0.45 \mu\text{m})^2$. For the low hole densities, the transmission of the structure approaches that of the gold itself ($T_{\text{gold}} \approx 3 \cdot 10^{-6}$). Hence, we need to correct for the transmission of the unperforated gold. Since both contributions are coherent we had to assume a phase relation to account for the interference, we choose the contributions to be in phase. This correction works for hole densities larger than $\rho_0/9$.

Figure 3.2 shows the scaled spectra for circular holes. The spectra for $\rho_0/2$, $\rho_0/3$, and $\rho_0/4$ overlap within 5%. In contrast, the scaled transmission of the densest sample at 685 nm is $37 \pm 1\%$ larger than that of the scaled transmission of the other three samples. This enhancement decreases gradually to $11 \pm 2\%$ at 600 nm, and to $18 \pm 5\%$ at 900 nm. For the square holes a similar but somewhat larger enhancement is found. We attribute the increase of the scaled transmission of the densest sample to the CW contribution discussed above.

Besides the hole density dependence of the spectra, the wavelength dependence of the transmission has attracted much interest too [55, 56, 62]. The long wavelength tail of the transmission can be fitted using the Bethe-Bouwkamp formula [55, 63], (see e.g. [43]). The fit results in a diameter of 130 nm, which is surprisingly close to the diameter of 120 ± 6 nm measured in the SEM pictures.

A remaining issue is whether the maximum in the transmission is a shape resonance. The transmission maximum appears to be dependent on the hole size, since transmission maxima of the (larger) square holes are at a larger wavelength (750 nm). In calculations and experiments on perfect electric conductor films such a size dependent transmission maximum is found too [43, 62, 64, 65]. In ref. [43] it is shown that a maximum is expected at

$kr \approx 1.6$ for zero thickness and at $kr \approx 2.1$ for a thickness equal to the radius. However, in our experiments a maximum is found at $kr = 0.8$, hence the origin of this feature is not yet resolved.

3.4 Polarization analysis of far field speckle pattern

We not only observe the zeroth order diffraction peak, but also a speckle pattern (order 10^{-3} , see Fig. 3.3a). If we illuminate the sample with polarized laser light, and place an analyzing polarizer, we see that the zeroth order can be suppressed while the speckle pattern remains visible (see Fig. 3.3 b). The speckle pattern intensity for the orthogonal polarization is roughly an order of magnitude smaller than that of the parallel polarization. Moreover the pattern has changed (only 2% correlation in speckle patterns). To the best of our knowledge, this speckle pattern has never been reported.

Before analyzing the speckle patterns in further detail, we calculate the contributions to the speckle intensity at an angle $\vec{\theta}$. In contrast to the analysis of the zeroth order, the contributions of the direct transmission will not be in phase. Two holes at position $\vec{r}_i$ and $\vec{r}_j$ will have a phase difference $\vec{k}_{||}(\vec{r}_i - \vec{r}_j)$. Hence, the transmitted intensity in the speckle pattern is:

$$\langle T(\theta) \rangle = N \left(|t_d|^2 + \langle |t_s|^2 \rangle \right). \quad (3.2)$$

In contrast to the zeroth order transmission, both contributions are now incoherent and therefore both scale with N . However, one would expect that the direct contribution only has the incident polarization, whereas the SPP contribution is partially depolarized.

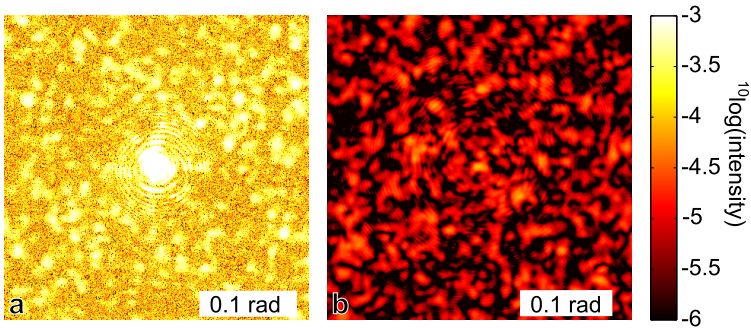


Figure 3.3: Angular transmission pattern of the sample of with $\rho/\rho_0 = 16$ illuminated with a weakly focussed beam. **a**, Using an analyzing polarizer parallel to the incident polarization, we see a zeroth order diffraction peak and a speckle pattern. **b**, By rotating the polarizer 90° the zeroth order is suppressed. All intensities are normalized to the maximum intensity of the zeroth order in **a**.

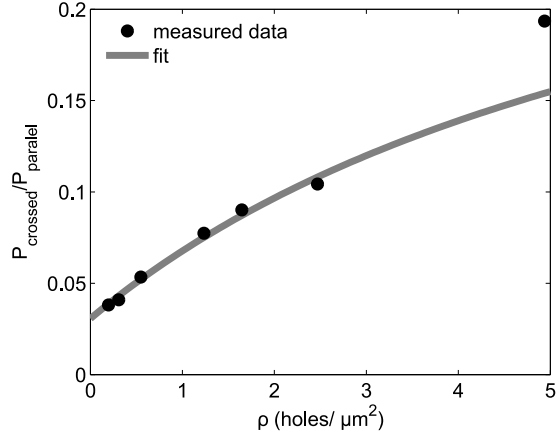


Figure 3.4: The ratio of the power in the speckle pattern for parallel over the orthogonal polarization. The ratio increases with hole density, showing that there is an indirect contribution.

Using a simple model we calculate the SPP contribution at hole i from all other holes j using: $t_{s,i} = \sum_j t_{s,ij}$. We assume that the light propagates along a straight line from hole j to i . The field will be damped due to absorption and light that is scattered out of the surface plasmon mode, making $t_{s,ij} = (C(\phi)/\sqrt{r_{ij}}) \exp((\alpha + ik_{spp})r_{ij})$. The excitation efficiency $C(\phi)$ is a function of the angle ϕ between the incident polarization and the propagation direction, and has unit $\sqrt{\text{m}}$. The loss rate due to scattering is denoted as α .

The exact form of $C(\phi)$ is unknown, but we can approximate it using a projection argument: $C(\phi) \propto \cos \phi$. Besides the excitation efficiency, there is a detection efficiency. Using a polarizer parallel or orthogonal to the incident polarization, this results in an extra factor of $\cos \phi$ or $\sin \phi$ respectively. Hence, the fraction of the average power in the surface modes for the parallel polarization ($C_{\parallel}^2 \propto \langle \cos^4 \phi \rangle$) is three times larger than in the orthogonal polarization ($C_{\perp}^2 \propto \langle \cos^2 \phi \sin^2 \phi \rangle$). The ratio between the power in the speckle pattern for the parallel and orthogonal polarization is thus:

$$\frac{P_{\perp}}{P_{\parallel}} = \frac{|t_{d,\perp}|^2 + \rho C_{\perp}^2/(2\alpha)}{|t_{d,\parallel}|^2 + \rho 3C_{\perp}^2/(2\alpha)}, \quad (3.3)$$

where $|t_{d,\perp}|^2$ is the depolarized part of the direct transmission. Thus at low hole densities, one measures the depolarization due to imperfections of the holes. As the hole density increases the relative amount of depolarized light will increase, due to the increased outcoupling of SPP.

For all seven samples we measured the ratio between the power in the speckle pattern for the parallel and orthogonal polarization. In Fig. 3.4 this ratio is plotted versus the hole density. The measured ratios increase with hole

density from 0.03 to 0.2. The fact that the depolarization is density dependent proves the existence of an indirect contribution. When fitting the data to the theory (line in Fig. 3.4), we see that the data point at the largest hole density can not be fitted properly. However we expect the CW to contribute to both the zeroth order and the speckle pattern, because the CW contribution is random but with a correlation between amplitude and phase.

From the fit we conclude that $C_{||}^2/(2\alpha |t_{d,||}|^2) = 0.14 \pm 0.02 \mu\text{m}^2$. This means that for a hole density of $1 \mu\text{m}^{-2}$ and parallel polarizers the SPP contribution is 14% of the direct transmission. From the fit we find: $|t_{d,\perp}|^2 = 0.031 \pm 0.005$, meaning that 3% of the directly transmitted light is depolarized by the holes. By imaging a low density sample, we observe that the transmission through individual holes can be blocked within experimental accuracy using a polarizer. However the blocking angle varies from hole to hole, ranging from -10° to 10° , which in rough agreement with the 3% found in Fig. 3.4.

3.5 Conclusion

We have shown that random patterns of subwavelength holes of variable hole densities are an ideal tool to unravel transmission processes. In the zeroth order transmission the direct transmission and quasi-cylindrical wave (CW) are the only relevant contributions. By analyzing a newly reported speckle pattern for different polarization states, we have quantified the SPP contribution to the transmission.

In future work the enhancement as a result of the CW can possibly be predicted using a recent theoretical calculation of the CW contribution near a single hole [46]. Moreover, the analysis of the speckle pattern could be done for different materials or wavelength ranges, to study the importance of the SPP and the CW there.

Speckle correlation functions applied to surface plasmons

The optical intensity transmitted through a random pattern of subwavelength holes in a metal film exhibits a speckle pattern. We study the variation of this speckle pattern as a function of wavelength. We find that the resulting speckle correlation function (SCF) separates in a wavelength dependent part and a wavelength independent background. The wavelength dependence is caused by surface plasmons excited at one hole and coupled out at another hole, while the constant background originates from light transmitted directly through the holes. By analyzing the SCF for a set of samples of varying hole density, we find the propagation length of the surface plasmons and the scattering losses induced by the holes.

F. van Beijnum, J. Sirre, C. R  tif, and M. P. van Exter, Phys. Rev. B **85**, 35437 (2012).

4.1 Introduction

Structuring materials on a scale comparable or smaller than the wavelength of light allows control over their optical properties in an unprecedented way. Famous examples are photonic crystals, metal hole arrays, and metamaterials. As the complexity of these structures increases, it becomes more challenging to understand their physics. For example, to quantitatively model the extraordinary optical transmission (EOT) [28] of metal hole arrays a large set of parameters is required that need to be calculated numerically [45]. Also the transmission of light through a single hole in a metal film is surprisingly complex [43].

In contrast, disordered media can be characterized relatively easy. By studying the variations of speckle as a function of wavelength or angle, speckle correlation functions (SCF) can be calculated analytically [66]. These functions, that can also be measured [67–71], provide insight in the dwell time inside a medium. Only three quantities are needed to describe the SCF in volume scattering: the transport mean free path, the energy velocity [72], and the sample thickness.

In this chapter we apply the framework of SCFs to surface plasmons. To this extent, we study the optical transmission through random patterns of subwavelength holes. These random patterns are previously studied using a broadband source [56, 73], but when illuminated with laser light the transmitted intensity exhibits a speckle pattern [57]. The intensity in this speckle pattern has two contributions: light that is directly transmitted through the holes and light transmitted via surface waves. By studying the change of these speckle patterns as we scan the wavelength of the laser we can record a SCF. Because of the two different contributions to the speckle intensity, we expect that these correlation functions are different from the ones found in three dimensional random media.

From the SCF we hope to infer a propagation length. As it is unclear whether the losses are dominated by absorption or scattering, we study a set of samples with different hole densities. We analyzed a set of nine samples for which the hole density is $1/(qa_0^2)$, where $a_0 = 450$ nm and $q = 1, 2, 3, 4, 9, 16, 25, 36, 81$. Each sample covers a square area of $400\text{ }\mu\text{m} \times 400\text{ }\mu\text{m}$. The average side length of these square holes is 125 ± 5 nm. The layer structure is as follows: on the glass substrate we subsequently deposited 150 nm gold and 20 nm of chromium, and we then perforated this metal layer. The function of the chromium layer is to damp the surface plasmons on the gold-air interface, limiting the analysis to one interface. Figure 4.1 shows scanning electron microscope images of three of the studied samples, with hole densi-

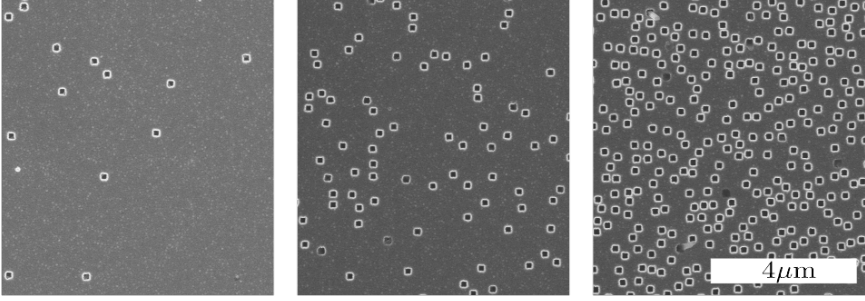


Figure 4.1: Scanning electron microscope images of three of the nine studied samples. From left to right the hole density is $0.19 \mu\text{m}^{-2}$, $1.2 \mu\text{m}^{-2}$, and $4.9 \mu\text{m}^{-2}$

ties of $0.19 \mu\text{m}^{-2}$, $1.2 \mu\text{m}^{-2}$, $4.9 \mu\text{m}^{-2}$, that is $q = 25$, $q = 4$ and $q = 1$ respectively.

4.2 Theory

Before showing the experimental results, we will first derive an expression for the expected correlation function. In the far field the speckle intensity is a function of wavelength and angle: $I(\lambda, \vec{\theta})$. We define the speckle correlation function as follows:

$$C \equiv \frac{\langle I(\lambda_0, \vec{\theta}_0) I(\lambda_1, \vec{\theta}_1) \rangle}{\langle I(\lambda_0, \vec{\theta}_0) \rangle \langle I(\lambda_1, \vec{\theta}_1) \rangle} - 1, \quad (4.1)$$

where $\langle \cdot \rangle$ denotes ensemble averaging [66]. We can rewrite the intensity correlation function in terms of the optical fields $E(\lambda, \vec{\theta})$, using Isserlis's theorem [74] for Gaussian random variables [75]:

$$C = \frac{\left| \langle E(\lambda_0, \vec{\theta}_0) E^*(\lambda_1, \vec{\theta}_1) \rangle \right|^2}{\langle I(\lambda_0, \vec{\theta}_0) \rangle \langle I(\lambda_1, \vec{\theta}_1) \rangle}, \quad (4.2)$$

where $*$ is the complex conjugate.

When measuring in the far field, at an angle $\vec{\theta}$, the light transmitted through the randomly positioned holes is a sum of all illuminated holes E_i . Depending on the position $\vec{x}_i$, the field from each hole acquires a random phase: $\vec{k}_{||}(\lambda) \vec{x}_i$, with $|\vec{k}_{||}(\lambda)| = 2\pi \sin(|\vec{\theta}|) / \lambda$. Hence the correlation is:

$$C \propto \left| \sum_{i, i'} \langle E_i(\lambda_0) E_{i'}^*(\lambda_1) e^{i[\vec{k}_{||}(\lambda_0) \vec{x}_i - \vec{k}_{||}(\lambda_1) \vec{x}_{i'}]} \rangle \right|^2, \quad (4.3)$$

where the proportionality sign indicates that the normalization is now omitted. Calculations can be performed with this expression, but the decorrelation from $e^{i[\vec{k}_{||}(\lambda_0)\vec{x}_i - \vec{k}_{||}(\lambda_1)\vec{x}_{i'}]}$ may have a strong influence on the correlation function [76]. Hence, we take $\vec{k}_{||}(\lambda_0) = \vec{k}_{||}(\lambda_1)$, such that the exponent becomes one for $\vec{x}_i = \vec{x}_{i'}$. The phase of this term is random when $\vec{x}_i \neq \vec{x}_{i'}$ and $\vec{k}_{||}$ is sufficiently large, i.e. if we are outside the zeroth diffraction order [57]. Hence, after ensemble averaging the terms with $i \neq i'$ are zero, and therefore the double sum over i and i' , reduces to a single sum.

