Near unity collection efficiency of light from single photon sources in planar antenna structures

Thomas Esselink
15035751

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The Hague University of Applied Sciences
Institute AMOLF
Superior: Prof dr A. Femius Koenderink AMOLF
           Ing. M. Kamp AMOLF
           J. H. R. Lambers HHS
           R. Buning HHS
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Preface

This report contains the work that I carried out for the last four months as part of my internship in Applied physics at AMOLF. The realization of this project has been a collective undertaking of many people that assisted in very diverse ways. I would like to thank Femius Koenderink and Marko Kamp for your time and trust. From the start I knew I have a lot to learn from both of you. Thanks to Dimitry Lamers for the help in the NanoLab Amsterdam. Thanks to the Resonant Nanophotonics group for the interesting discussions and meetings. Thanks to Jan Lambers for your time and your advice.

Thomas Esselink
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Abstract

A main challenge in single emitter microscopy is that it is difficult to efficiently collect all the emitted light from single photon sources. Due to the dipole nature of the single photon sources, it is hard to measure the brightness of these molecules in a calibrated manner, since the light is not emitted isotopically. Recent studies have shown however, that collection efficiencies of 96% for can be reached for single photon sources. In this report, a fabrication recipe is described for a planar antenna structure that should, in theory, also reach these collection efficiencies for single photon sources. The angular distribution of the radiated power from these emitters are measured with a Fourier microscope. Firstly, the Fourier microscope will be characterized. After this, experimentally measured radiation patterns of the planar antenna structure will be compared to calculated radiation patterns. The Fourier microscope is correctly characterized, resulting in quantitative comparisons between theory and measurements. However, the beaming effect of the planar antenna structures is not observed. Nevertheless, promising results are obtained that show an increase in collection efficiency between the emitters which are embedded in such structures and emitters which are embedded on a normal glass substrate.
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1. Introduction

The research institute where I am attending my internship is called AMOLF. It is a part of the institutes organisation of NWO. [1] The institute was established on 15 September 1949 and it is located in Amsterdam. The goal of AMOLF is to carry out leading fundamental research on the physics of functional complex matter. The research gives understanding about innovative functional materials that could supply solutions to societal challenges. AMOLF is divided into four different research programs: Nanophotonics, Nanophotovoltaics, Designer matter and Living Matter. These research programs are divided into several research groups. This internship is done in the research program of Nanophotonics and the research group is called Resonant Nanophotonics. This group studies the interaction of light and matter on the nanoscale, where structures can be made with a size in the same order of magnitude as the wavelength of light. [2]

Single photon emitters – i.e., single molecules or quantum dots that emit single photons at a time – are useful tools in the Resonant Nanophotonics group.

A main challenge in single emitter microscopy is that it is difficult to efficiently collect all the emitted light. To first approximation, a single emitter is so much smaller than the wavelength that is must be an omnidirectional source, meaning that only a small fraction of radiation can be captured by a lens. In fact, the dipole nature of emitters makes that the light is not quite emitted isotropically from the source but forms a Hertzian radiation dipole pattern. This makes the amount of photons that can be collected by a microscope objective highly dependent on the orientation of the dipole. While transmission and detection efficiencies of every part of our setups can be measured this uncertainty in dipole orientation makes it impossible to measure brightness of emitters in a calibrate manner.

Recent studies [3] [4] [5] [6] have shown that planar antennas can be used to modify the radiation patterns of emitters in such a way that the photons are emitted in a narrow cone, for instance towards the objective lens in a microscope. It has been shown that with a combination of an antenna structure and an objective lens with high numerical aperture (1.45) up to 99% of the emitted light can be collected. This makes it possible to measure the brightness of these molecules. Another advantage of this technique is that the brightness of measurements done with lower NA objectives can be greatly improved. This is especially useful in situations where longer working distances are required, for instance when measuring on single molecules in a cryostat.

The project will be divided in several parts. This results in a clear approach towards the project. First of all, a planar antenna structure has to be designed and manufactured. This will be realized by using a Matlab code. The Matlab code calculates the radiation pattern for a emitter in a multilayer structure. The thicknesses and materials of the multilayers can be implemented and optimized for an optimal collection efficiency. After fabricating the optimal structure found by theory, the profile of the layers can be measured. These measurements should confirm that the desired thicknesses are obtained.

