Exploring optical switching in subwavelength hole arrays

Lydwin van Rooyen
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Supervisor:
Prof. dr. L. Kuipers
Daily supervisor:
Ir. J.C. Prangsma
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Abstract

Fabrication of an ultrafast all-optical switch is one of the challenges in modern optics. In order not to limit the speed of data transfer with an electronic switching element, it is necessary to eliminate conversion of an optical signal to an electrical signal. Optically nonlinear materials, materials that have a strongly intensity-dependent refractive index, are the most likely candidates to make an all-optical switch.

A change in refractive index alone is not enough to make a good all-optical switch. It must be possible to block and unblock the signal with this change in refractive index. To do so, we here present an experiment in which a nonlinear material is used as a substrate for a metallic subwavelength hole array. The transmission spectra through such an array shows sharp, wavelength-dependent features. Furthermore, the transmission is much higher than expected based on the open fraction of the array, for certain wavelengths. The combination of the strong resonances of the array and the enhanced fields on the interface between the array and the nonlinear material should yield strong switching behaviour.

The location and shape of the features in the transmission spectrum of a metallic subwavelength hole array depend on the refractive index of the dielectric materials the array is in contact with. By exploiting the spectral features and the effects of a change in refractive index of the nonlinear substrate, we attempt to demonstrate the suitability of this system for building an all-optical switch.
Chapter 1

Introduction

In telecommunication, light is the premier medium to transport information. In order to control the path of the light, components like switches and waveguides are used. Ideally, such components do not decrease the speed of the information transfer. A beam splitter is an example of a device that can influence an optical signal nearly instantaneously. Optical switches, however, remain a challenge. Most current technologies require that an optical signal is converted to an electrical signal, and converted back to light after switching. Such a process limits the bandwidth of the switch.

Fast optical switches are needed to perform tasks like logic operations. They are difficult to make, since in order to not limit their bandwidth, all components have to be fast. Current technologies depend on electro-optic, magneto-optic, acousto-optic and all-optical effects, and entail varying the refractive index of the medium through which the light propagates, thus altering the beam path.

If we want to build an all-optical switch, we need a nonlinear optical material. In these materials, the refractive index changes as a function of the local intensity of light. The best candidates for optical switching have a high nonlinear response, ensuring that measurable effects occur even for intensities that are commonly used in electro-optical applications. The response also needs to be fast, to ensure that the reaction time of the device is not limited by the speed of the switching device. In nonlinear optical materials, the electronic response to the applied field is the only limiting factor to the speed, making them suitable to build the fastest possible switching devices.

A good nonlinear material alone is not enough to create a high-quality optical switch. A change in refractive index will cause light incident on the nonlinear material under an angle to be transmitted under a slightly different angle, or alter the percentage of transmitted light a little. The phase and the polarization of the light beam also change as a function of the refractive index when it passes through an interface. Those changes alone will not lead to switching behaviour, however.

A common way to use nonlinearity for switching is to consider the way nonlinear materials change the polarization of light traveling though them. A polarizing filter can
be used to detect changes in polarization, thus producing an optical switch. However, changes in angle and polarization are minute and hard to measure. The system that is investigated in this project therefore combines the characteristics of a nonlinear material with the properties of an ordered nano-structure commonly known as a subwavelength hole array, in order to combine the ultrafast intensity-dependent change in refractive index of an optically nonlinear material with the sharp wavelength-dependent resonances of the hole array.

The subwavelength hole array, essentially a grid of subwavelength holes in a metal film, was first investigated by Ebbesen [1]. According to Bethe’s theory of diffraction through small holes [2], the transmission of light through a subwavelength hole is significantly smaller than predicted from the intensity per surface area. However, for an array of holes in a metallic film, Ebbesen found that the intensity of the transmitted light can be much higher than predicted by Bethe. Moreover, for certain wavelengths the transmitted fraction of the incident light exceeds the open fraction of the film! This effect is now known as extraordinary transmission. In order to explain this phenomenon, Ebbesen concluded, the hole array must be considered an active element in the path of the incident beam. Apparently, the holes and the metallic substrate influence the way in which light is transmitted through the sample.

The wavelengths for which extraordinary transmission occurs in a hole array depend on the material the array is made of, on the materials the array has interfaces with, on the periodicity of the structure and on the shape and size of the holes ([1], [3], [4]). Although it is generally accepted that collective oscillations of the free electrons on the surface of the metallic array, surface plasmon polaritons, play an important role in the phenomenon of extraordinary transmission, the exact mechanics behind this process remain unclear.

When a metallic hole array is coated with a nonlinear material, the applied light intensity can cause a change in refractive index of the nonlinear coating, which can in turn cause a shift in the transmission spectrum. A wavelength that is transmitted well by the unswitched structure can be blocked by the switched structure. This makes the combination of the two aforementioned phenomena an interesting candidate for a fast all-optical switch, and a system well worth studying.

Zayats and co-workers from Belfast [5] presented some of the first experimental results on nonlinear-coated hole arrays. They show that optical switching can indeed be accomplished in the proposed structure. In this project, we investigate similar systems, and their responses to femtosecond excitations. We hope to develop a structure in which ultrafast all-optical effects are measurable, and we will explore the possibilities of using such a structure as an all-optical switch.
1.1 Overview of the thesis

In Chapter 2 a summary of the theoretical background needed to understand the experiments we did is presented, along with a first estimate of the effects we can expect.

The experiments themselves will be discussed in Chapter 3. In the first series of experiments, we try to characterize the nonlinear behaviour of the materials we use. The materials that are discussed are one conjugated polymer, poly-3-butoxy-carbonyl-methyl-urethane (3-BCMU), and two chalcogenide glasses, As$_2$S$_3$ and Ge$_{11.5}$As$_{24}$Se$_{64.5}$. All of these materials are documented to have a high optical nonlinear response. An experiment is performed to verify this.

We then present transmission measurements through the hybrid structure of a layer of nonlinear material on a metallic subwavelength hole array. We measure the intensity-dependent transmission of this structure.

In Chapter 4, the results of the experiments are discussed. An assessment of the experimental errors is made, and aided by those we draw some conclusions from the experimental results.

Finally, Chapter 5 contains some ideas and recommendations about further research on this topic.
Chapter 2

Theory

In this chapter, the essential theoretical background of the physics encountered in this research is given. In section 1, the theory of extraordinary transmission will be discussed. In section 2, the optical Kerr effect will be discussed in some detail. Finally, section 3 combines those two phenomena to discuss nonlinear transmission through hole arrays.