The field E_i is a sum of a direct contribution $E_{d,i}$ and a contribution via surface waves $E_{s,i}$. The field $E_{s,i}$ consists of contributions from all neighboring holes. Hence the amplitude and phase are random, and depend on the position of the surrounding holes. Although the phase is random, it changes gradually as the excitation wavelength changes. We can now evaluate Eq. (4.3) further by inserting $E_i(\lambda) = E_{d,i} + E_{s,i}(\lambda)$, and taking the ensemble average:

$$C = \frac{\left| \left\langle |E_d|^2 \right\rangle + \left\langle E_s(\lambda_0) E_s^*(\lambda_1) \right\rangle \right|^2}{\langle I_{tot} \rangle^2}, \quad (4.4)$$

with $\langle I_{tot} \rangle = \langle I_d \rangle + \langle I_s \rangle$, where $\langle I_d \rangle$ and $\langle I_s \rangle$ are assumed to be wavelength independent for the clarity of the expressions. Please note that the summation and the i -dependence disappear because the summation is replaced by the number of holes times the ensemble average. Also the ensemble average of the cross terms is zero, $\left\langle E_{d,i} E_{s,i}^* + E_{d,i}^* E_{s,i} \right\rangle = 0$, because $E_{s,i}$ and $E_{d,i}$ are independent and the ensemble average of $E_{s,i}$ is zero. Equation 4.4 is an essential result: the direct transmission is observable as a constant background correlation, while the contribution from surface waves does decorrelate in a limited wavelength range.

Next we calculate the wavelength dependent correlation caused by the surface waves. For simplicity, we neglect contributions from the quasi-cylindrical wave [36] and Norton wave [44–46], and only consider surface plasmons (SPP). We consider a particular hole i , and write the total SPP field as a sum of contributions from all the surrounding holes j . We describe the SPP propagation as a two dimensional surface wave. As it propagates it can be absorbed or scattered out by a hole such that:

$$E_{s,i} = \sum_j \frac{A_0(\phi_j)}{\sqrt{r_{ij}}} e^{(-\sigma\rho - \text{Im } k_{\text{spp}} + i \text{Re } k_{\text{spp}})r_{ij}}, \quad (4.5)$$

where $A_0(\phi)$ is a prefactor describing the excitation and outcoupling of the surface plasmon field, it has unit $V/\sqrt{m}$ and depends on the angle ϕ between the

incident polarization and the propagation direction [10, 57]; r_{ij} is the distance between hole i and hole j ; $\text{Im } k_{\text{spp}}$ characterizes the loss due to absorption; $\sigma\rho$ characterizes the loss due to scattering; σ is the scattering cross section and ρ the hole density. We are dealing with a two dimensional problem, and therefore the scattering cross section is a length, instead of an area.

When evaluating $\langle E_s(\lambda_0)E_s^*(\lambda_1) \rangle$ a double sum is found, for which the cross terms associated with interference originating from different holes $j \neq j'$ again average out. We rewrite the phase difference of the remaining terms as: $r_{ij}/\lambda_0 - r_{ij}/\lambda_1 = r_{ij}\Delta\lambda/(\lambda_0\lambda_1)$, as in ref. [75]. We then form concentric rings around hole i to find that the number of holes in each ring scales with the radius r . On the other hand the contribution from these holes scales as $1/r$ multiplied by an exponential, see Eq. (4.4). By replacing the sum over neighboring holes for an integral we find:

$$C = \frac{1}{\langle I_{\text{tot}} \rangle^2} \left| \langle I_d \rangle + \frac{\langle I_s \rangle \tilde{\lambda}}{i\Delta\lambda + \tilde{\lambda}} \right|^2. \quad (4.6)$$

The width of the Lorentzian,

$$\tilde{\lambda} = (\sigma\rho + \text{Im } k_{\text{spp}}) \frac{2\lambda_0}{\text{Re } k_{\text{spp}}}, \quad (4.7)$$

combines the scattering losses ($\propto \sigma\rho$) and the ohmic losses ($\propto \text{Im } k_{\text{spp}}$). The relative strength is determined by the ratio $\langle I_s \rangle / \langle I_d \rangle$, where $\langle I_s \rangle$ is given by:

$$\langle I_s \rangle \propto \frac{\rho \langle |A_0(\phi)|^2 \rangle_\phi}{\sigma\rho + \text{Im } k_{\text{spp}}}, \quad (4.8)$$

where the proportionality sign indicates that we omitted constant prefactors.

To summarize, we expect that we can measure a correlation function with a background that is independent of wavelength. Besides this background correlation, a wavelength dependent part is expected, caused by SPP propagating on the metal surface. The width of this wavelength dependent part is proportional to the losses that the SPP experiences. The hole density is a crucial parameter for the correlation function, it determines both the weight of the SPP part and the total loss that the SPP experience.

4.3 Experiments

The experimental setup is as follows: the light of a wavelength tunable Coherent 899 Ti-Sapphire laser is led through a single-mode fiber, and collimated to a beam with a diameter of a few millimeters. This beam is first polarized

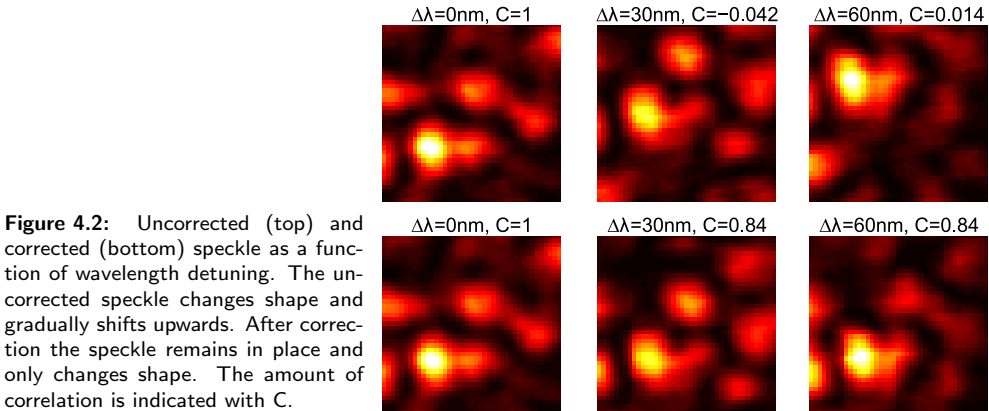


Figure 4.2: Uncorrected (top) and corrected (bottom) speckle as a function of wavelength detuning. The uncorrected speckle changes shape and gradually shifts upwards. After correction the speckle remains in place and only changes shape. The amount of correlation is indicated with C .

and then illuminates a $200\ \mu\text{m}$ pinhole that is imaged onto the sample with a magnification $3/8$, leaving a $75\ \mu\text{m}$ spot on the sample. This illuminated pinhole ensures that the size and position of the spot on the sample is wavelength independent. The light transmitted through the sample is collected using an aspheric lens ($f = 8\text{mm}$). The Fourier plane of this lens is imaged with a lens onto an intermediate plane and this intermediate plane is subsequently imaged onto a CCD. In the intermediate plane we block the zeroth order transmission with a black metal rod and we select the polarization parallel to the incident polarization. In all our experiments we scan the laser wavelength from $740\ \text{nm}$ to $810\ \text{nm}$.

We now first study the speckle that we see on the CCD. To derive Eq. (4.4), it was crucial to keep $\vec{k}_{||}$ constant with wavelength. Experimentally, this is achieved by rescaling the recorded images. In Fig. 4.2 we show the original and rescaled images as a function of wavelength, for a sample of hole density $1.2\ \mu\text{m}^2$. The top three images are shown as they are originally recorded. The zeroth order peak is located below the shown speckle. As the wavelength is tuned, the speckle moves up and changes.

In the bottom row the three images are corrected such that $\vec{k}_{||}(\lambda)$ is constant. For these images the speckle pattern changes gradually, but the speckle remains at approximately the same position. The effect on the correction is illustrated by the amount of correlation between a reference and the shown images. For the original images the correlation decreases rapidly from 1 to -0.042 and 0.014 , for the rescaled images the correlation is 1, 0.84 and 0.84 .

For rescaling the images we need to choose a point in the image for which $\vec{k}_{||}(\lambda) = 0$. Although we expected this point to be the zeroth order, we noticed that the correlation between two images recorded at different wavelengths is a few percent larger when we choose this point outside the zeroth order. The

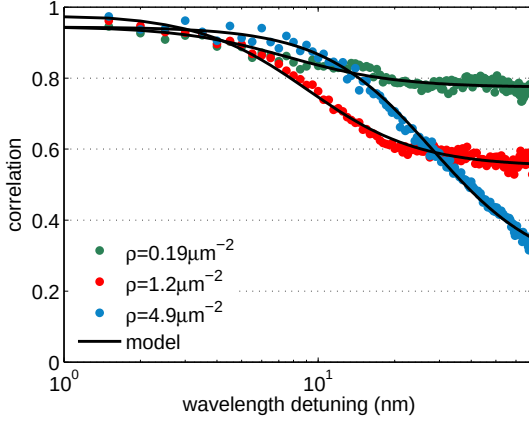


Figure 4.3: Measured correlation functions for three different hole densities. Each function combines a Lorentzian part with a wavelength independent background. With increasing hole density the Lorentzian becomes wider, while the background decreases.

origin of this effect has not been resolved so far. For our data processing we use the optimum point outside the zeroth order.

Now that we know how to rescale the images, we can measure the speckle correlation functions. Figure 4.3 shows three typical correlation functions, for the samples with hole densities $4.9 \mu\text{m}^2$, $1.2 \mu\text{m}^2$, and $0.19 \mu\text{m}^2$. The correlation function changes drastically with the hole density. Notwithstanding, it is shown that the correlation functions exhibit the predicted behavior: a wavelength dependent contribution and a constant background correlation. With increasing hole density, the background level decreases while the width of the wavelength dependent part increases. The decreasing background level is due to the increasing SPP contribution, as our model suggests. The increasing width implies shorter propagation lengths. This may be expected, as the scattering losses are proportional to the hole density.

To quantify our findings we have fitted the measured correlation functions to Eq. (4.6). Figure 4.3 shows that the theory describes the data well. The observation that our simple model describes the data might come as a surprise, as we omitted the wavelength dependence of both $\langle I_d \rangle$ and $\langle I_s \rangle$. Especially for $\langle I_d \rangle$ it is well known that it has a strong wavelength dependence, see e.g. [43] for a recent review. There are two ways to interpret our observation of a constant background correlation. First, because we normalize the recorded speckle patterns, see Eq. (4.1), a constant background correlation will also be found if the ratio $\langle I_s \rangle / \langle I_d \rangle$ is constant. So far, there is not much literature reported on the wavelength dependence of $\langle I_s \rangle$, but work on hole chains suggests that this is possible [36]. Second, the normalization makes the effect of the wavelength dependence smaller, as not only the nominator of the

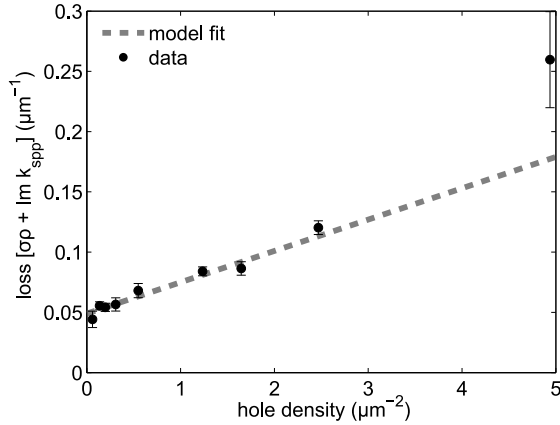


Figure 4.4: The loss plotted as a function of hole density. For all samples except the most dense, the loss increases linear with hole density. The loss at zero hole density is the surface surface plasmon absorption loss, that corresponds roughly with literature.

correlation changes but also the denominator. For a typical example, with $\langle I_s \rangle / \langle I_d \rangle = 1/4$ and a reduction of $\langle I_d \rangle$ to 70% of its original value, the background decreases to 62% instead of 67% for the constant background. To see this small decay, the scan range would have to be much larger. Finally, we also have an experimental argument for why the wavelength dependence of $\langle I_d \rangle$ and $\langle I_s \rangle$ are not relevant. If we scan in the opposite direction, namely 810 nm to 740 nm, instead of increasing the wavelength, we find practically the same correlation $C(\Delta\lambda)$.

4.4 Extracted fit parameters

Two fit parameters are extracted from the fit, the losses ($\sigma\rho + \text{Im } k_{\text{spp}}$) and the ratio $\langle I_s \rangle / \langle I_d \rangle$. As seen in Fig. 4.4 the loss increases linearly with hole density, as expected from Eq. (4.7). However, the loss of the highest hole density sample is larger than expected from a linear dependence. For this data point the error margin is also larger than the other data points, because the background level could not be fitted properly as the correlation still decreases within our scan range.

We fitted these loss values with a linear function, from which we find an offset of $0.049 \pm 0.002 \mu\text{m}^{-1}$ and the slope $\sigma = 26 \pm 2 \text{ nm}$. The cross section is reasonable, considering the hole side length of $125 \pm 5 \text{ nm}$. The offset corresponds reasonably to the values that can be calculated using the dielectric constant of gold at 740 nm reported by Johnson and Christy [77] ($0.056 \mu\text{m}^{-1}$) and Palik [54] ($0.065 \mu\text{m}^{-1}$).

In Fig. 4.5 the ratio $\langle I_s \rangle / \langle I_d \rangle$ is plotted versus hole density. We observe a nonlinear increase. Equation 4.8 shows that when $\sigma\rho$ is comparable to $\text{Im } k_{\text{spp}}$,

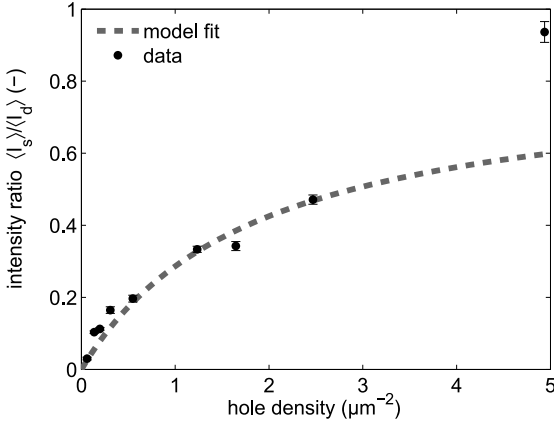


Figure 4.5: The surface plasmon contribution $\langle I_s \rangle$ relative to the direct contribution $\langle I_d \rangle$ increases non linearly with hole density. Using only the vertical scale as a free parameter, the data can be reasonably fitted to theory.

this nonlinear increase is expected. We fitted Eq. (4.8) to this data, using the values of $\text{Im } k_{\text{spp}}$ and σ we just obtained, leaving only the vertical scale as a free parameter. The fit result is reasonable. Interestingly, both the intensities at low hole density and at the largest density are underestimated by the fit.

Nevertheless, from fitting the intensity ratio we conclude that the scattering cross section found in Fig. 4.4, fits the data reasonably well. The high intensity for the most dense sample can imply an extra contribution to the field, that may be caused by the quasi-cylindrical wave [36, 45, 46]. However, to be able to make a sensible judgement about this, the correlation function should be studied theoretically, including and excluding the quasi-cylindrical wave. Moreover, it would be of great value to use a scanning laser with a larger range, to measure the full correlation function for this sample.

Another interesting question that arises from this data, is whether the underestimation of the low hole density is related to the Norton Wave [44–46]. At the distance probed with these low densities, roughly five propagation lengths ($\rho\sigma = \frac{1}{5} \text{Im } k_{\text{spp}}$), the Norton wave should have the same amplitude as the SPP. However, at these distances the field is only $7 \cdot 10^{-3}$ of its original strength and we wonder if these small fields contribute to the decorrelation seen. On the other hand, the Norton Wave becomes more than three orders of magnitude larger than the SPP field, for a distance of roughly ten propagation lengths [46].

Besides this set of random patterns with square holes, we have also analyzed a set of patterns of circular holes, with a diameter of 120 ± 6 nm. For these samples the signal is smaller than for square holes, and hence we could only measure the five most dense samples. The measured correlation functions

show a larger background correlation, when comparing patterns with the same hole density. From a similar analysis as for the square holes we found a cross section of 18 ± 1 nm and an absorption loss of 0.035 ± 0.002 . μm^{-1} . The loss of the most dense sample is now not as high as found for the square holes, and all found loss values fit the SPP model.