With the sample design and fabrication finished, the characterization of the set-up follows. The set-up used is a Fourier microscope. This is a microscope that allows us to measure both the real space and the angular response, also called Fourier space, of any sample. Both spaces will be characterized to ensure reliable measurements are acquired for the fabricated structures.

After this, radiation patterns of the planar antenna structures can be measured. First off, simplified structures embedded by emitters with an emission wavelength peak at 655 nm will be investigated. The
same is done for simplified structures embedded by emitters with an emission wavelength peak at 800 nm. The influence of the excitation wavelength and a protection layer for the emitters will be discussed also. Since the radiation pattern of an emitter depends on the height at which it is stationed, comparisons can be made with theory. Finally, radiation patterns for planar antenna structures will be measured and compared to theory.
2. Theory

This chapter presents the theoretical foundations of the project. First of all, the theory of an electric dipole is explained. After this, the interference of light and the transfer matrices of thin layers are explained and the basics of Fourier optics is explained. The information of these sections combined gives an idea of how a Matlab code can calculate the radiation pattern in a multilayer structure. In order to validate the code, a comparison is made with radiation patterns found in literature. Finally, a planar antenna structure is designed that will beam the light coming from a dipole and thus increasing the collection efficiency. Furthermore, a short introduction into Fourier optics will be given. This is necessary for understanding the basics of the set-up.

2.1. Electric dipoles

First of all, the theory of an electric dipole in an unbounded medium is introduced. This indicates how a dipole will emit its light in such a medium, which makes it clear why it is hard to efficiently collect the light. For this reason, a mirror is introduced and the dipole is placed in front. This will reflect a part of the light coming from the dipole, as a result a higher collection efficiency can be achieved. Therefore, it is interesting to investigate the radiation pattern that will occur after placing it in front of a mirror for making a structure design.

2.1.1. Dipoles in an unbounded medium

The combination of two electric charges with equivalent quantity but opposite sign on a certain distance from each other is called an electric dipole. [7] Electric dipoles can be described by their electric dipole moment. The electric dipole moment can be considered as a vector and is given by Eq. 1. The direction of the vector is from the negative charge towards the positive charge.

\[ \vec{p} = Q \vec{l} \]

where \( \vec{p} \) is the dipole moment in Coulomb meter, \( Q \) is the net charge on each atom in Coulomb and \( \vec{l} \) is the separation distance between each atom in meters. Numerous molecules have dipole moments since the distribution of the electrons is not uniform across the atoms. This is due to the difference in electronegativity for two atoms. An example is the water molecule, see Figure 1. Due to the non-uniformity of the electron density, electrons will spend more time at the oxygen atom then at the hydrogen atom, making the oxygen atom negatively charged and the two hydrogen atoms positively charged. Molecules with such a dipole moment are called polar molecules.
Figure 1: The electric dipole moment shown in a water molecule. The resulting dipole moment $\vec{p}$ is the vector sum of the two dipole moments, $\vec{p}_1$ and $\vec{p}_2$. [7]

Obviously, the electric dipole formed by the atoms of a molecule will induce an electric field. A way to visualize an electric field is by using a set of lines which gives the direction of the electric field for numerous points in space. These lines are called electric field lines. For a point dipole, these electric field lines will appear as shown in Figure 2. A point dipole is a dipole where the distance between the charges is so small in comparison with the working distances. Therefore, a molecular dipole can be interpreted as a point dipole when working on larger scales.

There are several types of dipoles that can occur within a molecule. One of these types is the transition dipole moment, which occurs in fluorophores, a name used to indicate any chemical compound that fluoresces. The transition dipole moment is formed due to a transition between two states. This transition takes place when molecules get excited by electromagnetic waves. Shortly after the excitation of the fluorophore by absorbing light with a specific wavelength, it will re-emit the light with longer wavelengths. From Figure 2 follows that, due to the dipole nature of a molecule the light is not quite emitted isotropically from the source but rather forms a Hertzian dipole radiation pattern. [8] Therefore, the direction of the emitted light highly depends on the orientation of the dipole.