2.1 Extraordinary transmission

In 1998, Ebbesen et al. [1] discovered that the transmission through an array of subwavelength holes in a metal film can be much higher than predicted by standard diffraction theory [2]. The transmission spectra of such structures show distinct minima and maxima at certain wavelengths (see figure 2.1). Because for certain colors the transmitted fraction of the incident light exceeds the open fraction of the film, the phenomenon was called extraordinary transmission.

Surface plasmon polaritons, collective oscillations of the free electrons on the metallic surface of a hole array, influence extraordinary transmission. In this section, we will explain the conditions under which they exist on subwavelength hole arrays.

2.1.1 Surface plasmon polaritons

Surface plasmon polaritons (SPPs) are collective oscillations of the free electrons of a metal interacting with an electromagnetic wave. They are strongly confined to the surface, their electromagnetic field decays exponentially with distance from the surface [6]. The presence of SPPs leads to a strong local field enhancement at the interface of a metal and a dielectric, and many possible applications have been discussed. An overview of effects related to metal films with subwavelength holes in them and their possible applications was published in Nature last year [7]. In particular, the phenomenon of extraordinary transmission has often been attributed to SPPs.
Figure 2.1: Transmission spectrum of an array of $20 \times 20$ holes in a 200 nm thick Au film on glass. The periodicity of the array is 510 nm in both directions. The hole size is $225 \times 150$ nm. Wood’s anomalies are distinctly visible at 550 nm and 800 nm. This measurement was done at AMOLF by the author.

**SPPs from Maxwell’s equations**

We can explain the existence of SPPs using Maxwell’s equations. We start from a situation as sketched in figure 2.2. We assume that both half-spaces are infinitely extended. For this derivation, we consider a real value of $\varepsilon$ for all materials. This is generally not true for a metal, but the derivation can be generalized for complex values of $\varepsilon$. Our sample differs from this situation in that it is an optically thick (250 nm) metallic film between two different dielectrics. The conditions we derive in this chapter can and must be applied to both metal-dielectric interfaces. We consider a TM-mode plane wave propagating in the $x$-direction. Such an electromagnetic wave has an electric field with components in the $x$- and $z$-directions, and a magnetic field in the $y$-direction. In this
section, the index $d$ will be used for the dielectric, while the metal will be designated by $m$. The electric and magnetic fields in the two media can be written as:

$$E_{d,m} = (E_{x,d,m}, 0, E_{z,d,m})e^{i(k_x x - \omega t)}e^{ik_{z,d,m}z}, \tag{2.1}$$

$$H_{d,m} = (0, H_{y,d,m}, 0)e^{i(k_x x - \omega t)}e^{ik_{z,d,m}z}, \tag{2.2}$$

In the absence of free charges, the normal component of the electric displacement $D_z$, is be conserved across the interface. Through Maxwell’s equations, $D_z$ is related to $E_z$ as $D_z = \varepsilon_0 \varepsilon E_z$. Hence, $E_z$ has a discontinuity across the interface. The other nonzero components, $E_x$ and $H_y$, are continuous.

Using $\nabla \cdot E = 0$, we find that

$$E_{z,d,m} = -E_{x,d,m}\frac{k_x}{k_{z,d,m}}. \tag{2.3}$$

We can find the relation between $E_x$ and $H_y$ by using that $\nabla \times E = -\mu \frac{\partial H}{\partial t}$. Using $\mu_1 = \mu_2 = \mu_0$, which is true in non-ferrous materials, we find that

$$H_{y,d,m} = \omega E_{x,d,m}\varepsilon_1\varepsilon_0/k_{z,d,m}. \tag{2.4}$$

From the boundary conditions, it follows that $H_y$ and $E_x$ must be continuous at the interface. Using that, we can now relate the relative permittivities to the normal components of the wavevectors as

$$\frac{\varepsilon_1}{k_{z,d}} = \frac{\varepsilon_2}{k_{z,m}}. \tag{2.5}$$

Using momentum conservation, we can derive the following constraints:

$$k_{z,d,m} = -i\sqrt{k_x^2 - \varepsilon_{d,m}k_x^2}, \quad k_x^2 > \varepsilon_{d,m}k_x^2, \tag{2.6}$$

where $k = \omega/c$. Substituting 2.6 into the boundary conditions, we find the dispersion relation for the electromagnetic wave we are considering.

$$k_x = k\sqrt{\frac{\varepsilon_{d}\varepsilon_m}{\varepsilon_d + \varepsilon_m}}. \tag{2.7}$$

Note that the dielectric constant of a metal, $\varepsilon_m$, is complex and depends on the wavelength. If the absolute value of the real part of $\varepsilon_m$ is larger than $\varepsilon_d$, which is generally true in the system we consider, the square root in eq. 2.7 is larger than unity. Hence, the in-plane wavevector $k_x$ is larger than the wavevector of the light in the dielectric. For a more in-depth treatment of this derivation, the reader is referred to [8].
Phase matching

We note that the wave vector $k$ in the dispersion relation 2.7 is a 2D wave in the $x - y$ plane. As the in-plane wavevector is larger than the wavevector of the light in the dielectric, it appears impossible to match the phase of this wave with that of a light beam. It is necessary to find a condition under which the phases match, because momentum must be conserved in the system. One way to solve this is by adding a periodic corrugation to the surface. Exploiting the reciprocal lattice vectors of the corrugation, we can match the wavevectors as follows:

$$k_{\text{SP}} = k_{//} + lG_x + mG_y,$$

(2.8)

where $k_{//}$ is the in plane component of the wave vector of the impinging light, $l$ and $m$ are integers and $G_x$ and $G_y$ are the unit reciprocal vectors in $\hat{x}$ and $\hat{y}$-directions respectively. $G_x$ and $G_y$ are defined by

$$G_x = \frac{2\pi}{a_x}\hat{x} \quad \text{and} \quad G_y = \frac{2\pi}{a_y}\hat{y}$$

(2.9)

with $a_x$ and $a_y$ the lattice periodicity in the $\hat{x}$ and $\hat{y}$-directions respectively. Note that eq. 2.9 only holds for simple periodic corrugations, like the rectangular corrugations we consider. Assuming that the light beam impinges perpendicularly to the surface of the metallic interface, $k_{//} = 0$, and that $a_x = a_y = a_0$ (square lattice), wave vectors that can couple to surface plasmons and excite SPPs in our structures obey

$$|k_{\text{SP}}| = |lG_x + mG_x| = \frac{2\pi}{a_0}\sqrt{l^2 + m^2}.$$

(2.10)

From the dispersion relation 2.7 we can now calculate the optical frequencies that satisfy the SP condition. Combining equations 2.7 and 2.10, we find a formula that predicts the surface plasmon polariton resonant condition

$$\lambda_{\text{vac/res}} = \frac{a_0}{\sqrt{l^2 + m^2}} \sqrt{\frac{\varepsilon_d \cdot \varepsilon_m(\omega)}{\varepsilon_d + \varepsilon_m(\omega)}}.$$

(2.11)

This derivation assumes that the corrugation of the metal surface is a small perturbation on the surface plasmon waves. If we fill in the respective dielectric constants of the dielectric materials in contact with both sides of the metallic film, we can predict the position of resonances of the grating. Those resonances correspond to minima in the transmission spectrum, the so-called Wood’s anomalies [9]. The transmission peaks, originating from resonances inside the holes, always occur slightly to the red of the Wood’s anomalies.
2.1.2 Rayleigh anomalies

A more mundane effect in transmission gratings is the existence of Rayleigh anomalies. These minima in transmission occur in all gratings, regardless of the material. This effect was first noticed Lord Rayleigh [10]. As a diffracted order of light grazes the surface, the reflection and transmission of the grating are suppressed. In hole arrays, which can be considered gratings, this effect also occurs.