4.5 Conclusion and discussion

We have derived a simple expression for the speckle correlation function for random patterns of subwavelength holes. The expression has two contributions, a constant background resulting from the direct transmission of the holes, and a wavelength dependent part due to surface plasmons propagating on the surface. The predicted behavior of a constant background and wavelength dependent part is also seen in the measured correlation function. By fitting the experimental results, we find that the propagation length of the surface plasmon decreases with increasing hole density, as the surface plasmon has a larger probability of being scattered out by the holes. This measurement yields the scattering cross section of the holes. Moreover the surface plasmon contribution increases with hole density, as more holes are available to excite surface plasmons. The results for the most dense sample are not consistent with the other samples, which implies that the quasi-cylindrical wave contribution might be visible in correlation functions.

We believe the application of speckle correlation functions to plasmonics can be very valuable. The experiments give insight in both the surface plasmon excitation and outcoupling by the holes. Moreover, it may be possible to study the influence of the quasi-cylindrical wave on the correlation functions, in theory and experiments. Also, the combination of a constant background and a wavelength dependent part is new compared to 3D random media [69]. Furthermore it is interesting that two different transmission processes can be separated using speckle correlation functions. It would be interesting to see if similar behavior is found when the 3D samples are made sufficiently thin compared to the transport mean free path, or when these media are modified such that very short paths through the medium exist [78].

Rayleigh scattering of surface plasmons by a subwavelength hole extracted from wavelength dependence of speckle patterns

Rayleigh scattering of light is well known for being inversely proportional to the fourth power of the wavelength, but so far it is unclear whether this scaling also applies to the scattering of surface plasmons at a subwavelength hole. We extract the scattering cross section of a surface plasmon scattering at a single hole from the transmission of random patterns of subwavelength holes. The measured scattering cross section for surface plasmon scattering at a single hole has a stronger wavelength dependence than the traditional λ^{-4} scaling found for small particles. Although this experimentally found scaling is consistent with recent theoretical work, the magnitude of the scattering cross section is about an order of magnitude larger than predicted.

F. van Beijnum, A. S. Meeussen C. R  tif, and M. P. van Exter, submitted for publication.

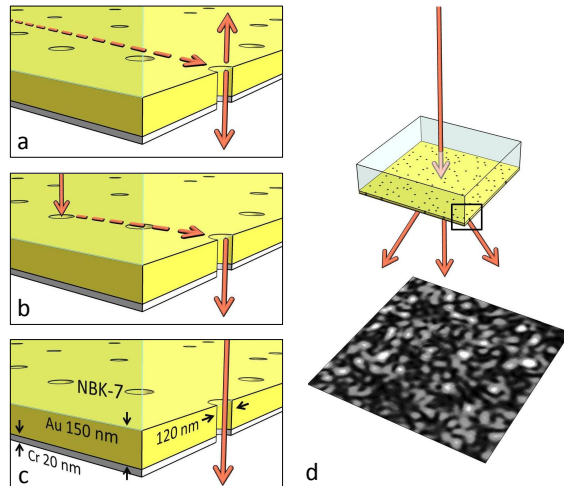
5.1 Introduction

Subwavelength holes are important building blocks for novel photonic structures, given that these holes are used in metamaterials [4, 79], photonic crystal slabs [80], sensors [29] and possibly thin film solar cells [81]. In the context of the extraordinary optical transmission [28], the transmission of light through single subwavelength holes in metal films has attracted much interest and its physics is surprisingly rich [43, 55, 82].

The excitation [10, 46] and scattering [83, 84] of surface plasmons by single subwavelength holes has been studied both theoretically and experimentally. The wavelength dependence of these scattering processes might reveal the underlying physics of surface plasmon scattering. Also, deep understanding of these scattering events is of paramount importance for recently developed microscopic models [36, 37]. So far, this wavelength dependence is only studied using metal hole arrays. One study reports the traditional [85] λ^{-4} dependence [86], while another study reports a λ^{-n} wavelength dependence where the power n depends strongly on hole size [87]. Both experimental observations contradict theories on surface plasmon scattering [83, 84].

For surface plasmons scattered at a single hole, the scattering cross section has unit length instead of an area [83, 84]. This is because the cross section is the scattered power divided by the incident power per unit width of the surface plasmon mode. This width is along the surface and perpendicular to the propagation direction [83, 84, 88]. Using the power per unit width has the advantage that it is independent of the surface plasmon mode size.

Figure 5.1: a-c, These experiments probe three scattering processes: **a**, the coupling of a surface plasmon to free space via a single hole; **b**, surface plasmon mediated transmission, where first a surface plasmon is excited at one hole and transmitted at another hole; **c**, direct transmission through a subwavelength hole. **d**, Random patterns of subwavelength holes are illuminated by a spectrally filtered supercontinuum laser source, of which we scan the wavelength. The change of the speckle pattern as a function of wavelength difference $\Delta\lambda$ is quantified by calculating the correlation $C(\Delta\lambda)$.



Because the scattering cross section for surface plasmons has unit length, the traditional expression [85] of a product of a volume squared and λ^{-4} can not be correct.

In this chapter we extract Rayleigh scattering of surface plasmons by single subwavelength holes from the transmission of random patterns of these holes. An important advantage of these random patterns is that most interference effects can be averaged, in contrast to the transmission of arrays which is entirely dominated by interference effects. In random patterns it is also straightforward to compare samples of different hole densities, which allows separating the ohmic and radiative losses of the surface plasmons [38].

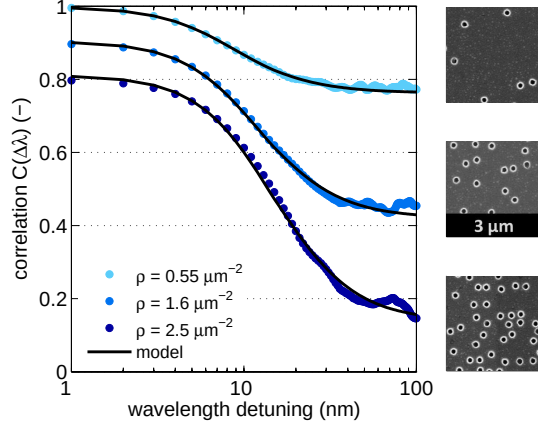
The results presented in this chapter revolve around three quantities, of which we measure the wavelength dependence: the surface plasmon absorption length L_{abs} , the scattering cross section σ , and the intensity ratio cross section A . The surface plasmon absorption length L_{abs} contains only the ohmic loss of the surface plasmons. The scattering cross section σ characterizes the radiative loss of a surface plasmon at a single hole (Fig. 5.1a). The intensity ratio cross section A describes the transmission of light via a surface plasmon where first a surface plasmon is excited and thereafter transmitted through the hole (Fig. 5.1b). This parameter A contains a normalization to the direct transmission (Fig. 5.1c). Before presenting the wavelength dependence of L_{abs} , σ and A , we show how we extract these quantities from the transmission of random hole patterns. Our approach is discussed in more detail in ref. [38].

5.2 Experiment

Our experiments are performed on a series of random patterns of subwavelength holes in a metal film. The series contains eight patterns of which only the hole density is varied. We choose the area per hole to be qa_0^2 , with $a_0 = 0.45 \mu\text{m}$ and $q \in [1, 2, 3, 4, 9, 16, 25, 36]$. The circular holes (diameter of $120 \pm 6 \text{ nm}$) perforate a 150 nm thick gold film which is deposited directly on glass, omitting the commonly used adhesion layer. A subsequently deposited 20 nm chromium layer damps the surface plasmons on the gold-air interface, allowing us to selectively study surface plasmons on the gold-glass interface (see Fig. 5.1c).

We illuminate these random patterns of subwavelength holes with monochromatic light and record the far field speckle intensity $I(\vec{\theta}, \lambda)$ (see Fig. 5.1d). The change of the speckle pattern with wavelength can be quantified by calculating the correlation between the measured speckle intensity at wavelengths λ_0 and $\lambda_1 = \lambda_0 + \Delta\lambda$, resulting in a correlation function $C(\Delta\lambda)$ [66, 68, 70, 72, 89, 90]. We perform these measurements in a large wavelength range using a supercontinuum laser source (Fianium Whitelase 400SC)

Figure 5.2: The measured correlation functions $C(\Delta\lambda)$ have a wavelength-dependent contribution caused by surface plasmons propagating on the gold-glass interface, and a wavelength-independent contribution resulting from light that is directly transmitted through the holes. The correlation function depends strongly on hole density: the width increases with hole density while the background decreases. For the clarity of the figure, the plots for $\rho = 1.6 \mu\text{m}^2$ and $\rho = 2.5 \mu\text{m}^2$ are offset by -0.1 and -0.2 respectively.



of which we select a narrow line (~ 1 nm) with a spectrometer.

Using a simple model, which assumes that only surface plasmons are excited at the holes and thus neglects the quasi-cylindrical wave contribution [35, 44, 47], we find an analytic expression for the correlation function:

$$C(\Delta\lambda) = \frac{1}{\langle I_d + I_s \rangle^2} \left| \langle I_d \rangle + \frac{\langle I_s \rangle}{1 - iL_{\text{tot}} \text{Re}[\Delta k_{\text{spp}}]} \right|^2. \quad (5.1)$$

Equation (5.1) contains two density dependent parameters: L_{tot} , the propagation distance of the surface plasmons which includes both radiative and nonradiative losses; $\langle I_s \rangle / \langle I_d \rangle$, which defines the intensity ratio between light transmitted via surface plasmons ($\langle I_s \rangle$) and directly through the holes ($\langle I_d \rangle$). The term $\text{Re}[\Delta k_{\text{spp}}]$ is the difference between the surface plasmon momenta at wavelengths λ_0 and λ_1 . To good approximation Eq. (5.1) is a Lorentzian with an almost wavelength independent background correlation $\langle I_d \rangle^2 / \langle I_d + I_s \rangle^2$.

Figure 5.2 shows three examples of measured correlation functions (on a log-linear scale) for three different hole densities. The scans in this plot are performed from 690 nm ($\Delta\lambda = 0$ nm) to 790 nm ($\Delta\lambda = 100$ nm). With increasing hole density the background correlation (i.e. at large $\Delta\lambda$) decreases while the spectral width of the correlation increases. The observation that the background correlation decreases shows that the efficiency of transmission via surface plasmons increases with hole density as a larger fraction of the excited surface plasmons is coupled out instead of being absorbed. This increase in outcoupling is also evidenced by the increasing spectral width, which is directly related to the losses of the surface plasmons.

The three fits in Fig. 5.2 are based on Eq. (5.1) and in good correspondence with the data. From each fit two density dependent parameters can

be extracted: L_{tot} and $\langle I_s \rangle / \langle I_d \rangle$. The density dependence of L_{tot} can be quantified in terms of two density-independent parameters:

$$L_{\text{tot}}^{-1} = L_{\text{abs}}^{-1} + \rho\sigma, \quad (5.2)$$

where L_{abs} is the surface plasmon absorption length in the absence of the holes and σ is a scattering cross section that describes the radiative loss of a surface plasmon at a single hole (see Fig. 5.1c). Because of the two dimensional nature of our system, the hole density ρ has unit per area and the scattering cross section σ has unit length. In the appendix we show that we can fit Eq. (5.2) to the measured density dependence of L_{tot} . This fit yields two density-independent parameters that apply to all structures: the absorption length of the surface plasmons L_{abs} and the scattering cross section σ at a single hole.

The second parameter that we obtain from the correlation functions is the intensity ratio $\langle I_s \rangle / \langle I_d \rangle$. In the appendix the experimentally obtained ratios are presented as a function of hole density. Using our model, we can express this intensity ratio in terms of the hole density ρ [38]:

$$\frac{\langle I_s \rangle}{\langle I_d \rangle} = \frac{A\rho}{\rho\sigma + L_{\text{abs}}^{-1}}, \quad (5.3)$$

where A is a third density-independent parameter: the intensity ratio cross section. Equation (5.3) fits the experimental data of the density-dependent intensity ratio, using only A as a free parameter (see appendix).

This parameter A comprises two different effects: first, the excitation of surface plasmons from free space at the glass side; second, the outcoupling to the air side (see Fig. 5.1b). The magnitude of A contains a normalization to the intensity transmitted directly through the hole (see Fig. 5.1c). The parameter A has unit length, which makes the right hand side of Eq. (5.3) dimensionless, as is the ratio on the left hand side.

To summarize, we can generalize the correlation functions $C(\Delta\lambda)$ of samples with different hole densities, using only three density-independent parameters: L_{abs} , σ and A . We measure the correlation functions for different values of the reference wavelength λ_0 , allowing us to measure the wavelength dependence of the parameters L_{abs} , σ and A . In particular, we try to understand the wavelength dependence of the scattering parameters σ and A using Rayleigh scattering of surface plasmons at single holes as microscopic model.

5.3 Results

In Fig. 5.3 we show the measured wavelength dependence of L_{abs} . The absorption length increases by approximately a factor four from $L_{\text{abs}} \approx 5 \mu\text{m}$ to

$L_{\text{abs}} \approx 20 \mu\text{m}$, when the wavelength is increased from 650 nm to 950 nm. The data matches very well with the theory for which we use literature values of the refractive index of gold [54, 77]. This correspondence is very important as it demonstrates the validity of our approach, both qualitative and quantitative.

In Fig. 5.4 we plot the extracted value for the scattering cross section σ as a function of wavelength. This scattering cross section shows a steep decline from slightly more than 100 nm at a wavelength of 675 nm to around 15 nm at 875 nm. This decline is significantly steeper than the traditional expression for Rayleigh scattering ($\sigma \propto \lambda^{-4}$) which is indicated by the dashed line.

Recently, an analytic expression is derived for the scattering cross section of surface plasmons scattered at a subwavelength hole [84]. For surface plasmons scattered to the photon field this expression is:

$$\sigma = \xi \frac{k^4 a^6}{d_{\text{spp}}} \quad (5.4)$$

where a is the hole radius, k is the wave vector in air, and d_{spp} is the mode size of the surface plasmon, i.e. the $1/e$ width of the intensity tail into the dielectric. The dimensionless proportionality constant ξ is radius independent for $ka \ll 1$. For a hole in a perfect electrical conductor slab of zero thickness $\xi = 0.24$. Hence, the expression is essentially equivalent to that for scattering of light by three dimensional particles, with the exception that the surface plasmon mode size comes in as a proportionality factor. This factor indicates that the hole is polarized more effectively when the surface plasmon mode is more compact. The wavelength dependent mode size of a surface plasmon at a metal-air interface is $d_{\text{spp}} \approx \sqrt{|\epsilon|}/(2k)$, with ϵ the dielectric constant of the metal (assuming $|\epsilon| \gg 1$).

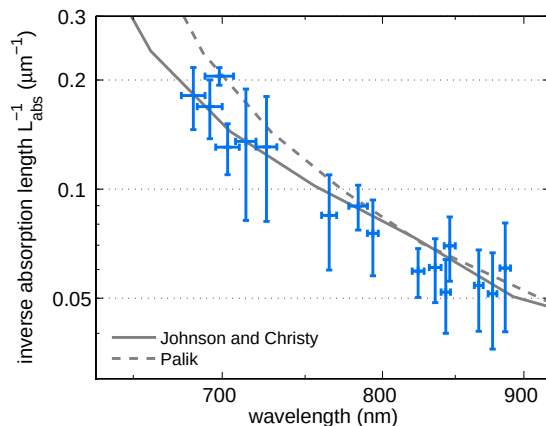


Figure 5.3: Inverse absorption length L_{abs}^{-1} as a function of wavelength, as extracted from our experiments. The obtained absorption length is in good agreement with theory showing both the validity of our experiment and the quality of the gold layer.

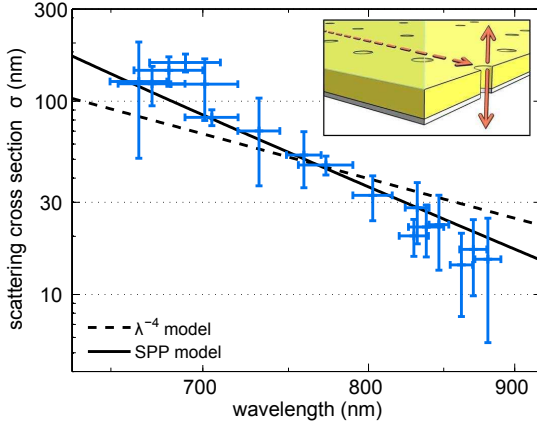


Figure 5.4: The scattering cross section σ , describing the radiative loss of a surface plasmon at a single hole, decreases almost a factor 10 in the measured wavelength range.