Figure 2: The electric field lines for a point dipole. [9]
2.1.2. Electric dipoles near a conducting plate.

In the previous section, an electric dipole in an unbound medium has been analyzed. Now, the performance of an electric dipole in front of an infinite perfect conductor will be studied. To study this, image carriers will be introduced. [10] [11] Image carriers are the mirror image of the real carriers and are therefore imaginary sources. The real carriers and the image carriers will form an equivalent system. Now, let’s suppose that a fluorophore has a vertical dipole with a certain spacing $h$ in meters with an infinite perfect conductor, depicted in Figure 3. The energy emitted from the dipole is determined by the properties in its unbounded state. For a certain point in space, in Figure 3 called $P_1$, a direct wave from the dipole will arrive there. Besides the direct wave, a reflected wave from the source arrives at point $P_1$. If the line is extended beneath the conducting plate, it appears to come from the imaginary dipole. Another point is shown, called $P_2$. The wave from the actual source reflects at point $R_2$ and travels further to point $P_2$. Again, the line gets extended and it appears to come from the same virtual source. This works for every other point in the space above the conducting plate.

Figure 3: A vertical dipole above a perfect conducting plate. With the method of image charging, it can be shown that the reflected waves seem to originate from the same virtual source. [11]

With the graphical representation of a vertical dipole near a perfect conductor from Figure 3, it is clear that the direct and reflected component of the electric field will interfere with each other.
The following equations, Eq. 2 and Eq. 3, for the distances can be introduced by using the cosine rule [11]:

\[ r_1 = \sqrt{r^2 + h^2 - 2rh \cos(\theta)} \]  \hspace{1cm} \text{Eq. 2}
\[ r_2 = \sqrt{r^2 + h^2 - 2rh \cos(\pi - \theta)} \]  \hspace{1cm} \text{Eq. 3}

where \( r \) is the radial distance in meters with respect to the origin, \( r_1 \) is the radial distance for the direct electric field component in meter, \( r_2 \) is the radial distance for the reflected electric field component in meters and \( \theta \) is the polar angle in degrees. Eq. 2 and Eq. 3 can be rewritten since it is a far-field representation, so \( r \gg h \), and by using the binomial expansion, resulting in Eq. 4 and Eq. 5 [11]:

\[ r_1 = r - h \cos(\theta) \]  \hspace{1cm} \text{Eq. 4}
\[ r_2 = r + h \cos(\theta) \]  \hspace{1cm} \text{Eq. 5.}

So, the pattern and the amplitude of the electric field not only depends on the field of the dipole but also on its height and the viewing angle, resulting from Eq. 4 and Eq. 5. This is also depicted in Figure 5. The height of the dipole is varied and it is plotted versus the viewing angle. Clearly, both the height and the viewing angle influence the radiation pattern coming from the vertical dipole. So, Figure 2 and Figure 5 show that when a dipole is placed in front of a mirror, the radiation pattern changes, as expected. More light is radiated into the upper half, meaning that the collection efficiency is increased. Therefore, placing the dipoles near a mirror, will be interesting for a structure that needs to act as an optical antenna.
2.2. Dielectric films

So, from 2.1. Electric dipoles, it follows that the collection efficiency increases when a dipole is placed in front of a mirror. However, metal mirrors do not have perfect reflection at optical frequencies, where metals are not perfect conductors. Nevertheless, the idea that radiation patterns of emitters can be optimized by planar structures does extend to optics, if the dipole is embedded in a stack metallic and dielectric films. To appreciate qualitatively how multilayer films can redirect light, it is useful to examine the basic concepts of interference in dielectric films. First, the basic concepts of interference in dielectric films are introduced. Then, the transfer matrix is introduced to analyze the propagation of electromagnetic waves through a multilayer. With these two ingredients, the performance of a multilayer stack can be investigated and defined.