For normal incidence, the location of the Rayleigh anomalies is given by

\[ \lambda_{\text{Rayleigh}} = \frac{a_0}{\sqrt{l^2 + m^2} \sqrt{\varepsilon_d}}. \]  

(2.12)

Note that the samples we used have two different metal-dielectric interfaces: one with either glass or air, and one with a nonlinear material. Equation 2.12 gives us the location of the Rayleigh anomalies induced by both interfaces, by filling in the appropriate dielectric constants.

Wood’s anomalies and Rayleigh anomalies occur at very similar wavelengths. In transmission spectra, it is therefore impossible to distinguish between the two. However, while the Wood’s anomaly influences the coupling of light into surface plasmon polaritons, the Rayleigh anomaly does not influence that process. For that reason, it is impossible to use the measured transmission minima to describe the coupling of light into surface plasmons.

2.1.3 Hole shape effects

So far, only the periodicity of the corrugation of the metal film has been taken into account. In practise, this corrugation is an array of holes with a diameter of the same order of magnitude as the lattice spacing and the wavelength. So far, we have assumed that the corrugations are point-like. Therefore, our model does not match reality as closely as we would like, and we need adjustments to relate model and reality.

If light of a frequency that resonates with the SPP crystal modes impinges on the array, a resonance will occur on both sides of the array. In the case of a free-standing film those resonances will be the same; in a hole array on a substrate they will be different. Either way, to get the energy from one side of the film to the other, the resonances have to combine into a single resonance condition. The frequencies of the combined resonance are different from the individual resonances of the surfaces. The interaction between the SPP modes on both sides of the array depends on the thickness of the layer [11].

In addition, it was shown that the size [4] and shape [3] of the holes are also important parameters. Using rectangular arrays, for example, leads to a redshift of the spectrum and an appreciable increase in the normalized transmission through the array. That the effects of shape and size are far from negligible can be seen when studying the
second-harmonic generation from a hole array. It was even found that for rectangular holes with a very precise aspect ratio, the second-harmonic generation from the array is increased by a full order of magnitude [12].

Summarizing, there are quite a few parameters that alter the transmission through a hole array, so that the simple derivation in section 2.1.1 does not lead to quantitatively realistic results. Still, theory can be used to predict the position of spectral minima, so that those minima can be attributed to specific orders of Wood’s and Rayleigh’s anomalies.

Polarization dependence

In arrays with holes that have a more complex symmetry than simply circular or square, like the rectangular holes we used in our samples, the polarization of the incident light strongly influences the transmission. Light polarized parallel to the long edge of the holes will be mostly blocked, while light polarized parallel to the short edge of the holes will be transmitted very well.

In the experiments described later, the polarization of the light was always chosen parallel to the short edge of the holes, to ensure maximum transmission.

2.2 Optical Kerr effect

The optical Kerr effect is a phenomenon that occurs in materials when very strong electromagnetic fields are present, for example the highly enhanced fields near a hole array. The polarization of the material, which in low fields scales linearly with the field strength, will become proportional to a higher power of the field strength, leading to a refractive index that changes as a function of the field strength.


2.2.1 Nonlinear susceptibility

If one applies an electric field to a medium, the response of the medium is described by the polarization \( \mathbf{P} \). This polarization is caused by the fact that the charge distribution in the medium will change due to the applied field. In general, the medium will need time to respond to an applied field. The polarization is therefore usually calculated in the frequency domain:

\[
\mathbf{P} = \mathbf{P}(\mathbf{E}(\omega)).
\]  

(2.13)

We will make the assumption that the medium is lossless and dispersionless, and therefore responds instantaneously to a change in the electromagnetic field. This assumption allows us to use constant values of the nonlinear susceptibilities, while we
would normally have to account for their dependence on the frequency of the applied field. We can also While these approximations are too strict for real media, it does allow us to develop an intuition for the mechanism of optical nonlinearity.

\( \mathbf{P}(\mathbf{E}) \) is usually not known analytically, but it can always be approximated using the Taylor expansion

\[
\mathbf{P}(\mathbf{E}) = \varepsilon_0 (\chi^{(1)} \mathbf{E}(t) + \chi^{(2)} \mathbf{E}^2(t) + \chi^{(3)} \mathbf{E}^3(t) + \ldots). \tag{2.14}
\]

Here, \( \chi^{(n)} \) are the \( n^{th} \) order electric susceptibilities of the material, which in general have the form of an \( (n+1)^{th} \) rank tensor. We ignore all terms with \( n > 4 \).

**Terms of \( \chi \)**

- In general, media are not self-polarized, so \( \chi^{(0)} = 0 \).

- For small enough electric fields, the change in polarization due to the applied field is to a good approximation linear:

\[
\mathbf{P}(\mathbf{E}) = \varepsilon_0 \chi^{(1)} \mathbf{E}. \tag{2.15}
\]

- For higher electric fields, e.g. when the surface plasmon polariton crystals we consider are excited with a pulsed laser, higher-order terms in eq. 2.14 may become relevant. In that case, the response of the polarization of a medium to an applied electric field is no longer strictly linear, and we enter the domain of nonlinear optics.

- In a centrosymmetric medium the contribution of \( \chi^{(2)} \) to the polarization must vanish. This can easily be seen by looking at the corresponding term in 2.14, assuming that the effect of an electric field on the polarization is instantaneous and ignoring the tensor nature of \( \chi^{(2)} \) and the vector nature of \( \mathbf{P} \) and \( \mathbf{E} \):

\[
\mathbf{P}^{(2)} \propto \chi^{(2)} \mathbf{E}^2(t). \tag{2.16}
\]

If we now assume inversion symmetry, reversing the electric field should lead to the inverse effect on the polarization:

\[
-\mathbf{P}^{(2)} \propto \chi^{(2)} [ -\mathbf{E}(t) ]^2 = \chi^{(2)} \mathbf{E}^2(t). \tag{2.17}
\]

So either \( \mathbf{E}^2(t) = -\mathbf{E}^2(t) = 0 \) (trivial) or \( \chi^{(2)} = -\chi^{(2)} = 0 \).