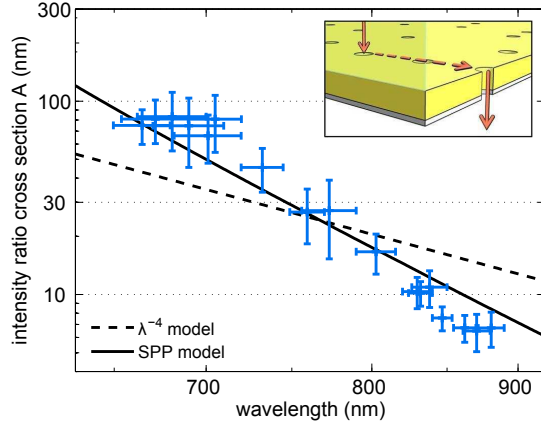
Fitting Eq. (5.4) to the data in Fig. 5.4, we see a much better correspondence than for $\sigma \propto \lambda^{-4}$. This is a very important result, as it shows that the wavelength dependence of the surface plasmon scattering can be understood and described well using a simple expression. The wavelength dependence of surface plasmon scattering at subwavelength holes can apparently be understood by combining Rayleigh scattering with the surface plasmon mode size.

The prefactor predicted by the theory [84] ($\xi = 0.24$) is roughly two orders of magnitude different from our results ($\xi = 63 \pm 27$), whereas the experiments published with the theory agree within a factor two [84]. The theory, however, is derived for a metal-air interface. By adapting the theory to a metal-glass interface, we find that Eq. (5.4) should be multiplied by $n^6 = 11.9$, with n the refractive index of glass (see appendix), thereby increasing the theoretical expectation to 2.8. In the appendix we speculate that the remaining order of magnitude can be explained by the field penetration into the metal, which is neglected when a perfect electrical conductor is assumed.

Given the promising results for the scattering cross section, we may also be able to understand the wavelength dependence of the intensity ratio cross section A . In Fig. 5.5 we plot the extracted value for A as a function of wavelength: A spans roughly an order of magnitude and is of comparable magnitude as σ , suggesting that A and σ may be related. Similar to σ , A has a stronger wavelength dependence than λ^{-4} .

In the appendix we derive a relation between A and σ , which is $A = \sigma \eta 3\lambda / (n 16 d_{spp})$. The efficiency η describes, for an incident surface plasmon, how much power is radiated to the substrate relative to the total power scattered out at this hole. The factor $3\lambda / (n 16 d_{spp})$ describes, for a magnetic

Figure 5.5: The intensity ratio cross section, A , represents transmission of light via a surface plasmon, that is excitation at one hole and transmission at another hole. This parameter also decreases almost a factor 10 in the measured wavelength range and is comparable in magnitude to the scattering cross section σ .



dipole, how much power is radiated to surface plasmons relative to the power radiated to the free space at the substrate side. This calculation assumes that both the surface plasmon excitation and outcoupling are mediated via the same (magnetic) dipole moment.

In Fig. 5.5 we plot a fit of $A = \sigma\eta 3\lambda/(n16d_{spp})$, using η as free parameter and the value of ξ we obtained from Fig. 5.4 which describes σ . We obtain a fitted value of $\eta = 0.67 \pm 0.19$, which is reasonable as we expect this efficiency to be close to, but smaller than, one. This demonstrates the consistency of the experimental data and the data analysis. We are able to relate two independent quantities (the intensity ratio and the spectral width) to the same scattering cross section σ using a simple efficiency factor.

These measurements are also performed on square holes with side length 125 ± 5 nm, showing the same wavelength dependencies for both σ and A (see appendix). For these square holes, the experimentally obtained values for ξ and η are very similar to those of circular holes.

5.4 Conclusions

The scattering cross section of surface plasmons scattered by a subwavelength hole is measured in the wavelength range of 650-900 nm. The reported wavelength dependence is stronger than Rayleigh scattering predicts, because a surface plasmon polarizes the hole less efficiently at larger wavelengths. Nonetheless, this behavior can be captured in a simple expression.

Additionally, the measured scattering cross section explains the ratio between surface plasmon mediated transmission and direct transmission of random hole patterns. Our results therefore imply that it may be viable to model particular complex plasmonic structures, like metal hole arrays, using only

physical parameters like the hole size, hole density and film thickness. However, first the magnitude of the measured scattering cross section needs to be understood as it is one order of magnitude larger than recent theoretical predictions.

The presented methodology of obtaining scattering cross sections from transmission measurements on samples of different hole densities is surprisingly powerful, and may prove to be fruitful outside plasmonics too. Moreover, we showed the advantage of using random patterns instead of arrays, as the randomness allows measurements at virtually any wavelength without changing the illumination angle and thus the character of the excited dipole moments.

Appendix

This appendix consists of four parts. We first present a derivation of the model used to fit our experimental correlation functions. Second, the parameters in our model are shown to be related the relevant magnetic and electric polarizabilities. In the third part we show the experimentally obtained density dependence of the propagation length and the intensity ratio, from which we obtained density independent parameters. In the last part, we present our results for square holes, presenting the wavelength dependence of the scattering cross section and of the intensity ratio cross section.

Model

In our experiments we calculate the correlation between two far-field speckle patterns $I(\lambda_0, \vec{\theta}_0)$ and $I(\lambda_1, \vec{\theta}_1)$, and express it as an experimental speckle correlation function (SCF). We have fitted the measured correlation function to a model containing two experimental parameters. In this section we show how we derived this model.

We calculate the correlation between two speckle patterns, which are both normalized to their mean intensity. Assuming the experimental correlation function is well described by an ensemble average, the correlation is expressed as follows:

$$C(\lambda_0, \lambda_1) \equiv \frac{\langle I(\lambda_0, \vec{\theta}_0) I(\lambda_1, \vec{\theta}_1) \rangle}{\langle I(\lambda_0, \vec{\theta}_0) \rangle \langle I(\lambda_1, \vec{\theta}_1) \rangle} - 1, \quad (5.5)$$

where $\langle \cdot \rangle$ denotes ensemble averaging. Using Isserlis theorem [74] Eq. (5.5) can be rewritten in terms of the electric fields $E(\lambda_0, \vec{\theta}_0)$ and $E(\lambda_1, \vec{\theta}_1)$, which simplifies further calculations:

$$C(\lambda_0, \lambda_1) = \frac{\left| \langle E(\lambda_0, \vec{\theta}_0) E^*(\lambda_1, \vec{\theta}_1) \rangle \right|^2}{\langle I(\lambda_0, \vec{\theta}_0) \rangle \langle I(\lambda_1, \vec{\theta}_1) \rangle}, \quad (5.6)$$

where $*$ denotes the complex conjugate. The field at a particular angle $\vec{\theta}$, is a summation of the fields at all individual holes i multiplied by a phase factor $\exp \left[i \vec{x}_i \vec{k}_{||}(\lambda) \right]$ that depends on the position of the hole $\vec{x}_i$, where $\left| \vec{k}_{||}(\lambda_0) \right| = (2\pi/\lambda) \sin \left| \vec{\theta} \right|$. Hence the total field $E(\lambda, \vec{\theta})$ is:

$$E(\lambda, \vec{\theta}) = \sum_i E_i(\lambda) \exp \left[i \vec{x}_i \vec{k}_{||}(\lambda) \right], \quad (5.7)$$

Inserting Eq. (5.7) into Eq. (5.6) results in a double summation (over i and i') of which the terms with $i \neq i'$ have an ensemble average of zero. For the terms with $i = i'$ we find $\left\langle E_i(\lambda_0) E_i^*(\lambda_1) \exp \left[i x_i (\vec{k}_{||}(\lambda_0) - \vec{k}_{||}(\lambda_1)) \right] \right\rangle$. This reduces to $\langle E_i(\lambda_0) E_i^*(\lambda_1) \rangle$ if we choose $\vec{k}_{||}(\lambda_0) = \vec{k}_{||}(\lambda_1)$. This condition can be achieved experimentally by rescaling the recorded images [38]. Hence our expression for the correlation function is now:

$$C(\lambda_0, \lambda_1) = \frac{|\langle E_i(\lambda_0) E_i^*(\lambda_1) \rangle|^2}{\langle I_i(\lambda_0) \rangle \langle I_i(\lambda_1) \rangle}, \quad (5.8)$$

This expression shows that we only have to consider the field at a single hole i , because the ensemble-averaged contribution of each hole is identical. Therefore we will now continue to derive an expression for the field at a single hole. We assume that this field has two contributions: a directly transmitted field $E_{d,i}(\lambda)$ and a surface plasmon field $E_{s,i}(\lambda)$, thereby neglecting the possible influence of the quasi-cylindrical wave [35, 44, 47] which is found at distances of roughly a wavelength from the hole. Inserting $E_i(\lambda) = E_{d,i}(\lambda) + E_{s,i}(\lambda)$ into Eq. (5.8) we obtain:

$$C \propto |\langle E_{d,i}(\lambda_0) E_{d,i}^*(\lambda_1) \rangle + \langle E_{s,i}(\lambda_0) E_{s,i}^*(\lambda_1) \rangle|^2. \quad (5.9)$$

To find this expression we use $\langle E_{d,i}^*(\lambda_1) E_{s,i}(\lambda_0) + E_{d,i}(\lambda_0) E_{s,i}^*(\lambda_1) \rangle = 0$, resulting from the random phase of the surface plasmon field.

We can specify $E_{s,i}(\lambda)$ further, assuming that the surface plasmons are radiated with an angular dependence $A_0(\lambda, \phi_j)$ from another hole j . These surface plasmons decay as a cylindrical wave ($1/\sqrt{r_{ij}}$) combined with an exponential decay due to radiative and nonradiative losses, given by the following expression:

$$E_{s,i}(\lambda) = \sum_j \frac{A_0(\lambda, \phi_j)}{\sqrt{r_{ij}}} \exp \left[-\frac{1}{2} L_{\text{tot}}^{-1}(\lambda) r_{ij} + i \text{Re } k_{\text{spp}}(\lambda) r_{ij} \right], \quad (5.10)$$

with $L_{\text{tot}}^{-1}(\lambda) = \rho\sigma(\lambda) + 2 \text{Im } k_{\text{spp}}(\lambda)$ the total propagation length, which has a contribution from radiative decay ($\rho\sigma$) and absorption $L_{\text{abs}}^{-1} = 2 \text{Im } k_{\text{spp}}(\lambda)$. Please note that the amplitude cross section used in previous work [38] is a factor two smaller than the (more common) intensity cross section used here. If we calculating the product $E_{s,i}(\lambda_0) E_{s,i}^*(\lambda_1)$ we find:

$$E_{s,i}(\lambda_0) E_{s,i}^*(\lambda_1) = \sum_j \frac{A_0(\lambda_0, \phi_j) A_0^*(\lambda_1, \phi_j)}{r_{ij}} \times \exp \left[-\frac{1}{2} (L_{\text{tot}}^{-1}(\lambda_0) + L_{\text{tot}}^{-1}(\lambda_1)) r_{ij} + i \text{Re } [\Delta k_{\text{spp}}] r_{ij} \right], \quad (5.11)$$

where:

$$\text{Re} [\Delta k_{\text{spp}}] = \text{Re} k_{\text{spp}}(\lambda_0) - \text{Re} k_{\text{spp}}(\lambda_1) = \frac{2\pi}{\lambda_0 \lambda_1} (\Delta \lambda n_{\text{eff}}(\lambda_0) - \Delta n_{\text{eff}} \lambda_1) \quad (5.12)$$

with $\Delta \lambda = \lambda_1 - \lambda_0$ and $\Delta n_{\text{eff}} = n_{\text{eff}}(\lambda_1) - n_{\text{eff}}(\lambda_0)$. For $\Delta n \lambda_0 \ll \Delta \lambda n_{\text{eff}}(\lambda_0)$ this is expressed as:

$$\text{Re} k_{\text{spp}}(\lambda_0) - \text{Re} k_{\text{spp}}(\lambda_1) = 2\pi \left(\frac{\Delta \lambda n_{\text{eff}}(\lambda_0)}{\lambda_0 \lambda_1} \right) \quad (5.13)$$

In case of an ensemble average, we can replace the summation in Eq. (5.12) with an integral using the average number of holes in an infinitesimal area: $\rho r dr d\phi$. This integral over a complex exponential results in what is essentially a complex Lorentzian. The resulting correlation function is:

$$C = \frac{1}{\langle I_d + I_s \rangle^2} \left| \langle I_d \rangle + \frac{\langle I_s \rangle}{1 - i L_{\text{tot}} \text{Re} [\Delta k_{\text{spp}}]} \right|^2, \quad (5.14)$$

where:

$$L_{\text{tot}}^{-1} = \frac{1}{2} (L_{\text{tot}}^{-1}(\lambda_0) + L_{\text{tot}}^{-1}(\lambda_1)), \quad (5.15)$$

$$\langle I_d \rangle = \langle E_d(\lambda_0) E_d^*(\lambda_1) \rangle, \quad (5.16)$$

$$\langle I_s \rangle = \rho L_{\text{tot}} \langle A_0(\lambda_0, \phi) A_0^*(\lambda_1, \phi) \rangle_\phi, \quad (5.17)$$

where $\langle \cdot \rangle_\phi = \int_0^{2\pi} \langle \cdot \rangle d\phi$. The factor A in the main manuscript is:

$$A \equiv \frac{\langle A_0(\lambda_0, \phi) A_0^*(\lambda_1, \phi) \rangle_\phi}{\langle E_d(\lambda_0) E_d^*(\lambda_1) \rangle} = \frac{\langle I_s \rangle}{\langle I_d \rangle} (\rho L_{\text{tot}})^{-1}. \quad (5.18)$$

To fit Eq. (5.14) we assume that $\langle I_d \rangle / \langle I_s \rangle$ and L_{tot} are wavelength independent within the scan range. Hence these fitted values are an average value over the wavelength range of interest. In the main manuscript it is shown that, although some approximations had to be made, the fit function works very well.

Relating model parameters to polarizability

Recent work has calculated the scattering cross section of the hole in terms of its polarizability [84]. This work shows that there are two relevant dipole moments in the surface plasmon scattering problem: a electric dipole oriented normal to the surface and a magnetic dipole oriented parallel to the surface.

In this section we will briefly discuss this calculation, trying to clarify the assumptions made. Hereafter we will try to relate the calculated scattering cross section to the intensity ratio cross section A extracted from our measurements.

The calculation assumes a surface plasmon on a metal dielectric interface, of which the power per length $P/L_\perp$ is calculated, analogous to the intensity in three dimensions. Using the induced dipole moments, the authors calculate the power radiated to free space P_{out} and to the surface plasmon field P_{spp} . When calculating this radiation, a closed film (i.e. without a hole) is assumed.

By dividing these radiated powers by $P/L_\perp$ a scattering cross section can be calculated for the scattering to free space (σ) and the scattering to surface plasmons (σ_{spp}) [84]:

$$\sigma = \frac{P_{out}}{P/L_\perp} \approx \frac{32\pi k_0^5}{3\sqrt{|\epsilon|}} (|\alpha_E|^2 + |\alpha_M|^2) \quad (5.19)$$

$$\sigma_{spp} = \frac{P_{spp}}{P/L_\perp} \approx \frac{8\pi^2 k_0^5}{|\epsilon|} (2|\alpha_E|^2 + |\alpha_M|^2) \quad (5.20)$$

Strikingly, both expressions have a different dependence on the dielectric constant ϵ of the metal. Our understanding of this is as follows: the magnitude of the dipole moment induced by an incident surface plasmon is inversely proportional to the mode size $d_{spp} \approx \sqrt{|\epsilon|}/(2k_0)$. The radiation to free space is comparable to that of a normal dipole, that is it has no dependency on ϵ . Radiation to the surface plasmon field, however, scales with the width of the angular spectrum [91] of the surface plasmon. This width is proportional to λ/d_{spp} , yielding an extra factor $\propto 1/\sqrt{|\epsilon|}$.