2.2.1. Interference in dielectric films

The common appearance of iridescence on the surface of soap bubbles and oily water are a result of the interference of light in single or multiple thin surface layers of transparent media. [12] These thin layers are also called thin films and have thicknesses in the sub-wavelength range. What kind of interference occurs depends on a variety of parameters, such as wavelength and film thickness. Suppose that there is a transparent film on top of substrate as shown in Figure 6. When the light beam hits the upper surface of the film at point A, a part is transmitted and a part is reflected. The transmitted light beam hits the film-substrate interface at point B and it will once again be either transmitted or reflected. The refracted beam leaves at point C and will travel parallel to the refracted beam at point A. However, a part of the light incident at C also reflects which is not shown in Figure 6. The refracted beam encounters numerous reflections within the film, until the irradiance of the beam eventually becomes zero.
Figure 6: Transparent film on top of a substrate, not shown in this figure. From this schematic, the optical-path difference can be determined between the two reflected waves on the boundaries of the film. [12]

From Figure 6 conditions for constructive and destructive interference can be derived. The interference is caused by the phase difference between point C and D. The phase difference between the two beams is due to the additional distance covered in the film, the so called optical-path difference. Constructive interference corresponds to a phase difference of $2\pi m$ and destructive interference corresponds to a phase difference of $(2m + 1)\pi$, where $m$ is an integer. [13] The beam in the film travels an extra distance ABC while the reflected beam travels a distance of AD. After the beams reach point C and point D, they will continue to travel parallel with respect to each other. The optical-path difference between the two beams is given by Eq. 6,

$$\Delta_p = n_f(AB + BC) - n_0(AD) \quad \text{Eq. 6.}$$

Where $\Delta$ is the optical-path difference in meters, $n_f$ is the refractive index of the film, $n_0$ is the refractive index of the external medium and AB, BC and AD are distances in meters. Eq. 6 can be rewritten as Eq. 7.

$$\Delta_p = 2n_f t \cos(\theta_t) \quad \text{Eq. 7}$$

where $t$ is the thickness of the film in meters and $\theta_t$ is the angle of refraction in degrees. The total optical-path difference is not only the result of the extra distance traveled. Reflections that occur on the boundaries of the layers introduce a phase shift and will therefore affect the total optical-path difference. [14] Thus, the total optical-path difference can be written as Eq. 8.

$$\Delta = \Delta_p + \Delta_r \quad \text{Eq. 8}$$

where $\Delta$ is the total optical-path difference in meters and $\Delta_r$ is the optical-path difference caused by the phase change on reflection in meters. The net phase difference that occurs can be calculated by Eq. 9.

$$\delta = k\Delta \quad \text{Eq. 9}$$

where $\delta$ is the net phase difference in radians. The wavenumber $k$ is better known as Eq. 10,
As mentioned before, when the net phase difference is equal to $2\pi m$, the light will constructively interfere and when the net phase difference is equal to $(2m + 1)\pi$, the light will destructively interfere. So, general conditions for constructive interference and destructive interference can be derived by using Eq. 8 and Eq. 9. In the case of constructive interference, Eq. 8 and Eq. 9 will result in Eq. 11, which is the general condition for this type of interference.

$$\frac{2\pi m}{\lambda} = \Delta_p + \Delta_r$$  \hspace{1cm} Eq. 11

A general condition for destructive interference can be determined in a similar way, this results in Eq. 12.

$$\frac{(2m + 1)\pi}{\lambda} = \Delta_p + \Delta_r$$

$$\left(m + \frac{1}{2}\right)\lambda = \Delta_p + \Delta_r$$  \hspace{1cm} Eq. 12

From Eq. 11 and Eq. 12 follows that one can achieve constructive interference or destructive interference by changing the wavelength for instance. Furthermore, the angle of refraction, the refractive index of the film, the refractive indices of the materials surrounding the film and the thickness of the film all affect the type of interference, this follows from Eq. 7. Therefore, the film can be designed in such a way, that you either have maximum reflectance, constructive interference, or maximum transmittance, destructive interference. In the case of destructive interference, the reflected waves are precisely out of phase which leads to elimination of the reflected light and thus maximizes the transmittance of the light. In the case of constructive interference, obviously, the opposite occurs.