- The third-order nonlinear susceptibility, \( \chi^{(3)} \), does not vanish in a symmetrical system.
While we are not necessarily working in a perfectly centrosymmetric medium, materials with a high value of $\chi^{(2)}$ are generally highly ordered, making them unsuitable for layer deposition. Consequently, we will choose materials in which the leading order nonlinear term is the third-order nonlinear susceptibility, $\chi^{(3)}$.

### 2.2.2 Nonlinear refractive index

To characterize the nonlinear response of a material, we can define a nonlinear refractive index. Intuitively, we can write the refractive index $n$ as

$$ n = n_0 + n_2 I, \quad (2.18) $$

where $n_0$ is the linear refractive index, $I$ is the intensity of the electric field in the sample and $n_2$ is a new constant, which we call the second-order index of refraction.

We can describe the interaction of light with a nonlinear optical medium by looking at the influence of the nonlinear part of the polarization on the propagation of a beam. Considering scalar fields only (this derivation can easily be adapted to describe vector fields as well), the electric field of light with a frequency $\omega$ is given by

$$ E(t) = E_0 \cos \omega t. \quad (2.19) $$

We can substitute this in the Taylor expansion of the polarization, eq. 2.14, ignoring the $\chi^{(0)}$ and $\chi^{(2)}$ terms and the tensor nature of the polarization:

$$ P(t) = \varepsilon_0 \chi^{(1)} E_0 \cos \omega t + \varepsilon_0 \chi^{(3)} E_0^3 \cos^3 \omega t + \ldots \quad (2.20) $$

Some algebraic manipulation of the $\cos^3 \omega t$ term leads to a more manageable form:

$$ \cos^3 \omega t = \cos \omega t (\cos^2 \omega) = \cos \omega t \left( \frac{1}{2} + \frac{1}{2} \cos 2\omega t \right) $$

$$ = \frac{1}{2} \cos \omega t + \frac{1}{2} \cos \omega t \cos 2\omega t $$

$$ = \frac{1}{2} \cos \omega t + \frac{1}{4} \cos \omega t + \frac{1}{4} \cos 3\omega t $$

$$ = \frac{3}{4} \cos \omega t + \frac{1}{4} \cos 3\omega t. \quad (2.21) $$

The last term in eq. 2.21 describes third harmonic generation, and is left out of the further derivation.

We now have

$$ P(t) = \varepsilon_0 (\chi^{(1)} + \frac{3}{4} \chi^{(3)} E_0^2) E(t). \quad (2.22) $$

Noting that in general the refractive index $n$ can be written as
where $\chi_{\text{eff}} = \chi^{(1)} + \frac{3}{4} \chi^{(3)} E_0^2$, we see that the change in polarization affects the refractive index of the material.

Manipulating expression 2.23 further, we find that

$$n = \sqrt{1 + \chi_{\text{eff}}},$$

which, using a Taylor expansion, can be approximated by

$$n \approx n_0 \left(1 + \frac{3\chi^{(3)}}{8n_0^2} E_0^2 \right) = n_0 + \frac{3\chi^{(3)}}{8n_0^2} E_0^2.$$  \hspace{1cm} (2.24)

If we now use the intensity

$$I = \frac{1}{2} \varepsilon_0 n_0 c E_0^2,$$  \hspace{1cm} (2.25)

we can substitute $E_0^2$ by

$$E_0^2 = \frac{2I}{\varepsilon_0 n_0 c}.$$  \hspace{1cm} (2.26)

If we then substitute eq. 2.26 into eq. 2.24, we find an expression that has the same form as eq. 2.18:

$$n = n_0 + \frac{3\chi^{(3)}}{4\varepsilon_0 n_0^2 c} I.$$  \hspace{1cm} (2.27)

We now have an expression for the second-order index of refraction, related to the third-order nonlinear susceptibility:

$$n_2 = \frac{3\chi^{(3)}}{4\varepsilon_0 n_0^2 c}.$$  \hspace{1cm} (2.28)

Typical values of $n_2$ found in strongly nonlinear bulk materials are $10^{-10} - 10^{-13}$, corresponding to $\chi^{(3)}$-values of the order of $10^{-8} - 10^{-12}$ [6].

### 2.3 Nonlinear transmission

Now that we know how a high value of $\chi^{(3)}$ can influence the refractive index of a material when illuminated with a high intensity, and how the refractive index of a dielectric coating influences the transmission through a hole array, we can predict the effects of using such a material as a substrate for a hole array.
Indeed, both 2.11 and 2.12 depend directly on the dielectric constant of the interface materials of the array. The dielectric constant is defined as the square root of the (complex) refractive index of a material. Thus, a change in refractive index implies a change in dielectric constant, which in turn induces a change in the positions of the maxima and minima in the transmission spectrum of a hole array.

The materials that were available during this project have bulk $n_2$ values of around $10^{-13}$ cm$^2$/W. We chose materials that are nonresonant in the range of the laser we used, so that no frequency-dependent absorption effects interfere with the phenomena we try to measure.

When we illuminate these samples with a light intensity near their damage threshold of roughly 100 GW/cm$^2$, we find a change in refractive index of the order of $10^{-2}$. While this is a small effect, it should be possible to see the induced change in the transmission spectrum through an array. In particular, if we study an array at a wavelength that corresponds with the steep flank of a high transmission peak, even a very small shift of the peak will result in an appreciable change in transmission for that wavelength.

We can estimate this effect, by considering a gold hole array coated with a nonlinear material with a given dielectric constant. We choose realistic values for the height and width of the peak from transmission spectra measured before. We assume that the unswitched structure has a transmission peak at 800 nm. Using eq. 2.11, assuming that the dielectric constant of gold for this wavelength is $\varepsilon_m=6.9$ and using a value of $\varepsilon_d=2.6$ for the dielectric constant of the nonlinear material, we see that a peak position of 800 nm corresponds to a period of 582 nm.