Expressing the equations in terms of d_{spp} and λ/d_{spp} yields:

$$\sigma \approx \frac{1}{d_{spp}} (16/3)\pi k_0^4 (|\alpha_E|^2 + |\alpha_M|^2) \quad (5.21)$$

$$\sigma_{spp} \approx \frac{\lambda}{d_{spp}^2} \pi k_0^4 (2|\alpha_E|^2 + |\alpha_M|^2) \quad (5.22)$$

where the factor $(16\pi/3) (|\alpha_E|^2 + |\alpha_M|^2)$ is what we call η in the main manuscript. If the electric polarizability is negligible, the ratio between these two cross sections is $\sigma_{spp}/\sigma = 3\lambda/(16d_{spp}) \approx 0.5$, for gold at 800 nm. This ratio is the power radiated to the surface plasmon field relative to that of the free space modes for a magnetic dipole. The same ratio can also be found by comparing the density of modes of the surface plasmon field to that of free space modes.

The theory in ref. [84] assumes that the surface plasmon is on a metal-air interface. In our experiment, however, it is a gold-glass interface, which

has some consequences for the quantitative agreement between the theory and experiment. For Rayleigh scattering, the expression is in terms of the wave vector inside the medium [92] ($k = n2\pi/\lambda$), hence we expect k_0^4 has to be replaced by k^4 . Additionally, $d_{spp} \approx \sqrt{\epsilon}/(2n^2k_0)$, contains the refractive index squared and the λ in the expression $\sigma_{spp}/\sigma = 3\lambda/(16d_{spp})$ should be λ/n . The modifications together yield an increase of n^6 of the scattering cross section σ .

A last step in calculating σ is finding the appropriate values for α_M and α_E . Rotenberg *et al.* calculated these values using a hole in a perfect electric conductor. In this paper, the calculated polarizability per cubed radius a^3 is plotted as a function of the size parameter a/λ . For $a/\lambda \ll 1$ the polarizability per cubed radius is constant. For the magnetic polarizability a shape resonance is found at $a/\lambda \approx 0.2$, and for larger a/λ the polarizability per cubed radius decreases. The polarizability depends on the film thickness: a 42% increase is found comparing a infinitely thick film with a zero thickness film. For the calculations in the main manuscript, we use the zero thickness value of $\alpha_M = 0.106a^3$ and $\alpha_E = 0.054a^3$.

Using these values for the polarizability the measured scattering cross section roughly an order of magnitude larger than predicted. To explain this difference between the theory and the experiment, we consider the penetration of the optical field into the metal. This penetration is neglected in calculating the polarizability of the hole, as it is calculated assuming a perfect electrical conductor. This penetration depth (~ 25 nm) is small compared to most holes in previous experiments ($a = 25$ nm – 500 nm) [84], where a good agreement between theory and experiment is reported. Somewhat speculatively, we estimate the effect of the penetration depth on the polarizability by modeling a hole in a metal of finite conductivity as a hole in a perfect electrical conductor whose radius is increased with the penetration depth of the field. If this approximation is valid, this would increase the expected value of ξ by a factor $(85/60)^6 = 8.1$ to $\eta = 23 \pm 10$. This would reduce the difference between theory and experiment to a factor 2.7 ± 1.2 .

Finally, we wish to relate the intensity ratio cross section A to the scattering cross section σ . In the main text we showed that the ratio A/σ is the maximum ratio between surface plasmon mediated transmission and directly transmitted light as $\langle I_s \rangle / \langle I_d \rangle \approx A/\sigma$ in the limit of high hole density. In this limit, the power flow simplifies as all excited surface plasmons are coupled out before they can be absorbed.

The power flow in the high density limit is sketched in Fig. 5.6. We assume an incident plane wave with power P_{in} which polarizes a hole and thereafter

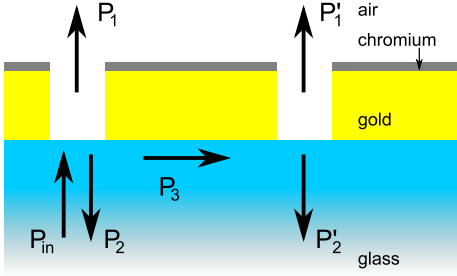


Figure 5.6: A sketch of the power flow, in the limit of high hole densities. An incident plane wave with power P_{in} induces a dipole moment. This dipole radiates into three channels: through the hole (P_1), into the substrate (P_2) and to a surface plasmon mode P_3 . For high densities there is no absorption loss, and hence all power in the surface plasmon mode is scattered out at other holes. At these holes the ratio between light scattered through the hole and into the substrate is defined to be P'_1/P'_2 .

radiates into three channels: P_1 through the hole, P_2 back into the substrate and P_3 into the surface plasmon field. In the high hole density limit the entire surface plasmon field is eventually scattered out, either through the hole (P'_1) or back into the substrate (P'_2), such that $P_3 = P'_1 + P'_2$.

The transmission via surface plasmons P_s is equal to P'_1 , but expressing it as $P_s = P_3 P'_1 / (P'_1 + P'_2)$ will prove to be more useful. Given that the directly transmitted power P_d is equal to P_1 , we find:

$$\frac{P_s}{P_d} = \frac{P_3}{P_2} \frac{P'_2}{P'_1 + P'_2} = \eta \frac{P_3}{P_2}. \quad (5.23)$$

where we assume $P'_1/P'_2 = P_1/P_2$, which physically means that the excitation and outcoupling are mediated via the same (magnetic) dipole moment. Note that we also introduced an efficiency $\eta = P'_2/(P'_1 + P'_2)$, which quantifies how much of the outcoupling is to the substrate relative to all light scattered out. At this point it is important to realize that ratio P_3/P_2 is equal to the ratio $P_{spp}/P_{out} = \sigma_{spp}/\sigma$ calculated in ref. [84]. Hence, $P_3/P_2 = \sigma_{spp}/\sigma = 3\lambda/(16nd_{spp})$. This allows us to relate A to σ :

$$\frac{A}{\sigma} = \frac{P_s}{P_d} = \eta \frac{\sigma_{spp}}{\sigma} = \eta \frac{3}{16d_{spp}} \frac{\lambda}{n}. \quad (5.24)$$

This is a very important result as we now found that the entire experiment can be described using a single scattering cross section σ , an efficiency η and some known prefactors. We expect η to be smaller but close to one, yielding a fairly accurate prediction for the relation between A and σ . This efficiency η , which has not been studied yet, basically quantifies how efficiently power is radiated to the substrate relative to the total radiative loss.

Density dependence of the loss and the intensity ratio

In this section we present the density dependence of L_{tot} and $\langle I_s \rangle / \langle I_d \rangle$ for three wavelength ranges: 705 ± 15 nm, 803 ± 13 nm and 881 ± 9 nm. We show that the measured dependencies are the same as those expected and presented in the main manuscript. Also, we show that these plots clearly reveal the strong wavelength dependence of $L_{\text{abs}}(\lambda)$, $\sigma(\lambda)$, and $A(\lambda)$ presented in the main text.

In Fig. 5.7a the fit parameter L_{tot} is plotted as a function of hole density for three different wavelengths. First, it is important that the expected density dependence ($L_{\text{tot}}^{-1} = L_{\text{abs}}^{-1} + \rho\sigma$) is satisfied, which is the case. Second, we can see that the inverse propagation length is not only density dependent, but also wavelength dependent. The slope σ decreases from 705 ± 15 nm to 803 ± 13 nm by a factor four, and the slope almost vanishes at 881 ± 9 nm. Third, the axis cutoff that resembles the surface plasmon absorption loss, decreases with wavelength by more than a factor two. This is consistent with the theoretically expected dependence.

In Fig. 5.7b we show the density dependence of the intensity ratio $\langle I_s \rangle / \langle I_d \rangle$. The data for each wavelength shows the expected dependence ($\langle I_s \rangle / \langle I_d \rangle = A\rho L_{\text{tot}}$), which is plotted as a solid line with only the vertical scale A as a free parameter. For this fit we use the values of L_{abs} and σ that we obtained for each wavelength in Fig. 5.7a to describe L_{tot} : $L_{\text{tot}}^{-1} = L_{\text{abs}}^{-1} + \rho\sigma$.

To relate Fig. 5.7b to the presented values for A in the main manuscript, we compare the three curves in Fig. 5.7 at low densities. There is a factor four difference between the top curve ($\lambda = 705$) and the lowest curve ($\lambda =$

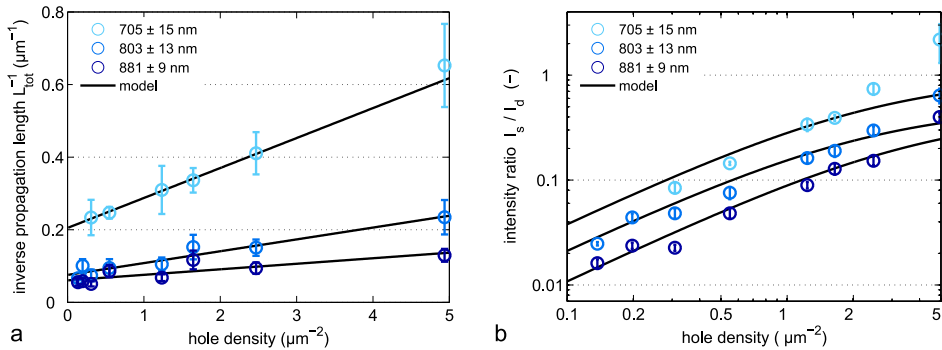


Figure 5.7: **a**, The inverse propagation length L_{tot}^{-1} as a function of hole density for three different wavelength ranges. Both the axis cutoff, i.e. the absorption, and the slope decrease with wavelength. **b**, The density dependence of the intensity ratio $\langle I_s \rangle / \langle I_d \rangle$. For each hole density the intensity ratio decreases with wavelength. In the low density regime the intensity ratio increases linearly.

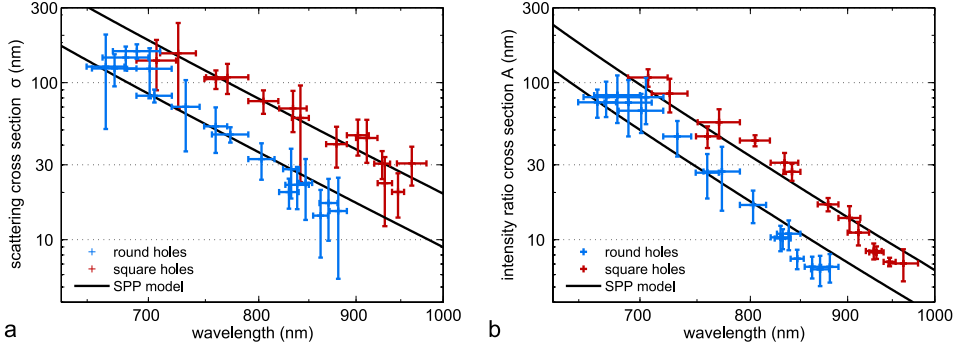


Figure 5.8: **a**, The measured values of the scattering cross section σ for circular and square holes. For both types the predicted wavelength dependence reproduces the data accurately. The prefactor for the round holes is smaller however. **b**, The measured values of the intensity ratio cross section A for round and square holes. Also for this parameter the predicted wavelength dependence describes the data of both round and square holes.

881). The propagation length, which is dominated by absorption at these low densities, increase roughly a factor two in this wavelength range. Hence the factor four difference between the curves in Fig. 5.7b resembles the order of magnitude change of A presented in Fig. 5.5.

Last, we note that the ratio $\langle I_s \rangle / \langle I_d \rangle$ shows an outlier at the highest hole density for each wavelength, which is systematically larger than the fitted model. These outliers are expected because we neglected the influence the quasi-cylindrical waves [35, 44, 47] in our model. Additionally, our model neglects the heavily damped surface plasmons on the gold-air interface, which is also not valid from high densities. For these reasons, these outliers are not used to obtain the fitted value for A .

Analysis of square holes

In addition to the round holes presented in the main text, we have also studied square holes. These square holes have a rib size (125 ± 5 nm) slightly larger than the diameter of the round holes (120 ± 6 nm). We are interested whether the shape has any influence on the magnitude of the scattering cross section its wavelength dependence.

In Fig. 5.8a we plot the results for the scattering cross section of the square holes, along with the results for round holes presented in the main text. The measured scattering cross section σ is larger for the square hole than that of the round holes, but its wavelength dependence is very similar. The suggested wavelength dependence $\sigma = \xi k^4 a^6 / d_{spp}$ accurately fits the experimental data, where we choose a the rib length divided by two. The prefactor ξ is found to

be 1.7 ± 1.3 larger for the square holes, where the large error bar is mostly the result of the error in the hole size.

In Fig. 5.8b we plot the results for the intensity ratio cross section, also with the results of the round holes. The value of A is larger for the square holes too. We fit the expected wavelength dependence of $A = 3\eta\lambda\sigma/(16nd_{spp})$, using the value of ξ just found and leaving only η as a free parameter. We find $\eta = 0.60 \pm 0.13$, which is comparable to that of round holes.

In conclusion, the data for the square hole shows the same wavelength dependence of σ and A . The prefactors η and ξ obtained for the square holes do not differ significantly from those found for round holes.

Loss compensation of extraordinary optical transmission

Surface plasmons offer many new possibilities in photonics, but applications are often limited by absorption or radiative loss. Compensating these losses will further improve the applicability of surface plasmons. In this context, long range surface plasmons have been successfully amplified [93, 94], and amplified spontaneous emission of surface plasmons is reported [21, 95]. Simultaneously, various kinds of surface plasmon lasers [96] have been studied experimentally [12, 40, 97, 98]. Despite the large interest in surface plasmon lasers, lasing may also be a nuisance when improving a lossy metallic system. Such problems may occur in metamaterials [22], like negative index materials [79, 99, 100] or $n=0$ metamaterials [9]. Also metal hole arrays, known for their extraordinary optical transmission [28], could show unintended laser action which may be detrimental to their performance. In this chapter we show strong loss compensation in extraordinary optical transmission and identify a number of challenges in loss compensation of resonant plasmonic systems in general.

6.1 Introduction

The metal hole array is an ideal system to study loss compensation for several reasons. First, the extraordinary optical transmission (EOT) is thoroughly studied, see [43] for a recent review. Second, the role of surface plasmons in the EOT phenomenon is also quantitatively understood nowadays [36, 37]. Third, the EOT has potential applications, like biosensing [39, 101]. Last, EOT is also an example of a Fano resonance [102], often exploited in plasmonic nanostructures [103–106].

Figure 6.1 shows the essence of the experiment, which is measuring the optical transmission of a metal hole array that has a semiconductor (InGaAs) gain layer placed in close proximity of the metal. The gain layer is pumped optically with a 1064 nm laser beam. The experiment is performed in a Helium flow cryostat to increase the potential gain of the semiconductor. The gain material can amplify the surface plasmons, which are excited at the holes, and thereby increase the optical transmission. The luminescence of the gain material is also transmitted through the holes and is emitted in all directions, which gives rise to background luminescence (red arrows).

6.2 Evolution with pump power

Figure 6.2 shows the key result of this chapter, namely the evolution of the transmission with pump power. The inset shows the transmission when the system is not pumped (red curve), with a transmission maximum of 8.6×10^{-3} . This spectrum shows the well-known Fano line shape. The dashed grey curve shows the estimated transmission in the absence of any surface waves, which is 2.3×10^{-3} at 1500 nm. Hence the transmission of the unpumped system is enhanced by a factor 3.7 by the presence of surface waves.

When we increase the pump power the maximum transmission increases

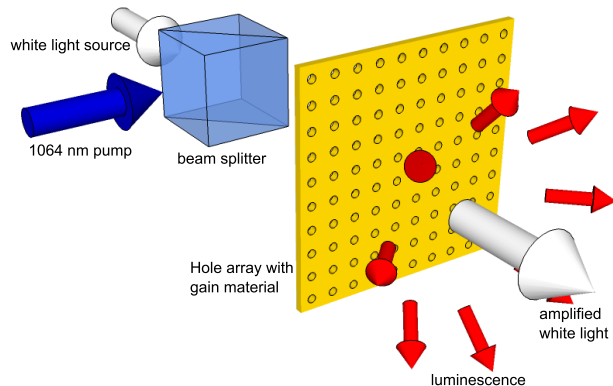


Figure 6.1: A sketch of the experiment. We record the white light transmission spectrum of a hole array that has a gain layer in close proximity of the metal, while pumping the gain layer with a laser. The luminescence from the pumped gain material is also recorded (red arrows).