### 2.2.2. Transfer matrix

Usually, the amplitude of the reflection and transmission coefficients of light on a single interface is described by the Fresnel equations, Eq. 13 - Eq. 16 [14]:

\[
\begin{align*}
    r_s &= \frac{E_{r,s}}{E_{0,s}} = \frac{\cos(\theta_i) - \sqrt{n^2 - \sin(\theta_i)^2}}{\cos(\theta_i) + \sqrt{n^2 - \sin(\theta_i)^2}} \\  \\
    t_s &= \frac{E_{t,s}}{E_{0,s}} = \frac{2\cos(\theta_i)}{\cos(\theta_i) + \sqrt{n^2 - \sin(\theta_i)^2}} \\  \\
    r_p &= \frac{E_{r,p}}{E_{0,p}} = \frac{\sqrt{n^2 - \sin(\theta_i)^2 - n^2 \cos(\theta_i)}}{\sqrt{n^2 - \sin(\theta_i)^2 + n^2 \cos(\theta_i)}} \\  \\
    t_p &= \frac{E_{t,p}}{E_{0,p}} = \frac{2\cos(\theta_i) n}{\sqrt{n^2 - \sin(\theta_i)^2 + n^2 \cos(\theta_i)}}
\end{align*}
\]

where $r_s$ is the reflection coefficient for s-polarized light, $t_s$ is the transmission coefficient for s-polarized light, $r_p$ is the reflection coefficient for p-polarized light, $t_p$ is the transmission coefficient for p-polarized light, $E_{r,s}$ the reflected electric wave for s-polarized light in volts per meter, $E_{t,s}$ the transmitted electric wave for s-polarized light in volts per meter, $E_{r,p}$ the reflected electric wave for p-polarized light in volts per meter, and $E_{t,p}$ the transmitted electric wave for p-polarized light in volts per meter.
wave for s-polarized light in volts per meter, $E_{r,p}$ the reflected electric wave for p-polarized light in volts per meter, $E_{t,p}$ the transmitted electric wave for p-polarized light in volts per meter, $E_{0,s}$ the incident electric wave for s-polarized light in volts per meter, $E_{0,p}$ the incident electric wave for p-polarized light in volts per meter, $\theta_i$ is the angle of incidence in degrees and $n$ is the ratio of the refractive indices $n_2/n_1$, where $n_2$ is the refractive index of medium 2 and $n_1$ is the refractive index of medium 1. These coefficients however, do not instantly provide an answer for the fraction of power transmitted or reflected from the incident wave, while this information is often necessary. The fraction of power reflected is given by Eq. 17 and Eq. 19 and the fraction of power transmitted is given by Eq. 18 and Eq. 20:

\[ R_s = |r_s|^2 \quad \text{Eq. 17} \]
\[ T_s = n|t_s|^2 \quad \text{Eq. 18} \]
\[ R_p = |r_p|^2 \quad \text{Eq. 19} \]
\[ T_p = n|t_p|^2 \quad \text{Eq. 20} \]

where $R_s$ is the reflectance for s-polarized light, $T_s$ is the transmittance for s-polarized light, $R_p$ is the reflectance for p-polarized light and $T_p$ is the transmittance for p-polarized light. For unpolarized light, a numerical mean can be taken. In this case, the total reflectance turns into Eq. 21 and the total transmittance turns into Eq. 22.

\[ R = \frac{1}{2}(R_s + R_p) \quad \text{Eq. 21} \]
\[ T = \frac{1}{2}(T_s + T_p) \quad \text{Eq. 22} \]

where $R$ is the total reflectance and $T$ the total transmittance. Due to conservation of energy, the total reflectance and the total transmittance must add to unity, described in Eq. 23:

\[ R + T = 1 \quad \text{Eq. 23} \]