If we now induce a change in the refractive index of the nonlinear material, changing it from 2.6 to 2.61, we find the new peak position, 801.11 nm. If we now take the maximum of the difference between the values of two Gaussians around the old and new peak position, we find that the difference in normalized transmission can get as high as 5%. That is certainly an effect that should be readily measured. It is illustrated in figure 2.3. It can easily be seen that the largest difference in transmission between the two peaks can be found on the steep flanks of the Gaussians. To maximize the difference between two shifted peaks, peaks that are higher and narrower in spectral width should be used.
Figure 2.3: Gaussian transmission peaks around 800 nm (blue solid curve) and 801.11 nm (red dashed curve). The difference is largest on the steep flanks of the peak. This is illustrated by the black solid curve, which shows the difference between the two Gaussians. It has extrema on the steep flanks of the Gaussian curves. The difference graph is scaled up by a factor of 2 to show more detail.
Chapter 3

Experimental

In the previous chapter, we have derived that a layer of an optically nonlinear material on a metallic hole array should yield a measurable nonlinear response in transmission. We will now describe the design and conduction of an experiment that should show that effect. We use a metallic subwavelength hole array on a substrate of an optically nonlinear material, and we measure the transmission through that sample as a function of the intensity impinged onto it.

In the first section of this chapter, the preparation of the samples we used is discussed. In the second and third sections, the nonlinear materials we studied are characterized using two different methods. In the fourth section, the transmission spectra of our samples are measured and explained. In the final section, we describe the experiment devised to show the optical Kerr effect in transmission, and we present the results of that experiment.

3.1 Sample fabrication

Two different kinds of sample were used during this project.

The first consists of a optically thick (200 nm) layer of amorphous Au on a glass substrate (purchased from Ssens, product number 14-2-00). It was spincoated with a thin layer of 3-butoxy-carbonyl-methyl-urethane. This sample will be referred to as the 3-BCMU-sample.

The second consists of a 200nm layer of Ge$_{11.5}$As$_{21.5}$Se$_{64.5}$ (GeAsSe), made with pulsed laser deposition on glass and coated with a 250 nm layer of Au (samples provided by Prof. Luther-Davies of the Australian National University, Au layer evaporated at AMOLF). Henceforth, we will refer to this sample as the GeAsSe-sample.

Using focused ion beam (FIB) milling, 20 × 20 arrays of 225 (W) × 150 (H) nm holes were milled into the Au films (see figure 3.1). The periodicity a of the arrays was varied between 300 nm and 585 nm. The aspect ratio was chosen such that the
transmission is close to an order of magnitude higher than in a similar sample with round holes, due to the hole shape [3]. The FIB system used is an FEI Helios NanoLab 600. The milling resolution of this system is 5 nm.

The spincoating of the 3-BCMU sample was done after milling the holes. The 3-BCMU polymer is a powder at room temperature, and can be solved in chloroform. The 3-BCMU layers were made by spincoating a 10 g/L solution of 3-BCMU in chloroform for 40 s at 3000 rpm. The thickness was measured with a profiler, and is approximately 150 nm. The polymer was kindly provided by Prof. Licchelli of the Università di Pavia.

When trying to mill through GeAsSe, it turned out to be very ‘soft’; the milling depth is difficult to control. In order to obtain a well-controlled sample, the holes in the GeAsSe-sample were milled through both the Au layer and the GeAsSe layer. Furthermore, when milling through a layer of material, redeposition of material that was milled away leads to some deviations from the desired shape and size of the holes. The holes become slightly trapezoidal. Since this deformation will be present in all samples, we will assume that the effects, if measurable, will cancel when comparing data.

In figure 3.2, a cross section of both samples is sketched schematically. In sample 1, the size of the holes is large compared to the average molecule size of 3-BCMU, so we can assume the holes are filled completely. All layers except for the glass are 150-250 nm thick. The glass layer is approximately 1 mm thick. FIB milling leads to a small layer of glass being etched away at the end of each hole. This is estimated to be less than 30 nm.
3.2 Thermal effects

3.2.1 Setup

To study the nonlinearity of the spin-coated 3-BCMU-layers, we set up an interferometric experiment, based on the classical Michelson-Morley experiment (figure 3.3). In this setup, a beam of light is split into two beams, traveling at a right angle with respect to each other. At the end of each of the two paths, a mirror is placed to reflect the beams back to where they were split. A difference in path length between the two arms leads to an interference pattern, that can be imaged with a camera.

One of the mirrors in the interferometer was coated with 3-BCMU. The interferometric pattern was optimized to match the path lengths in the two arms as much as possible. Subsequently, a beam of green light (a diode-pumped solid state laser at 532 nm wavelength), was focused onto the spot where the other laser beam hits the 3-BCMU-coated sample.

![Diagram of Michelson-Morley interferometer with 3-BCMU coating](image)

Figure 3.3: A Michelson-Morley interferometer with a layer of 3-BCMU on one of the mirrors and a green pump beam to induce a change in refractive index in the polymer layer.

3.2.2 Results

If the refractive index of the 3-BCMU-layer changes, the time it takes for a light beam to pass through the 3-BCMU layer changes accordingly. The effective path length $ct$ changes to $c(t + \Delta t)$, and the phase changes accordingly. Hence, changing the refractive index of the 3-BCMU layer will lead to a mismatch in phase in the two branches of the interferometer. That should be readily visible. If the change in refractive index is due to the Kerr effect, it should disappear very quickly after the pump beam that induces the change in refractive index is removed.
Indeed, switching on the green laser leads to an observable change in the interference pattern. However, if the green laser is blocked, the interference pattern needs several camera frames (~100ms) to return to its previous state. This strongly suggests that the effect we see is not an ultrafast optical Kerr effect, but a thermally induced change in refractive index. Since the 3-BCMU polymer absorbs green light very well, it is expected that considerable heating is caused by the pump laser.

![Figure 3.4: A part of the interference pattern before (left) and after (right) switching on the green pump beam. On the spot where the pump beam incides with the (much larger) probe beam, a change in the interference pattern can be seen.](image)

We confirmed that the effect is caused by heating by replacing both beams with a pulsed laser at 810 nm, which has a much higher peak intensity than the green laser but a lower average intensity. The switching of the pump beam had no visible effect on the observed interference pattern.

An equivalent experiment in which the 3-BCMU layer was replaced with a wafer of As$_2$S$_3$ chalcogenide glass, which should have a comparable $\chi^{(3)}$, also revealed no visible change in the interference pattern, neither in the continuous wave nor in the pulsed experiment. Since these glasses have a much lower absorbance than 3-BCMU for the wavelengths we used, this also suggests that the effect we saw in the 3-BCMU layer was completely thermal.

Consequently, we concluded that the bistability demonstrated by Zayats et al. [5] is no evidence of the optical Kerr effect. Instead, given the experimental details of their experiment, it is not unlikely that they see the thermal effect we observed in our interferometric setup.

If the 3-BCMU layer has a measurable Kerr effect, it is likely obscured by the thermal effect in this experiment. In order to distinguish between ultrafast effects and thermal effects, we need to perform an ultrafast, time-resolved experiment.
3.3 Pump-probe measurements

3.3.1 Setup

Since the optical Kerr effect is an ultrafast effect, in order to distinguish it from thermal effects we need an experiment that has a high enough time resolution. The so-called pump-probe technique allows us to measure ultrafast effects due to a short excitation of the nonlinear material. Furthermore, we can use it to find the damage thresholds of the samples.