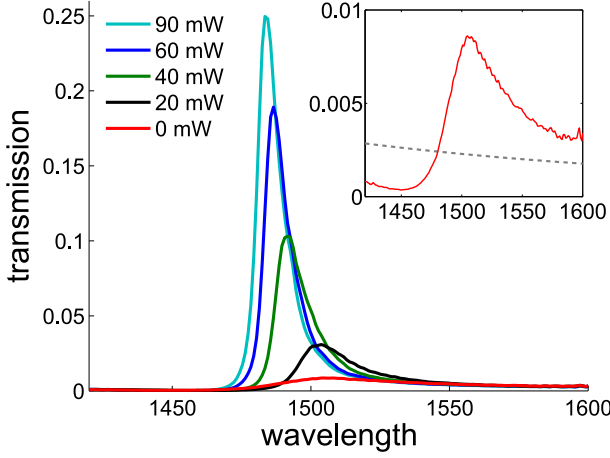


Figure 6.2: Measured transmission as a function of pump power. The transmission maximum gradually increases with pump power from 8.6×10^{-3} (see inset) to 0.25, which is a 31 fold enhancement. The lattice spacing of this sample is $a_0 = 450$ nm; 90 mW corresponds to roughly 7 kW/cm^2

dramatically, while it remains practically constant at other wavelengths. At 90 mW pump power, the transmission has reached a value of 0.25, corresponding to a 31 fold increase of the transmission. The transmission enhancement as a result of surface waves is now 109, instead of 3.7. As a reference, in a recent experiment without loss compensation we found an 17 fold enhancement for a hole array on glass [37]. Hence we conclude that loss compensation can dramatically increase the EOT.

6.3 Background luminescence

In Fig. 6.3 we show that the increased transmission is indeed due to amplification of the incident white light, because at the transmission maximum the transmitted intensity is much larger than the background luminescence. The transmission plotted here is $T = (I_{out} - I_{bg})/I_{in}$, where I_{in} is the incident white light intensity, I_{bg} is the background signal when only the pump is switched on, and I_{out} is the intensity recorded when both light sources are on. The background luminescence plotted in Fig. 6.3 is I_{bg} divided by the value of I_{in} at 1490 nm. We have also studied the background luminescence at an angle where the white light is absent, to check that the background is not affected by switching the white light source on.

At the transmission minimum the background signal exceeds the transmitted signal, which imposes a challenge on measuring the transmission minimum accurately. To limit this problem, the light source used in a loss compensated transmission experiment has to be sufficiently bright. Here, we use a superluminescent diode, which is much brighter than the halogen lamp that is generally used for EOT experiments.

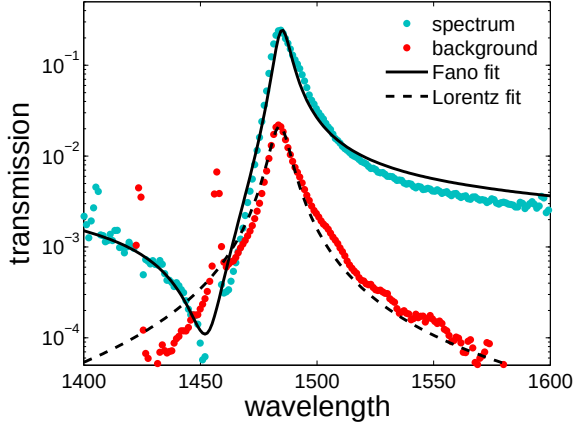


Figure 6.3: Comparison between the collected background luminescence and the transmitted white light. At the transmission maximum, the background luminescence is an order of magnitude smaller. At the transmission minimum, however, the magnitude of the background luminescence exceeds that of the transmitted white light. Please note that the plots are on a semilog scale.

We fitted the background luminescence using a Lorentzian lineshape, which describes the luminescence to good approximation. The resonance wavelength and spectral width of the Lorentzian is the same as that of the Fano resonance that is fitted to the transmission spectrum. This shows that both resonances have the same origin, namely surface waves propagating on the metal dielectric interface.

The background luminescence in Fig. 6.3 exhibits two narrow linewidth resonances, at ~ 1425 nm and ~ 1460 nm. These are resonances with a subradiant character [107]. We have recently shown that such resonances can show laser action. Fortunately, this laser emission is donut shaped and therefore it has limited emission normal to the surface where we measure the transmission. Nonetheless, this laser action may hinder loss compensation in two ways. First, the lasing peak will largely exceed the transmitted signal and therefore subtraction of the background may become inaccurate. Second, when the device starts lasing the gain of the semiconductor will not increase anymore with a further increase of pump power, because all additional pump power is lost to the lasing mode [21, 108]. Hence, these two narrow linewidth resonances exemplify that loss compensation can lead to lasing effects at one wavelength that hinder further loss compensation at other wavelengths [22].

6.4 Quantitative analysis

We now compare the transmission enhancement for a set of three different samples with different lattice spacings a_0 : 470, 460 and 450 nm, of which the 450 nm sample is discussed in Figs. 6.2 and 6.3. This quantitative analysis will address two questions: What limits the observed enhancement? How much gain is supplied by the semiconductor material? The transmission spectra of

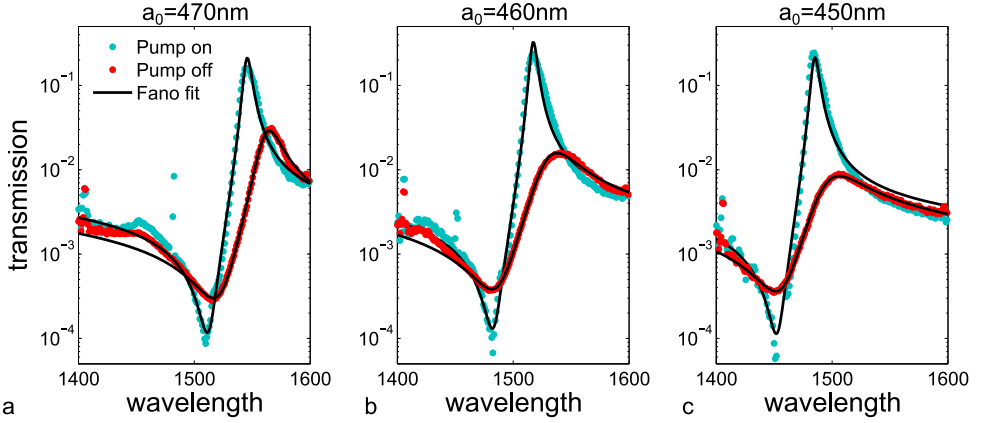


Figure 6.4: Transmission for samples of different lattice spacing: $a_0 = 470$ nm (a), 460 nm (b), and 450 nm (c). The transmission at maximum pump power (90 mW) is compared to that without pumping. The data is fitted to a Fano resonance.

these three samples are plotted on a semilog scale in Figs. 6.4a, 6.4b and 6.4c, where the spectra with the pump on and off are compared. Each sample is pumped strong enough to be close to full inversion within the bandwidth of the Fano resonances. Noteworthy is that the transmission at the minimum decreases in presence of the pump, showing that pumping can also decrease the transmission.

The six smooth curves in Fig. 6.4 are Fano fits to the data. Our Fano expression is an approximate version of the microscopic model for EOT, which is recently developed [36] and verified [37]. The expression for these fits is as follows:

$$T = \left| t\omega^2 + \frac{\alpha\omega^4}{\omega - \omega_0 + i\gamma} \right|^2 \quad (6.1)$$

The parameter t is proportional to the transmission in absence of any surface wave, α quantifies the combined excitation and outcoupling, which also has a phase difference with respect to t , ω_0 is the resonance frequency and γ is the combined ohmic and radiative loss of the surface plasmon. For convenience we expressed ω , ω_0 and γ in eV.

In Table 6.1 we show the key fit values for the three plots in Fig. 6.4, comparing the pumped and unpumped resonances. For the unpumped samples, this linewidth increases with decreasing lattice parameter. This is as expected: at reduced wavelengths the absorption losses in the gold and the unpumped gain medium increase, while also the scattering losses induced by the holes increase. For the pumped samples, the increased ohmic and radiative losses

Table 6.1: Fit parameters corresponding to the Fano fits in Fig. 6.4. Dimensions of λ_0 and $\Delta\lambda$ are nm, and eV^{-4} and eV^{-2} for $|\alpha|$ and t , respectively

Parameter	λ_0	$\Delta\lambda$	$ \alpha $	t
470 nm, off	1562	12	2.8×10^{-3}	73×10^{-3}
470 nm, on	1545	4	2.4×10^{-3}	87×10^{-3}
460 nm, off	1530	20	2.7×10^{-3}	80×10^{-3}
460 nm, on	1517	4	2.5×10^{-3}	90×10^{-3}
450 nm, off	1494	24	2.1×10^{-3}	70×10^{-3}
450 nm, on	1485	4	2.1×10^{-3}	81×10^{-3}

could explain that the linewidth does not become smaller than 4 nm.

The difference between the pumped and unpumped linewidth is the gain experienced by the surface plasmon. This difference increases more than a factor two from 8 to 20 nm when a_0 decreases. The 20 nm difference can be converted into the net gain that is experienced by the surface plasmon using:

$$\frac{4\pi n_{\text{eff}}}{\lambda^2} (\Delta\lambda_{\text{off}} - \Delta\lambda_{\text{on}}) = L_{\text{off}}^{-1} - L_{\text{on}}^{-1} \quad (6.2)$$

where L_{off} and L_{on} are the effective propagation lengths without and with pump, respectively. The difference $L_{\text{off}}^{-1} - L_{\text{on}}^{-1}$ of $3.5 \times 10^3 \text{ cm}^{-1}$ for the $a_0 = 450 \text{ nm}$ sample, is the change in inverse absorption length of the surface waves. To relate it to the material gain, this value needs to be divided by the confinement factor, estimated to be 0.32 for our system. For a conservative estimate of the material gain at this wavelength and at full inversion, we also have to divide $L_{\text{off}}^{-1} - L_{\text{on}}^{-1}$ by two. Hence, the estimated material gain is $5.5 \times 10^3 \text{ cm}^{-1}$ for the 450 nm sample and $2.2 \times 10^3 \text{ cm}^{-1}$ for the 470 nm sample. These numbers are reasonable for a semiconductor operated at high carrier densities and low temperatures [97, 109].

The parameter α , which models the excitation and outcoupling of the surface plasmons shows little dependence on pump power for each sample. This shows that the measured enhancement and the spectral width are consistent, and in agreement with $T_{\text{max}} \approx |\alpha|^2 \omega^8 / \gamma^2$. The parameter t is also more or less constant for all six cases, as expected for the nonresonant transmission.

Finally, we note that there is much room for improvement of our results, for example by carefully maximizing the thickness of the gain layer, placing the gain layer closer to the metal interface, or using quantum wells. The system would also become more practical when electrical pumping is implemented and when the structures can be operated at room temperature.

6.5 Conclusion

We have demonstrated the improved performance of a plasmonic system using a semiconductor gain material. We increase the transmission of a metal

hole array by a factor 31. Our quantitative analysis shows that we experimentally obtain the large but expected material gain. Three challenges are identified that are generic to loss compensation in plasmonic systems: (1) subtraction of the background luminescence can be troublesome; (2) to observe any transmission signal, the signal beam needs to be sufficiently intense compared to the background luminescence; (3) gain saturation will occur when the structure lases unintentionally. Therefore, loss compensated systems need to be designed with much care, taking the physics of the gain medium and optical resonances into account.

6.6 Methods

The sample is fabricated as follows. On a semi-insulating indium phosphide wafer a lattice-matched indium gallium arsenide layer is grown (105 nm), which is subsequently covered with a thin (15 nm) layer of indium phosphide. Hereafter a 5 nm protective silicon nitride layer is grown using plasma enhanced chemical vapor deposition. On these layers we fabricate the metal hole array by depositing 100 nm gold and 20 nm titanium on a lithographically defined array of dielectric pillars. To provide sufficient adhesion of the gold onto the silicon nitride, we deposit a very thin (average thickness smaller than 0.5 nm) titanium adhesion layer in between these layers. The last step is to etch the pillars away, leaving the subwavelength holes (diameter 160 nm).

The setup for transmission measurements is as follows. We illuminate the sample with two beams, at the sample the pump beam is roughly 40 μm diameter and the white light signal roughly 12 μm diameter. To create a pump spot of uniform intensity we illuminated a pinhole with the pump laser, and imaged this pinhole on the sample. The diameter of this pinhole is roughly two times smaller than that of the laser beam. The signal beam is generated using a superluminescent diode with a center wavelength of 1550 nm and a spectral width of 110 nm. The transmitted light is collected using a microscope objective. The far field of the objective is imaged onto a single mode fiber, which is subsequently led to a grating spectrometer with a linear array. The angular resolution of the setup is ~ 4 mrad. This low angular resolution helps to minimize the collected laser emission from the hole arrays.

Because the sample is placed inside a cryostat, we can not move the sample in and out of the beam without changing the alignment. For this reason we use the signal at unperforated areas of the sample as a reference. Later, we measure the transmission spectra of the unpumped samples outside the cryostat to determine the correct prefactor for the transmission measurement.

Surface plasmon lasing observed in metal hole arrays

Surface plasmons in metal hole arrays have been studied extensively in the context of extraordinary optical transmission, but so far these arrays have not been studied as resonators for surface plasmon lasing. We experimentally study a metal hole array with a semiconductor (InGaAs) gain layer placed in close (20 nm) proximity of the metal hole array. As a function of increasing pump power we observe an intense and spectrally narrow peak, with a clear threshold. This laser emission is donut shaped and radially polarized. Three experimental observations support that the system shows surface plasmon lasing. First, the full wavelength dispersion of the observed resonances can be understood using a single surface plasmon mode of the system. Second, the polarization of these resonances is as expected for surface plasmons. Third, the magnitude of the avoided crossing, which results from mode coupling at the holes, has a similar magnitude as found in simulations using surface plasmons.

F. van Beijnum, P. J. van Veldhoven, E. J. Geluk, M. J. A. de Dood, G. W. 't Hooft, and M. P. van Exter, submitted for publication.

7.1 Introduction

Surface plasmon lasers and spasers [96] have recently attracted much interest (see e.g. [97] for a review) and are attractive candidates for nanoscale lasers. Lasing is reported in different nanoscale resonators: metal-coated nanopillars [11], metal-coated nanorings [110], semiconductor nanowires on a silver film [40, 111], and gold nano spheres [12]. However, claims of surface plasmon lasing are often hard to substantiate and experimental observations can be misinterpreted as (surface plasmon) lasing [112]. Therefore, studying surface plasmon lasing in a simple and well known model system will contribute to our understanding of surface plasmon lasing and the limitations thereof.

A metal hole array is a thoroughly studied plasmonic system, mainly in the context of extraordinary optical transmission [28]. For surface plasmon lasing, however, hole arrays have not yet been considered. Nonetheless, experiments on hole arrays without a gain material suggest that the holes in the array could provide the required feedback for lasing. For example, the wavelength dispersion of the resonant transmission reveals that the surface plasmons couple with the array, which results in an avoided crossing [28, 49, 50, 113]. This coupling behavior can be related to transmission and reflection coefficients of surface plasmons, using a recently developed [36] and experimentally verified [37] microscopic theory.