So, the Fresnel coefficients can be calculated for every interface the light beam undergoes. However, when a multilayer stack of thin films is used, calculating the total reflectance can get real time-consuming. For this reason, the transfer matrix is introduced. The transfer matrix represents the multilayer stack and analyzes the propagation of electromagnetic waves. [15] The matrix treats all the beams in a film, transmitted and reflected, as summed, rather than looking individually at the reflected and transmitted beams in the film. These summed up beams fulfill the boundary conditions that follow from Maxwell’s equations. The boundary conditions state that magnitudes of the electric and magnetic field are continuous across the boundaries of the media. In Figure 7, a similar case is depicted as in Figure 6. However, Figure 7 illustrates how the electric field and the magnetic field from a light beam behave when incidence on a single thin film. The transfer matrix is given by Eq. 24:

\[
\begin{bmatrix}
E_a \\
B_a
\end{bmatrix}
= \begin{bmatrix}
\cos(\delta) & i\sin(\delta) \\
\sqrt{\gamma_1} \sin(\delta) & \gamma_1 \cos(\delta)
\end{bmatrix}
\begin{bmatrix}
E_b \\
B_b
\end{bmatrix}
\]

\[ \text{Eq. 24} \]

where $E_a$ is the electric field at interface a in volts per meter, $E_b$ is the electric field at interface b in volts per meter, $B_a$ is the magnetic field at interface a in Tesla’s, $B_b$ is the magnetic field at interface b in Tesla’s and $\gamma_1$ is a variable given by $\gamma_1 = n_f \sqrt{\varepsilon_0 \mu_0} \cos(\theta_{t1})$ with $\varepsilon_0$ the vacuum permittivity in farads per meter, $\mu_0$ the vacuum permeability in henries per meter, $n_f$ the refractive index of medium 2 and $\theta_{t1}$ the angle of incidence in degrees.
\( \mu_0 \) the vacuum permeability in Henry per meter and \( \theta_{t1} \) the angle of reflection of the first interface in degrees. The 2 x 2 matrix given by Eq. 24 represents an individual interface of a film layer. So when more layers are present, all individual 2 x 2 matrices can be multiplied, resulting in a transfer matrix that represents the whole stack of layers, shown in Eq. 25.

\[
\begin{bmatrix}
E_a \\
B_a
\end{bmatrix} = M_{1}M_{2}...M_{N}\begin{bmatrix}
E_N \\
B_N
\end{bmatrix} \tag{Eq. 25}
\]

where \( M_{1} \) is representing the transfer matrix for the first layer, \( M_{2} \) is representing the transfer matrix for the second layer, \( M_{N} \) is representing the transfer matrix for the Nth layer, \( E_{N} \) is the electric field at the back interface of the Nth layer in volts per meter and \( B_{N} \) is the electric field at the back interface of the Nth layer in Tesla’s. Now, Eq. 24 will be rewritten in such a way, that the matrix elements of the 2 x 1 matrix only contain the electric field. This can be done because the electric and magnetic field are related by Eq. 26.

\[
B = n\sqrt{\varepsilon_0 \mu_0}E \tag{Eq. 26.}
\]

where \( B \) is the magnitude of the magnetic wave in Tesla, \( n \) is the refractive index of the material where the electromagnetic wave moves through and \( E \) is the magnitude of the electric wave in volts per meter.

Furthermore, the elements of the 2 x 2 matrix will be denoted as a variable, resulting in Eq. 27:

\[
\begin{bmatrix}
E_0 + E_{r1} \\
\gamma_0(E_0 - E_{r1})
\end{bmatrix} = \begin{bmatrix}
m_{11} & m_{12} \\
m_{21} & m_{22}
\end{bmatrix}\begin{bmatrix}
E_{t2} \\
\gamma_s E_{t2}
\end{bmatrix} \tag{Eq. 27}
\]

where \( E_0 \) is the incident electric field in volts per meter, \( E_{r1} \) is the reflected electric field of the first interface in volts per meter, \( E_{t2} \) is the transmitted electric field at the second interface in volts per meter, \( \gamma_0 \) is a constant given by \( \gamma_0 = n_0\sqrt{\varepsilon_0 \mu_0} \cos(\theta_1) \) with \( n_0 \) the refractive index of the medium surrounding the film and \( \gamma_s \) is a constant given by \( \gamma_s = n_s\sqrt{\varepsilon_0 \mu_0} \cos(\theta_{t2}) \) with \( n_s \) the refractive index of the substrate and \( \theta_{t2} \) the angle of reflection of the second interface in degrees. The variables \( m_{11}, m_{12}, m_{21} \) and \( m_{22} \) match with the matrix elements from Eq. 24. Eq. 27 can be written as two equations, Eq. 28 and Eq. 29:

\[
E_0 + E_{r1} = m_{11}E_{t2} + m_{12}\gamma_s E_{t2} \tag{Eq. 28}
\]

\[
\gamma_0(E_0 - E_{r1}) = m_{21}E_{t2} + m_{22}\gamma_s E_{t2} \tag{Eq. 29}.
\]