In the pump-probe experiment we performed, a short pulse of light impinges on a sample under a certain angle. This pump pulse is tuneable in wavelength and energy, and is used to excite the sample and induce effects that only occur at high field strengths. A second (probe) pulse is then applied to the sample on the same spot. It is at least ten times lower in peak intensity than the pump pulse, so that the response to the probe of the material stays linear to a good approximation. The reflectance of the sample is measured with that probe pulse. The probe pulse can also be tuned in wavelength. The delay between the pump and the probe can be varied, thus allowing us to monitor the changes in reflectance of the sample as a function of time.

The measurements were performed at AMOLF, in the photonic bandgaps group. The light is generated by a regeneratively amplified Ti:Sapphire laser (Spectra Physics Hurricane) which drives two optical parametric amplifiers (OPA, Topas). Both OPAs have a continuously tunable output wavelength between 475 and 2600 nm, with pulse durations of 150 fs and a pulse energy of at least 20 $\mu$J. The reflectivity was calibrated by measuring the reflectivity of a gold mirror.

The optical Kerr effect leads to a change in refractive index, and a change in refractive index leads to a change in reflectivity (following Fresnel’s laws). If the $\chi^{(3)}$ of our sample is high enough, this should lead to a dip or a peak in the reflectance versus delay graph measured by the pump-probe experiment.

We performed pump-probe measurements on three different materials that are documented to have a high nonlinear response: a spun layer of 3-BCMU polymer, a thin layer (250 nm) of GeAsSe chalcogenide glass and a thick wafer (2 mm) of As$_2$S$_3$ chalcogenide glass. The goal of this experiment was to find out which of these materials would be the most promising candidate to combine with a hole array to find switching.

3.3.2 Results

Pump-probe measurements were done on three different samples. All of these samples were bare layers of nonlinear material, with no gold film or hole arrays present. The first was a glass wafer spin coated with 3-BCMU, the second a 2 mm thick wafer of As$_2$S$_3$ chalcogenide glass and the third a glass wafer with a 250 nm layer of GeAsSe.
Figure 3.5: Left figure: Reflectivity as a function of the delay between the pump and the probe pulse, on a thin film of GeAsSe. The incident power is 68 GW/cm$^2$.
Right figure: Change in reflectivity as a function of the delay between the pump and the probe pulse, on a thick wafer of As$_2$S$_3$. The incident power is 104 GW/cm$^2$.
Pump and probe pulses overlap where $\Delta t = 0$. Pump wavelength is 1650 nm. The different graphs represent different probe wavelengths. Graphs have been offset with respect to each other.

Chalcogenide glass. The linear refractive indices of these substances are 1.6, 2.4 and 2.6 respectively. Using Fresnel’s equation for light at normal incidence,

$$R = \left| \frac{n_1 - n_2}{n_1 + n_2} \right|^2,$$  \hspace{1cm} (3.1)

where $R$ is the reflectivity, $n_1$ is the linear refractive index of the first medium (air) and $n_2$ of the nonlinear medium, we expect reflectances of 5%, 17% and 20%, respectively. Here we assume that the situation can be approximated with two half-spaces, one of air and one of the nonlinear materials. That assumption is correct for the As$_2$S$_3$ chalcogenide glass, but likely not valid for the thin films of the other samples. The expected reflectances were confirmed for intensities of at least an order of magnitude smaller than the pump pulses we use, before the pump-probe measurements were done.

To approximate the change in refractive index induced by the pump pulses, we measure the change in reflectivity as a function of time. The refractive index of the nonlinear material and the change in reflectivity are related as

$$\Delta R = \frac{\delta R}{\delta n_2} \Delta n_2.$$  \hspace{1cm} (3.2)

In all the experiments, a pump beam with a wavelength of 1650 nm was used. The probe beam was varied between 840 nm and 1140 nm. The intensity of the pump beam was
varied to see how the effects scale and to find the damage threshold.

3-BCMU

When doing pump-probe experiments on 3-BCMU, it quickly became apparent that an irreversible change takes place when the polymer layer is irradiated, even when using only low-intensity probe pulses. Furthermore, repeating measurements on different spots led to different results. We concluded that either the polymer layer is so fragile we cannot use it for this kind of measurement, or the quality of the layer is too low to give consistent data.

Note that the measurements described in section 3.2 were done at peak intensities that were very low compared to the pump pulses used here.

GeAsSe

When pumping the GeAsSe layer with peak intensities up to 40 W/cm², no visible changes in reflection can be seen where pump and probe coincide. At higher intensities, the reflectivity decreases sharply when the pump pulse hits the sample (figure 3.5). This change does not revert during the experimental time frame.

Investigation of the sample after measurements were performed revealed that the GeAsSe layer was damaged. We assume that this damaging takes place immediately after the pump beam first hits the sample, and that the change in reflectivity we see is a free carrier effect in the damaged sample. Chalcogenide glasses are known to crystallize after melting. In the crystalline form of GeAsSe we do not expect appreciable Kerr nonlinearities.

While this experiment allowed us to estimate the damage threshold of this glass rather well, we did not find a measurable nonlinear response. Investigating the validity of 3.1 might shed some light on the effects that do occur.

As₂S₃

Pump-probe measurements on the As₂S₃ wafer showed evidence of an ultrafast response to the pump beam. In the right graph of figure 3.5, a sharp dip can be seen in two of the three graphs. The fact that the peaks are not exactly centered at 0 ps is due to small alignment errors in the setup. The reflectivity recovers within 1 ps from the incidence of the pump pulse, which is fast enough to exclude the possibility of a thermal effect.

At this stage, it is not clear why the graphs look so different for different probe wavelengths. In some nonlinear materials, the change in refractive index due to an incident electric field changes from positive to negative as a function of incident wavelength. If that is the case, the switching wavelength may be close to 840 nm (black curve).
Conclusion

Of the three materials, only As$_2$S$_3$ showed clear evidence of Kerr nonlinearity. Therefore, we initially chose this material to fabricate our samples with. Unfortunately, it turned out to be impossible to create good quality samples on As$_2$S$_3$ with simple fabrication techniques. The thin layers of GeAsSe were better suited to sample fabrication, and literature documents a similar $\chi^{(3)}$ for both chalcogenide glasses. We believe that in the pump-probe experiment the nonlinear response of GeAsSe was obscured. In particular, the thin layer of GeAsSe may have lowered the damage threshold, thus breaking the material before the nonlinear response became measurable.