More generally, it is shown that two-dimensional photonic crystals with gain can show laser action [114–116]. In the far-infrared a photonic crystal laser is demonstrated where the mode volume is reduced using a surface

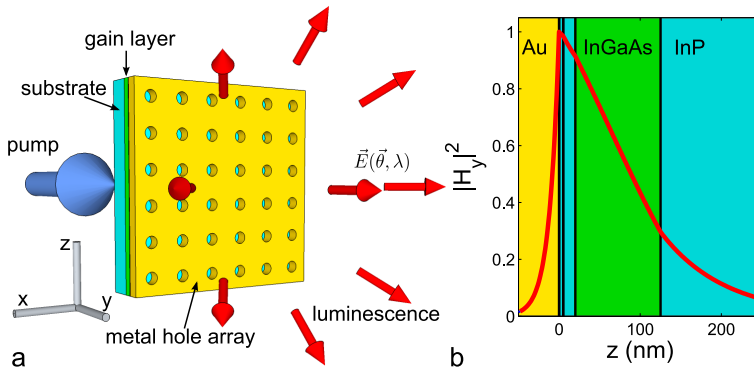


Figure 7.1: **a**, Sketch of our experiment: a semiconductor layer, in close proximity of a metal hole array, is pumped optically. We perform an angle-resolved study of the luminescence transmitted through the hole array. **b**, The absolute value squared of the magnetic field H_y of the designed surface plasmon mode (solid curve) exhibits a field maximum on the gold-dielectric interface. The 105 nm InGaAs gain layer is placed roughly 20 nm from the gold, using a InP and SiN spacer layers.

plasmon mode [117]. At these wavelengths, however, the surface plasmon absorption is modest [97]. Hence, a major challenge to obtain lasing in metal hole arrays at optical frequencies is to overcome both the absorption losses and the radiative losses of the surface plasmons.

In this Chapter we demonstrate surface plasmon lasing in metal holes arrays and substantiate our claim of surface plasmon lasing with three different experimental observations. The experiment, illustrated in Fig. 7.1a, studies an optically pumped semiconductor gain layer placed in close proximity of a metal hole array. The luminescence that is transmitted through the hole array is recorded. After discussing this experiment in more detail, we first present the observation of laser action in this system. Thereafter we present measurements on the luminescence that are resolved by angle, wavelength and polarization thereby revealing the nature of the laser action.

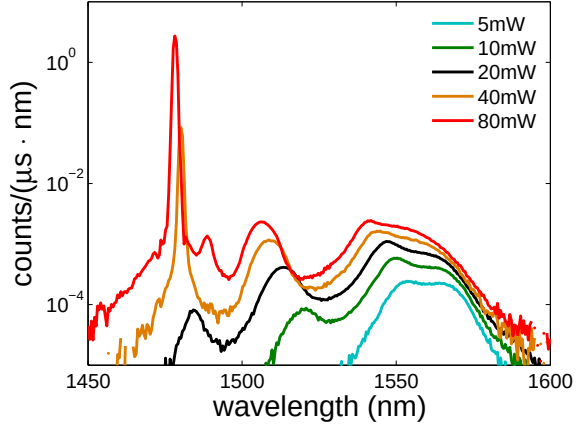
7.2 Experiment

In Fig. 7.1b we show the layer stack used in our experiment. On a semi-insulating indium phosphide (InP) wafer a lattice-matched indium gallium arsenide (InGaAs) layer is grown, which is subsequently covered with a thin (15 nm) layer of InP. Hereafter a 5 nm protective silicon nitride (SiN) layer is grown using plasma enhanced chemical vapor deposition. On these layers we fabricate the metal hole array by depositing 100 nm gold and 20 nm titanium on a lithographically defined array of dielectric pillars. To provide sufficient adhesion of the gold onto the SiN, we deposit a very thin (average thickness smaller than 0.5 nm) titanium adhesion layer in between these layers. The last step is to etch the pillars away, leaving the subwavelength holes (diameter 160 nm).

To estimate the gain required to compensate the absorption loss of the surface plasmons, we calculate the complex effective refractive index of the layer structure numerically [88]. The spatial mode overlap between the gain layer and the surface plasmon is shown in Fig. 7.1b, solid red curve. Using literature values for the complex refractive index of the gold, InP and InGaAs layers and a measured value of the SiN layer, we find that a material gain of $3 \times 10^3 \text{ cm}^{-1}$ is required to compensate the absorption loss of the surface plasmon mode. This required gain is large, but realistic for bulk InGaAs [109].

The real part of the calculated effective refractive index is closely related to the wavelength of the resonances in a metal hole array [36, 37]. For the 105 nm InGaAs layer the surface plasmon has a calculated effective refractive index of $n_{\text{eff}} = 3.43$. The InGaAs layer thickness is chosen to maximize the gain, while maintaining single mode operation. For our system the transverse electric waveguide mode is well below cut-off, as this mode is only supported

Figure 7.2: Luminescence spectra as a function of pump power, plotted on a semilog scale. For increasing pump power the bandwidth of the luminescence increases until the device starts lasing. Above threshold, the emission of the non-lasing resonances starts to saturate at a maximum intensity.



for InGaAs thicknesses above ~ 170 nm.

Our experimental setup is as follows. We pump the InGaAs gain layer using a continuous wave laser at a wavelength of 1064 nm. To pump the structure with a uniform intensity we illuminate a $600\ \mu\text{m}$ diameter pinhole with the gaussian laser beam, and image this aperture on the sample with a 20x demagnification. The sample is placed in a Helium flow cryostat to study the structure at temperatures down to 5 K. The cryostat has windows on both sides of the hole array sample, allowing us to pump the structure on one side and study the luminescence at the other side.

We record the luminescence as a function of the angle $\vec{\theta}$, by imaging (Magnification = 0.25) the Fourier plane of a microscope objective with a long-working-distance (Mitotoyu, focal distance 10 mm, NA=0.4) on a single-mode fiber placed on a xyz-stage. The angular resolution of our setup is approximately 4 mrad, which is determined by the modal-field diameter of the single-mode fiber. By scanning the fiber through the Fourier plane and spectrally resolving the fiber output (~ 1 nm resolution), we can create an image of the Fourier plane for each emitted wavelength. In addition to the far-field imaging, we can also adjust the optics such that the fiber is placed in an image plane of the sample, allowing us to make spatial images of the laser for each wavelength.

7.3 Laser threshold

In Fig. 7.2 we plot the recorded spectra as a function of the pump power, at an angle $(\theta_x, \theta_y) = (0\ \text{mrad}, 48\ \text{mrad})$. For most pump powers three maxima in the luminescence are seen. These maxima are associated with different resonances of the hole array, of which the origin will be discussed later. As the

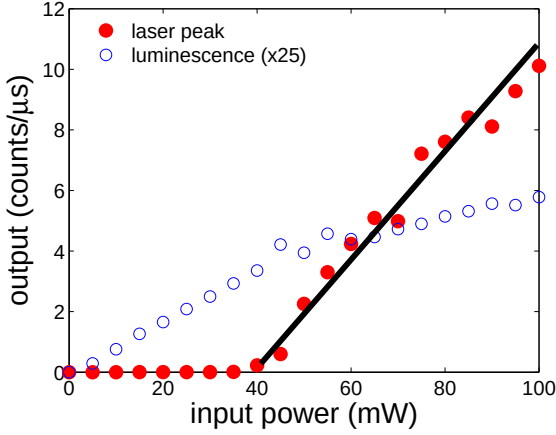


Figure 7.3: The output in the lasing peak and in the luminescence in the range of 1485 – 1600 nm. The power in the lasing peak shows a clear threshold (red). The black line is a guide to the eye. The luminescence outside the lasing peak starts to level off, as expected for lasing in semiconductor devices (blue).

pump power is increased from 5mW (cyan) to 20mW (black) the luminescence maxima at ~ 1560 nm and ~ 1520 nm increase in intensity, and a third peak becomes visible at ~ 1480 nm. Increasing the intensity further to 40 mW (orange) the ~ 1480 nm peak increases dramatically. A further increase in the pump power to 80 mW (red) shows an increase of the ~ 1480 nm peak by more than an order of magnitude, while the intensity of the 1510 nm peak increases only a factor two. The dramatic increase of the ~ 1480 nm peak suggests that the structure is lasing.

The existence of a clear laser threshold is supported by Fig. 7.3, where we plot the recorded intensity integrated over the spectral peak at ~ 1480 nm as a function of input power. A distinct laser threshold is seen at roughly 40 mW pump power. We also plot the integrated luminescent intensity in the wavelength range of 1485 – 1600 nm. The integrated luminescence first increases linearly with the input power but for powers larger than the threshold it starts to level off, indicating the expected carrier pinning [11, 108].

The results presented in Figs. 7.2 and 7.3 are for a laser that has a lattice spacing of 470 nm, and lases at 1478 nm. We also fabricated lasers with a lattice parameter of 460 nm and 450 nm. Both devices show lasing too, with the laser wavelengths of 1450 nm and 1419 nm respectively. Hence the ratio between the laser wavelength and lattice parameter is a constant, namely 3.15 ± 0.01 . This clear relation between the lattice constant and the laser wavelength shows that the hole array is used as a resonator.

7.4 Dispersion

To reveal the origin of the three resonant peaks in Fig. 7.2, we will now study the wavelength dispersion of these resonances. This dispersion has been

intensively studied in the context of the extraordinary optical transmission [28, 49]. In these experiments the resonant wavelength of the transmission is studied as a function of the angle of incidence. We use angle-resolved measurements of the luminescence, below laser threshold, to study the dispersion of the resonances in our structure. A study below threshold is convenient, as it allows us to study the angle-resolved luminescence without a lasing peak that saturates our detector.

The emission angle as a function of wavelength can often be understood using a simple model that assumes uncoupled traveling waves. For two neighboring holes on either the x -axis or y -axis this traveling wave has a phase difference of $k_x a_0$ or $k_y a_0$ respectively, with $\vec{k} = (k_x, k_y)$ the wave vector of the traveling wave. This linear phase results in a plane wave emitted at an angle (θ_x, θ_y) . Relating k_x and k_y to the total momentum, $k^2 = k_x^2 + k_y^2$, the emission angle can be calculated [49]:

$$n_{\text{eff}}^2 k_0^2 = |k|^2 = |k_{||,x} - m_x G|^2 + |k_{||,y} - m_y G|^2. \quad (7.1)$$

with $G = 2\pi/a_0$, n_{eff} the mode index of the traveling wave, (m_x, m_y) the diffraction order, $k_{||,x} = k_0 \sin \theta_x$ and $k_{||,y} = k_0 \sin \theta_y$. Equation (7.1) describes a circle of radius k around the point $(m_x G, m_y G)$. In our measurements, the mode index is the only unknown parameter. Equation (7.1) describes uncoupled traveling waves, but often suffices to understand the main features of the dispersion. For the relatively weakly interacting surface plasmons, the avoided crossing can only be observed when curves of different (m_x, m_y) intersect.

In Fig. 7.4 we show the measured angle-resolved luminescence for nine wavelengths, along with a plot of Eq. (7.1) for $n_{\text{eff}} = 3.26$. We plot the images with decreasing wavelength, i.e. increasing k . The general behavior is well predicted by the simple model and shows the square symmetry of the lattice. As a function of k the circles increase in radius, thus move towards the optical axis (1600 nm – 1540 nm), cross it (1540 nm – 1520 nm) and thereafter move away from it (1520 nm – 1440 nm). At the angular position where the theoretical lines intersect, coupling is found (1520 nm – 1440 nm), changing the far field drastically. From the correspondence between the figures and the theory for uncoupled modes we conclude that one value of n_{eff} is required to understand all far-field luminescence patterns. Given that our layer stack only supports a single mode, the surface plasmon, this is the first experimental observation that supports our claim of surface plasmon lasing.

The found value of $n_{\text{eff}} \sim 3.26$ is low compared to the value we expect for the surface plasmon mode (3.43). This difference is partially due to cooling the sample and a carrier induced refractive index change. Furthermore, the

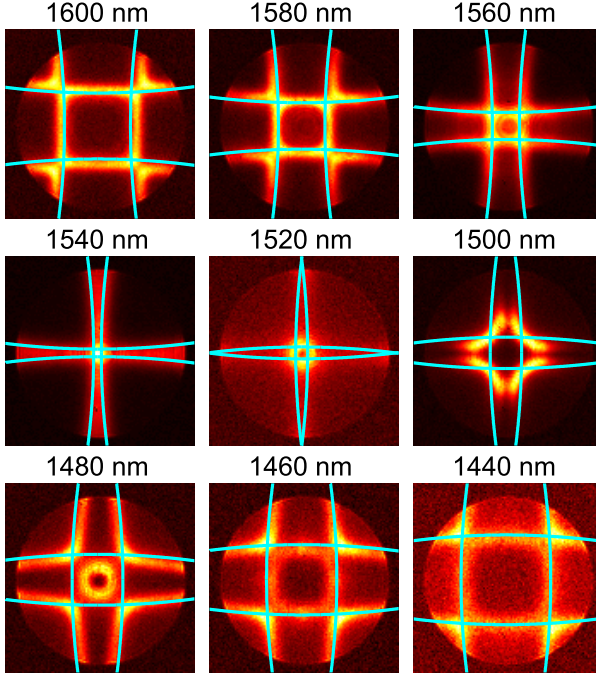


Figure 7.4: Far field emission pattern for nine wavelengths. The device is operated at 150K, where the gain is too small to reach threshold. These images reveal the dispersion of the modes inside the metal hole array. The cyan lines correspond to the dispersion of uncoupled traveling waves with an effective index of 3.26, showing that only one mode is needed to understand the dispersion of the luminescence maxima. The circle shows the numerical aperture of the microscope objective (NA=0.4), the optical axis is the center of this circle.

predicted value of n_{eff} is very sensitive to the thickness of the low-index layer of SiN. Increasing the thickness of this layer 10 nm already lowers the index of the surface plasmon from 3.43 to the experimentally observed value of 3.26. An alternative explanation for the low value of n_{eff} is a thin layer of oxide on the InP.

The luminescence in Fig. 7.4 is recorded without using a polarizer. By studying the dispersion of the resonances for p - and s -polarization separately, we can show whether the observed resonances are mediated via a transverse electric (TE) or a transverse magnetic (TM) mode [118]. Performing this measurement, we find three p -polarized resonances and one s -polarized resonance if we scan along θ_y while setting $\theta_x = 0$ (see appendix). The observed polarization dependence of the resonances is consistent with a TM mode around $\lambda/a_0 = n_{\text{eff}}$. Because a surface plasmon is a TM-polarized mode, this provides the second observation that substantiates our claim of surface plasmon lasing.

From this dispersion we can also quantify the magnitude of the coupling that the mode experiences from the holes. As discussed in the appendix the avoided crossing has a splitting of roughly 65 nm, which compares very well with the simulated avoided crossing for surface plasmons (70 nm), while the splitting for a TE mode is less than 1 nm. Hence, the magnitude of the splitting

of the avoided crossing is the third experimental observation that supports a claim of surface plasmon lasing.

7.5 Polarization of laser emission

Last, we find that the laser emission is donut shaped, as shown Fig. 7.5a, which shows the emission above threshold for a temperature set at 5 K. This behavior compares well to transmission experiments and simulations on these hole arrays, where it is shown that this resonance vanishes when excited at normal incidence [36, 107]. The diameter of the ring we observe is 120 mrad, which is Fourier related to a diameter of roughly 16 μm on the sample. The donut shaped emission reproduces in all lasing samples, although the beam quality varies from sample to sample.

To study the local polarization of the lasing mode we show an image of the emission when the vertical polarization is selected using a polarizer (Fig. 7.5b). This image, on the same intensity scale as Fig. 7.5a now shows two lobes, instead of a ring. If we set the polarizer at an arbitrary angle of 30 degrees (Fig. 7.5c) the lobes rotate along with the polarizer, showing that the laser emission is radially polarized. This radial polarization is consistent with the polarization analysis performed in the appendix, which shows that for angles $(0, \theta_y)$ the polarization is vertical and for angle $(\theta_x, 0)$ the polarization is horizontal.

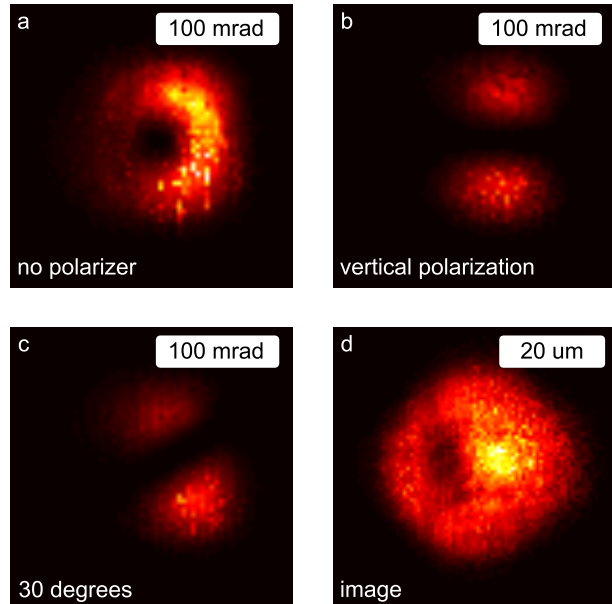


Figure 7.5: a-c, Polarization analysis of the angle resolved intensity at the lasing wavelength. **a**, No analyzing polarizer. **b**, Vertical polarization selected. **c**, Thirty degrees from vertical. **d**, Direct image of the luminescence at the lasing wavelength. The structure lases at all positions where it is pumped.