Eq. 13 - Eq. 16 show that the reflection and transmission coefficient both are defined as a reflected or a transmitted electric field divided by the incident electric field. By dividing Eq. 28 and Eq. 29 with \( E_0 \), the reflection coefficient and transmission coefficient are obtained. Eq. 30 and Eq. 31 show the reflection and transmission coefficient in terms of matrix elements.

\[
r = \frac{\gamma_0 m_{11} + \gamma_0 \gamma_s m_{12} - m_{21} - \gamma_s m_{22}}{\gamma_0 m_{11} + \gamma_0 \gamma_s m_{12} + m_{21} + \gamma_s m_{22}} \tag{Eq. 30}
\]

\[
t = \frac{2\gamma_0}{\gamma_0 m_{11} + \gamma_0 \gamma_s m_{12} + m_{21} + \gamma_s m_{22}} \tag{Eq. 31}
\]

Eq. 30 and Eq. 31 allows one to analyze the propagation of an electromagnetic wave through a single thin film or a multilayer stack expressed by the transfer matrix. So, equations are derived that show what the reflection and transmission coefficients are for a certain stack of layers. When these equations are implemented in Matlab, a structure can be designed that will guide the light from the dipole in the upper space. Qualitatively, the idea of the calculation method is that the radiation of a point dipole is seen as the sum of contributions of different emission angles, as if the point source emits rays in all different
directions. For each angle, the transfer matrix method for multilayers is used to calculate the transmission and reflection of rays in to two half spaces around the multilayer stack in which the dipole is assumed to be placed. This provides an angular radiation diagram.

![Diagram of light beam incident on a thin film showing the magnitudes of the electric field and the magnetic field at the boundaries of half spaces](image)

*Figure 7: A light beam incident on a thin film. The magnitudes of the electric field and the magnetic field are shown at the boundaries of a and b.* [15]

### 2.3 Matlab calculation benchmarks

In the sections 2.1 and 2.2, the theoretical foundations for the Matlab code are explained. In this code, the radiation pattern of a dipole inside a multilayer can be calculated. In order to verify the code, radiation patterns from literature are used to make a comparison. The following parameters for the electric field can be adjusted in the code: the refractive indices for the half infinite space on the top, the layer with the dipole and the half infinite space on the bottom, the wavelength the dipole emits, the thickness of the layer with the dipole and the height of the dipole within this layer. Furthermore, a stack of two materials can be implemented, the refractive index of both materials can be chosen, the thickness of these layers and how many times the two materials are repeated. A schematic of the structure of what it looks like in Matlab is shown in Figure 8. The code uses these thicknesses and refractive indices to calculate the electric field at certain angles. This is done by calculating the transmission and reflection coefficients of the materials, which follow from a set of matrices.
The code has been made by a former master student. However, two things needed to be added to make the code work for the application of the project. In the original code, radiation patterns are calculated for a dipole with a fixed dipole orientation. Since in practice, the dipole orientations can’t be chosen, will the code have to sum the radiation pattern for a chosen number of random dipole orientations. A collection of dipole orientations will then represent the ensemble of random dipole orientations. This is done, by calculating the intensity for a number of dipole moments and average it over the dipole directions. This intensity is shown in Eq. 32.