Sample fabrication on As$_2$S$_3$ would also be possible, using different fabrication techniques. If it turns out that the GeAsSe samples do not yield measurable nonlinearity, it is advisable to make As$_2$S$_3$ samples.

3.4 White light spectra

3.4.1 Setup

To characterize the hole arrays on the GeAsSe sample, we measured the transmission spectrum through each of them. We were looking for hole arrays that have interesting features in the transmission spectrum in the wavelength range of the pulsed laser. In order to see any nonlinear effects, we need features in the spectrum that will change quickly with a small change in refractive index.

The transmission setup (figure 3.6) uses a spectrograph in combination with a thermoelectrically cooled CCD camera to obtain measurements with a very low instrumental error. A Xe arc lamp is focused on the sample as a light source. This lamp yields a broad and stable spectrum to characterize the transmission of the arrays.

3.4.2 Results

In figure 3.7, the white light transmission spectra as measured through the two arrays we chose is shown. The measured spectrum was divided by the spectrum of a reference hole of $10 \times 10 \, \mu m$, to normalize it with respect to the spectrum of the lamp.

We can use eqs. 2.11 and 2.12 to attribute the features in these spectra to the theory described earlier. Since the intensity of the white light source is low, we may assume that the refractive index of the GeAsSe chalcogenide glass can be safely approximated by its linear refractive index, 2.6. Substituting $\varepsilon_{d,1}=1$ (vacuum) and $\varepsilon_{d,2}=2.6$ (chalcogenide), and using data for the wavelength-dependent $\varepsilon_m$ of gold that was measured on gold coatings at the University of Twente, we can predict Wood’s anomalies.

We can now attribute the minima in the transmission spectrum to Wood’s anomalies. The peaks occur to the red side of Wood’s anomalies. The minima that are important
in this spectral range are the \((l, m) = (1,0), (0,1)\) and \((1,1)\) minima of the metal-GeAsSe interface. The \((1,0)\) and \((0,1)\) minima overlap, and are designated by \((1,0)\) in figure 3.7.

Based on the spectra we measured in this setup, we selected two arrays that are interesting for nonlinear transmission measurements. The first (blue line in fig. 3.7) has a Wood’s anomaly close in the spectral range of the laser (between 780 nm and 840 nm). The second (red line in fig. 3.7) features a sharp flank in that same wavelength range.

The reason for choosing the first array is that while we can attribute the peaks in the spectrum to the Wood’s anomaly closest to them, there are effects in the array that cause the peaks to broaden or to shift. While we can be sure that a change in the refractive index of the interface material will have an effect on the shape and position of the peaks, we cannot predict that effect precisely. The Wood’s anomalies, on the other hand, only depend on the periodicity and the refractive indices in the sample. Hence, in a sample where our laser wavelength is usually in a Wood’s anomaly of the spectrum, we are sure that something will happen when the refractive index of the nonlinear material changes.

The reason for choosing the second array is that on a flank, the transmission changes most rapidly if the refractive index of the nonlinear material changes. Although it is not possible to predict very accurately what will happen to the shape and position of a peak when the refractive index of the substrate material changes, this position should at least yield large effects.

### 3.5 Transmission as a function of intensity

If the refractive index of a nonlinear material changes as a function of the intensity of the electromagnetic field and the transmission spectrum of a hole array shifts as a function of the refractive index of the interface materials, we can devise a simple experiment to
Figure 3.7: Normalized transmission through hole arrays with hole sizes $225 \times 150$ nm, with periods of 540 nm (blue solid line) and 495 nm (red dashed line). Both hole arrays are on the GeAsSe sample. The vertical dashed lines designate the wavelength range of the laser.

measure this effect.

Consider a metallic hole array between two sheets of a material with a dielectric constant $\varepsilon_1$. The transmission of this array for a laser pulse centered around $\lambda=800$ nm is $T_1$. Now we replace this material with a different material, with dielectric constant $\varepsilon_2$. The new transmission is $T_2$.

In our experiment, we can alter the dielectric constant of our nonlinear material by increasing the intensity of the light we use. At low intensities, the nonlinear material will behave like material 1, while at high intensities it behaves as material 2. The percentage of transmitted light will change as a function of the intensity, leading to a transmission curve that is not linear as a function of the intensity.

The intensity-dependence of the transmission through our sample is the effect we are looking for in the final experiment, described in this section.

### 3.5.1 Setup

The measurements aimed at finding nonlinearities were done using a pulsed mode-locked Ti:Sapphire laser (Spectra Physics Tsunami) with a pulse width of approximately 100 fs and a bandwidth of 10 nm, around an operational wavelength between 780 nm and 840 nm. The intensity of the beam can be varied using two neutral density filter
wheels. The intensity was continuously monitored with a power meter by splitting off a small part of the beam.

The setup used to measure the transmitted power is similar to the setup used in the white light transmission measurements (see figure 3.6). The measured spectra were integrated to yield the transmitted power.

Having chosen suitable arrays, the white light source was replaced by a pulsed laser beam. From the pump-probe experiments, we estimated the damage threshold of Ge-AsSe to be around 40GW/cm$^2$. In order to see as high a nonlinear response as possible, we ramped up the intensity of the pulsed laser from the lowest measurable intensity to peak intensities close to the damage threshold.

Using an objective (NA 0.25) to focus the beam on the array, we obtained a lower size limit of our focus of 2 $\mu$m $\times$ 2 $\mu$m. That focus is smaller than our sample, illuminating approximately 20 holes. That is sufficient to excite SPPs on the hole array and induce extraordinary transmission.

The maximum incident power we used was 250 mW, at peak wavelengths of 800 and 810 nm. Using the pulse duration (100 fs) and the repetition rate of the laser (80 MHz), and assuming a focus size of $(2\mu m)^2$, we can estimate the power per area impinging on the nonlinear material. In each second, there are $80 \cdot 10^6$ pulses of $100-10^{-15}$s, so there is light for $8 \cdot 10^{-6}$ s during each second. In those pulses, the peak power is $250 \text{ mW} / 8 \cdot 10^{-6}=31.25 \text{ kW}$. If we focus that power on a $(2\mu m)^2$ focus, we find a peak intensity of 781 GW/cm$^2$. That is much higher than the damage threshold of GeAsSe. If the light interacts with the GeAsSe, we should be able to at least see the effects of damage to the glass layer. If there are any nonlinear effects that can be measured, they should also be well within range of the laser power.

### 3.5.2 Results

For a wide range of intensities, we measured the total transmission through both arrays. We did this by measuring the spectrum of the pulse with a spectrometer, while making sure the power hitting the spectrometer never got high enough to damage the spectrometer, by attenuating the light with neutral density filters. The spectra were then corrected for the filters and the integration time, and summed to yield the total intensity. The results of two of such series of measurements are shown in figures 3.8 and 3.9, for a slightly focused laser beam and for a sharply focused laser beam respectively.