In Fig. 7.5d we also show an real-space image of the laser mode. The round circular spot is comparable to the size of the pumped area ($\sim 30 \mu\text{m}$), albeit somewhat larger ($\sim 40 \mu\text{m}$). This size difference between the lasing spot and pump spot is probably due to carrier diffusion out of the pumped region, which is expected to be a few micrometer. The observed emission is really laser light, as a similar image at a non lasing wavelength is three orders of magnitude less intense. When comparing Figs. 7.5a and 7.5d, we note that the far-field spot is not Fourier related to the image of the lasing spot. This suggests that the laser has multiple spatial modes, a hypothesis that is supported by the observation that we can reduce the size of the pump spot and still obtain lasing.

7.6 Conclusion

We have demonstrated surface plasmon lasing in metal hole arrays. The luminescent input-output characteristic shows clear threshold behavior and the integrated emission levels off, which indicates carrier density pinning [11, 108]. The laser emission is not linearly polarized but radially polarized and thus donut shaped. We find three experimental observations that support our claim of surface plasmon lasing: first, the angle-resolved luminescence shows that all observed resonances are mediated via one mode, while the layer stack only supports a surface plasmon; second, the polarization of the four observed resonances is as expected for surface plasmons; third, the dispersion shows a large avoided crossing of which the magnitude is almost the same as found from simulations using surface plasmons.

Using metal hole arrays as laser resonators could be very important in understanding surface-plasmon lasing, as we consider it to be an ideal model system. The metal hole arrays are well-known structures, hence deviations from the expected behavior can be interpreted in terms of new physical effects induced by the lasing surface plasmon. The metal hole arrays provide the ability to study the band structure and perform polarization analysis, which yields insights into the laser physics. Further miniaturizing the laser, for example by increasing the feedback provided by holes, could make the laser more interesting for photonic integration.

Appendix

In this Appendix we perform a polarization-resolved study of the resonances found in the far-field luminescence, presented in Fig. 7.4. We hereafter compare these measurements to simulations of the angle-dependent transmission of these arrays.

Figure 7.6 shows the measured dispersion of the resonances for p - and s -polarization. These measurements are performed by scanning the fiber along the θ_y direction and at $\theta_x = 0$. We then select either the vertical (corresponding to p) or horizontal (corresponding to s) polarization using a polarizer.

For the p -polarization, we find three resonances. For two of these resonances the emission angle depends linearly on $\sin \theta_y$, except for small angles. This dispersion characteristic is very comparable to that seen in a one dimensional system where TM-polarized traveling waves propagate in the plus or minus y -direction. Close to the optical axis an avoided crossing with a splitting of 65 nm is found. The third resonance seen for the p -polarization shows very little dispersion. In literature it is shown, using two-dimensional coupled mode theory, that such a low dispersion mode is expected for the p -polarization [118]. For the s -polarization, only one resonance is found. Also this low-dispersion resonance is predicted by two-dimensional coupled mode theory [118]. This s -polarized resonance is degenerate on the optical axis with a p -polarized resonance. This polarization dependence has been observed experimentally in several transmission measurements on hole arrays [28, 49, 50, 107, 113]

Although the dispersion of the measured resonances is associated with a TM mode, it is not necessarily a surface plasmon. To study this in more detail, we perform rigorous coupled-wave analysis using commercially available code (Rsoft Diffractmod). This software allows us to simulate the angle-dependent

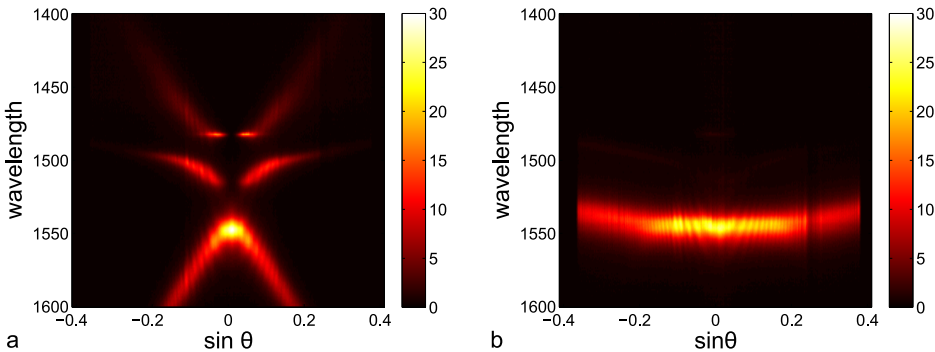


Figure 7.6: Wavelength dispersion of the resonances as a function of the radiation angle ($0, \theta_y$) for p -polarization (a) and s -polarization (b). At each non-zero angle we observe three p -polarized resonances and one s -polarized resonance in this wavelength range. The scale bar shows the counts per millisecond.

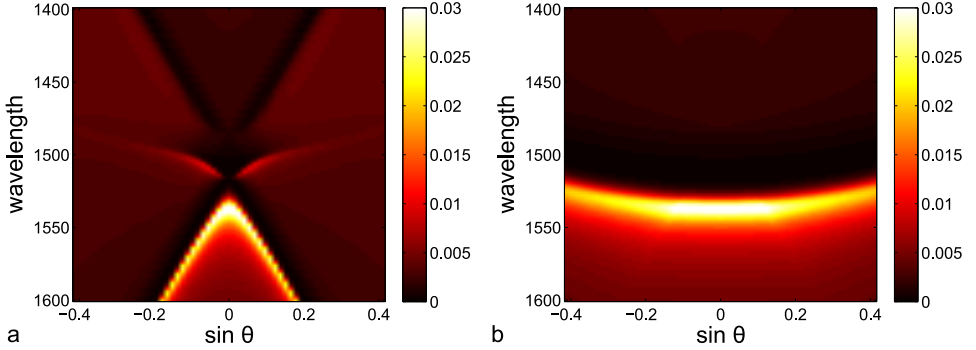


Figure 7.7: Simulated angle dependent transmission spectra for p -polarization (a) and s -polarization (b). As with the measurements, three resonances are visible for p -polarization at a given angle, and only one for s -polarization. In addition, the splitting found in the simulations is comparable to that found in the experiment.

transmission of the metal hole array with the layer structure that we designed. Using the original design, Fig. 7.1, we found that the resonance is shifted with respect to the observed resonance wavelengths. From the dispersion data presented in the main manuscript we already concluded that if the lasing is mediated via surface plasmons, their effective index is lower than anticipated.

As discussed in the main manuscript, the measured effective refractive index of the surface plasmon does not match the expected value. To shift the simulated resonance wavelength to that of the measurement we replaced the SiN layer with a 15 nm layer with an index of 1.8, which shifts the transmission resonance close to the observed luminescence maximum. The gain layer is set to have a gain of 3000 cm^{-1} . For this simulation, the effective mode index of the surface plasmon mode that exists on the metal-dielectric interface is calculated to be $n_{\text{eff}} = 3.21$.

The simulated angle-dependent transmission is presented in Fig. 7.7, where Fig. 7.7a shows the p -polarization, and Fig. 7.7b shows the s -polarization. The product of the mode index ($n_{\text{eff}} = 3.21$) and the lattice parameter (470 nm) is 1510 nm, which is roughly in the center of the avoided crossing. In contrast to the luminescence, this angle-dependent transmission contains a Fano resonance, because there is interference between light that is directly transmitted through the hole and light transmitted via surface plasmons.

Two important observations can be made from the comparison between the simulated resonances and the measurements. First, both the simulation and measurement shows that the system has three p -polarized resonances and one s -polarized resonance. Second, the magnitude of the splitting ($\sim 70 \text{ nm}$) simulated for p -polarization agrees reasonably well to that of the measurements ($\sim 65 \text{ nm}$). Hence, the measured dispersion appears to be a signature of

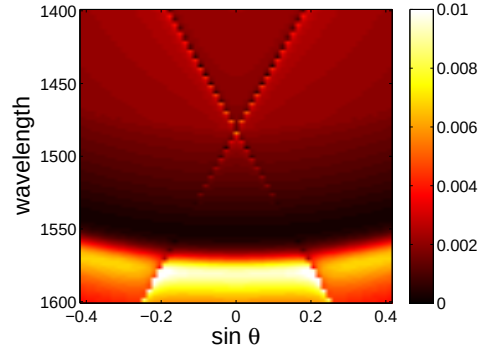


Figure 7.8: Simulated angle-dependent transmission spectra for a TE mode, simulated for s -polarization. Most importantly, the splitting on the optical axis is practically negligible for this mode.

surface plasmons. This is additional evidence that the lasing is mediated via surface plasmons.

To be sure that the splitting and the dispersion is unique for surface plasmons we will now introduce another mode by increasing the InGaAs from 105 nm to 180 nm thickness. This will introduce a guided TE-mode, and it will increase the effective index of the surface plasmon from 3.21 to 3.30. Hence, the surface plasmon resonances will be red-shifted by approximately 42 nm. To make the surface plasmon mode less prominent we now set the gain in the InGaAs layer to zero.

In Fig. 7.8 the simulated dispersion for this thicker gain layer is seen, for s -polarization. Three resonances are seen, two narrow resonances that intersect at ~ 1480 nm and one broad resonance at ~ 1580 . The broad resonance is the surface plasmon resonance, also seen in Fig. 7.7 for s -polarization, but shifted by the expected 40 nm. The narrow resonances that intersect on the optical axis are mediated via the introduced TE-mode. The resonances are much sharper than for surface plasmons, because the guided mode experiences less absorption loss. The avoided crossing for this TE mode is too small to be observed, showing that a TE mode in our system does not experience strong feedback from the holes.

In conclusion of this appendix, we have shown three aspects of the dispersion that show that we observe a surface plasmon mode in our experiments. First, the measured dispersion corresponds closely to simulations in which we know we are dealing with surface plasmons: three p -polarized resonances are observed and one s -polarized resonance is observed. Second, we extracted the magnitude of the avoided crossing which is almost identical in experiments and simulations. This shows that the avoided crossing has a magnitude that is to be expected for surface plasmons. Finally, we show that a TE mode has a negligible avoided crossing.

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Samenvatting

In dit proefschrift bestuderen we de verstrooiing, demping en versterking van oppervlakteplasmonen. Een oppervlakteplasmon is licht dat gevangen is aan het oppervlakte van een metaal. Oppervlakteplasmonen zijn een goed voorbeeld van het gebruik van metalen voor optica op de nanoschaal. De bijzondere brekingsindex van een metaal biedt vele mogelijkheden die momenteel worden verkend, zoals een exotische eigenschap als een negatieve brekingsindex. Het grote nadeel van metalen voor optische toepassingen is dat een metaal geneigd is licht om te zetten in warmte. Hierdoor gaat het licht, en de informatie die het licht bevat, verloren. Hierdoor is de voortplantingslengte van een oppervlakteplasmon vele miljoenen keren kleiner is dan van licht in een glasvezelkabel. In dit proefschrift onderzoeken we of dit verlies van oppervlakteplasmonen tegengegaan kan worden door oppervlakteplasmonen te versterken terwijl ze zich voortplanten.

In onze experimenten bestuderen we oppervlakteplasmonen op dunne maar ondoorzichtige metaallagen, die we hebben doorboord met minuscule gaten. Elk van deze gaten laat een kleine beetje licht door, maar de gaten kunnen ook samenwerken waardoor ze gezamenlijk meer licht door de gaten loodsen dan de som van de individuele bijdragen. Dit verschijnsel heet ‘buitengewone optische transmissie’, en wordt grotendeels verklaard doordat er oppervlakteplasmonen worden gegenereerd bij de gaten. Als deze oppervlakteplasmonen een ander gat tegenkomen krijgt het licht een tweede kans om door de metaallaag heen te gaan, wat de toename in de transmissie verklaart. Met behulp van deze methode bestuderen wij oppervlakteplasmonen en proberen we de verliezen van de oppervlakteplasmonen te begrijpen en te compenseren.

In Hoofdstuk 2 laten we zien dat er niet alleen oppervlakteplasmonen worden aangeslagen bij het belichten van een gat, maar ook een tweede golf, de quasi-cylindrische golf. Het bestaan van deze tweede golf was al aangetoond, maar het was nog niet experimenteel bewezen dat het ook belangrijke consequenties heeft voor buitengewone optische transmissie. In Hoofdstuk 2 laten we zien dat deze tweede golf buitengewone optische transmissie aanzienlijk vergroot als de gaten op zeer kleine afstand staan. Bij grotere afstanden tussen de gaten wordt de buitengewone optische transmissie gedomineerd door oppervlakteplasmonen.

In Hoofdstukken 3 en 4 bestuderen wij buitengewone optische transmissie

wanneer de gaten niet geordend worden geplaatst, zoals gebruikelijk, maar in een willekeurig patroon. We vinden dat orde niet noodzakelijk is voor het verhogen van de transmissie door middel van oppervlakteplasmonen, hoewel orde het effect wel doet toenemen. Verrassend is dat de wanordelijke patronen eenvoudiger te modelleren zijn dan geordende patronen.

In Hoofdstuk 5 bestuderen we een oppervlakteplasmon dat door een gat wordt verstrooid naar licht in de vrij ruimte. Onze analyse laat zien dat de golflengte-afhankelijkheid van dit proces heel goed vergelijkbaar is met een klein deeltje dat licht verstrooit, beter bekend als Rayleigh verstrooiing. De theorie van Rayleigh verstrooiing is oorspronkelijk ontwikkeld om de blauwe kleur van de lucht te verklaren. Ons experiment laat zien dat het mogelijk wordt om de buitengewone optische transmissie te begrijpen in termen van ontwerpgrontheden zoals de gatgrootte en de brekingsindex van het metaal.

In Hoofdstuk 6 bestuderen we de buitengewone optische transmissie in een nieuwe context. We proberen de hoeveelheid licht die door de metaallaag gaat te vergroten door de oppervlakteplasmonen te versterken. Hiervoor plaatsen we een bijzonder materiaal in de buurt van de metaallaag: een zogeheten ‘versterkingsmateriaal’. Dit materiaal kan licht versterken met behulp van energie die door een externe bron wordt aangeleverd, de pomp. We bestuderen vervolgens de buitengewone optische transmissie terwijl we de door de pomp aangeleverde energie vergroten. Door het pompen neemt de buitengewone optische transmissie ruim dertig keer toe. We tonen vervolgens aan dat deze toename komt door de versterking van oppervlakteplasmonen.

In Hoofdstuk 7 laten we tot slot zien dat het versterken van oppervlakteplasmonen een bijzonder effect geeft: in onze experimenten gaan de oppervlakteplasmonen in het rooster als een laser samenwerken. Deze waarneming geeft aan dat de versterkte oppervlakteplasmonen helemaal zonder verlies over het metaaloppervlakte reizen. Behalve verliesloze oppervlakteplasmonen is terugkoppeling ook een noodzakelijk element voor een laser, met andere woorden de oppervlakteplasmonen moeten door spiegels worden weerkaatst. Bij deze laser zijn het de gaten die voor deze terugkoppeling zorgen.

Samenvattend, in dit proefschrift hebben we laten zien dat de absorptieverliezen van oppervlakteplasmonen in een gatenrooster volledig gecompenseerd kunnen worden, wat tot laserwerking leidt. De laser kunnen we grotendeels begrijpen door de opgedane kennis over de verstrooiing van oppervlakteplasmonen aan gaten. In vervolgonderzoek kunnen we bestuderen of deze oppervlakteplasmonlasers nog veel kleiner gemaakt kunnen worden. Daarnaast is het interessant om te analyseren voor welke toepassingen een plasmonisch systeem met verliescompensatie van meerwaarde is.

Curriculum Vitæ

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List of publications

- Frequency bandwidth of light focused through turbid media.
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