It is clear that the noise level makes it hard to say whether a nonlinear fit is better than a linear one. The noise from the spectrometer is very low, so the error must be mostly attributed to experimental constraints, such as misalignments in the setup, laser fluctuations and dust particles. If we compare the measured pulse shape of high-power transmitted pulses to low-power pulses, no shape differences can be detected, suggesting that no measurable nonlinear processes occur. Similar results were obtained for different
Figure 3.8: Transmission intensity as a function of laser power for a hole array with a period of 540 nm. Laser pulses with a spectral width of 10 nm, centered around 800 nm, were used to measure this data. A lens with a focal length of 35 mm was used to focus the laser power on the sample.

laser wavelengths and different arrays, suggesting that one or more of the criteria for seeing a nonlinear effect on the transmission through these arrays were not met.

The black arrows in figure 3.8 point at the places where the measured graph shows jumps in transmission. Those places coincide with the changing of the neutral density filter used to filter the transmitted intensity before it hits the spectrometer. It is very likely that those jumps are caused by an error in the calibration of the neutral density filters placed before the spectrometer. Unfortunately, time constraints did not allow a precise calibration of all the filters in the setup.
Figure 3.9: Transmission intensity as a function of laser power for a hole array with a period of 495 nm. Laser pulses with a spectral width of 10 nm, centered around 800 nm, were used to measure this data. An objective with NA 0.25 was used to focus the laser power on the sample.
Chapter 4

Conclusions

The experiment we carried out had suitable parameters and a high enough resolution to show nonlinear effects in our GeAsSe-sample. Nevertheless, when comparing the magnitude of what we expected to measure with the errors we may have introduced through sample fabrication, alignment and analysis, we cannot conclusively state whether nonlinear effects were present in the experiments or not. If anything interesting took place in the experiment, it may well have been obscured by the measurement errors.

By eliminating the factors we address in this chapter, a new experiment may be devised, that should be able to shed more light on the presence and magnitude of nonlinear effects in this structure.

Improved sample design

So far, we have assumed a metallic hole array covered with a homogeneous slab of optically nonlinear material of infinite thickness. However, the sample we used (sample 2 in figure 3.2) is a gold hole array on a substrate with the same periodic corrugations as the array. The holes in the substrate may influence the behaviour of the sample.

We characterized the arrays by measuring white light spectra through each of them. While we did find spectra with interesting features in the frequency domain of our laser, the peaks there were rather broad and low in transmission. That could be optimized.

- The major difference between our sample and the ideal sample is the fact that the focused ion beam (FIB) milled through the metal and the GeAsSe chalcogenide glass. The reason for this difference, as mentioned before, is the considerable ‘softness’ of GeAsSe to FIB milling. The SPPs that are responsible for extraordinary transmission, existing on the interface of gold and GeAsSe, now need to be supported by two different kinds of dielectric.

We cannot predict what sort of effects, if any, this change in sample design has on the transmission through the arrays. However, it would be instructive to study the
differences in transmission through these samples and samples on a homogeneous substrate.

- While we had a wide enough variety in transmission spectra to choose samples that have interesting features at our laser wavelength, there were none that had very high peaks or steep flanks in the right wavelength range. This is due to the choice of parameters when designing the samples. If samples had been available with peaks that are an order of magnitude higher or narrower, implying flanks that are an order of magnitude steeper, any nonlinear effects would have been much more pronounced and likely easier to measure. Based on the white light spectra measured through the various arrays on the Ge-AsSe sample, we know that a larger periodicity than the ones we used will be necessary. A period of around 650 nm should yield a suitable transmission spectrum.

**Improved error reduction**

In our data (see figures 3.8 and 3.9) we have found that the spread of the points is much larger than can be expected from the precision of the spectrometer. The detection error should be equal to $\sqrt{n}$, where $n$ is the number of detector counts, which is so small the error bars would hardly be visible. Apparently, there are other sources of errors in the experiment, which might obscure any nonlinear effects.

- Due to the sharp focusing used to obtain a high enough power to measure nonlinear effects, the experiment became very susceptible to small changes in alignment. Simply touching the setup table was sometimes enough to obscure transmission almost completely. Because of this sensitivity, we assume that small changes in alignment, for example those induced by changing filters, may already have large effects on the measured transmission. If a more robust and better alignment can be obtained, the measurements should be more stable to changes like these.

- Before analyzing the data, we characterized the transmission through the neutral density filters we used, using a power meter. It was clear that there were some deviations from the documented attenuation factors, likely due to small wavelength dependences. The calibrations we did were not very precise, however. If the filters are calibrated with a more sensitive method, for example the camera of the spectrometer, their effects on the intensity and the measured transmission can be corrected for better. This should also decrease the error margins of the measurements significantly.

- A differential measurement would allow direct comparison of the laser beam and the transmission of that beam through a hole array. A laser beam can be split into
two paths, leading to a pair of calibrated photodiodes that measure their intensity. If one of the beams is sent through a pinhole and the other goes through a hole array on a nonlinear substrate, the nonlinear effects can be monitored at the same time as the laser fluctuations. That would allow us to correct for laser fluctuations.
Chapter 5

Recommendations

- A sample can be made that has a steep flank in the transmission spectrum near the laser wavelength. A much steeper flank than the one we measured on should yield a much higher nonlinear response, that will be easier to measure. Making samples with a larger period than the ones we used should shift the high peak we now see at around 650 nm further into the red, yielding a more optimal transmission spectrum.

- To obtain samples in which the layer of nonlinear material is left intact, other fabrication methods should be explored. A good candidate for high-resolution sample fabrication is electron beam lithography with liftoff. Such a method will make it possible to deposit a gold hole array on a layer of nonlinear material without the risk of melting.

- As mentioned before, more robust alignment of the setup would allow more precise measurements. Investigating the effect of filters could also shed some light on the large errors. Perhaps a gradual neutral density filter, that can be adjusted to change the intensity that gets through, would yield a better result than a filter wheel. Also, assuring that the focus is smaller than the sample and making sure that the focus is in the middle of the sample instead of on the edge should make the alignment more robust to small disturbances.

- Heterodyne detection, e.g. mixing the output signal with a signal of known frequency and observing the difference frequency, would allow the detection of even very small nonlinear changes in transmission as a function of intensity. However, if the nonlinear changes are so minute that such a detection scheme is necessary to find them, the system is likely to be unsuitable for optical switching.

- Replacing the pulsed laser by a continuous wave laser, where pulses are created by placing a chopper in the beam, could also shed new light on the behaviour of the sample.
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Bibliography