$$I(\theta, \varphi \mid \theta_d, \varphi_d) \quad \text{Eq. 32}$$

where $I$ is the intensity per steradian in the far field directions of $\theta$ and $\varphi$, for a given dipole orientation aiming at $\theta_d$ and $\varphi_d$. Where $\varphi$ is the azimuthal angle in degrees, $\theta_d$ is the dipole orientation over the polar angle and $\varphi_d$ is the dipole orientation over the azimuthal angle. With this modification of the code, the code calculates the radiation pattern of random fluorophore ensembles i.e., the emitted flux as a function of viewing angle in the far field. Now, the other addition is that the code needs to calculate what fraction a microscope objective lens can collect for a given numerical aperture. To this end, the emission as a function of the angle for all angles $\theta$ and $\varphi$ (now referring to emission direction and not dipole orientation) that fall in the opening angle of a lens are summed. This means an integration over $\varphi$ from 0 to $2\pi$, and over $\theta$ from 0 to $\theta = \sin^{-1}(NA)$ where $NA$ is the numerical aperture of a microscope objective. To obtain a fraction, the result is divided by the total integrated emission. The code now calculates the fraction of the radiation pattern collected for a given numerical aperture for a dipole with a number of orientations in a multilayer structure.

With the code finished, a comparison can be made with the literature to check the code, where. First off, the radiation pattern is calculated for a dipole with a fixed dipole orientation. In Figure 9 and Figure 10, radiation patterns are depicted for a dipole on top of a waveguide in the polar coordinate system. The dipole has an orientation of 60° with respect to the vertical axis, it emits a wavelength of 488 nm, medium 1 and medium 2 both have a refractive index of 1, medium a and medium b both have a refractive index of $\sqrt{5}$ and an overall thickness of 80 nm, and medium 3 has a refractive index of 1.5. The height of the dipole above the interface is varied and is denoted in the top left of every plot. Figure 9 is taken from [17], whereas Figure 10 is retrieved by the code. Both figures show agreement with each other.
To be completely sure that the code works correct for this case, another comparison is made but with different parameters. Figure 11 A and Figure 11 B show the radiated power as function of the angle. The dipole again has a 60° dipole orientation with respect to the vertical axis, it emits a wavelength of 633 nm, the height of the dipole is 20 nm, medium 1 and medium 2 both have a refractive index of 1 and medium 3 has a refractive index of 1,5. Figure 11 A is taken from [17], whereas Figure 11 B is retrieved by the code. Again, both figure show superb agreement with each other.

After this, the calculated radiation patterns for random dipole orientations will be verified with literature. Figure 12 A and Figure 12 B show the angular distribution of power radiated for a random dipole orientation. Medium 1 and medium 2 both have a refractive index of 1, medium 3 has a refractive index of \( \sqrt{2} \) and the height of the dipole is 0 nm. Figure 12 A is taken from [18] and shows the radiation pattern for s-polarized, p-polarized and the sum of s-polarized and p-polarized light. Figure 12 B is obtained from the code and shows the radiated power for the sum of s-polarized and p-polarized light. Again, both figures match with each other. However, the vertical axis shows some dissimilarity in the values due to the withhold of parameters in [18].

![Figure 9: Radiation patterns for a dipole with a dipole orientation of 60° with respect to the vertical axis on top of a waveguide, taken from [17]. It is presented in a polar coordinate system. On the top left of every image, the height of the dipole above the waveguide is denoted.](image)
Figure 10: Radiation patterns for a dipole with a dipole orientation of 60° with respect to the vertical axis on top of a waveguide, obtained by calculations. It is presented in a polar coordinate system. On the top left of every image, the height of the dipole above the waveguide is denoted.

Figure 11: Angular distribution of the radiated power for a dipole with a dipole orientation of 60° with respect to the vertical axis. Figure A is taken from [17] and figure B is obtained by calculations. Note the shift in definition of angle, shifting the curves by 90 degrees.
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Figure 12: Angular distribution of the radiated power for a number of random dipole orientations. Figure A is taken from [18] and figure B is obtained by calculations.

With the code now being verified by literature, an optimal design can be realized. As mentioned before in 1. Introduction, recent studies [3] [4] [5] [6] have shown that planar antennas can be used to modify the radiation patterns of emitters in such a way that the photons are emitted in a narrow cone. This enhances the collection efficiency of the photons emitted by the dipole. The planar antenna structure in [5] demonstrates the beaming effect with a highly reflecting mirror, a dielectric spacer layer embedded with fluorophores and a thin metal layer. Since this structure only consists of three layers, it is relatively easy to make. Also, the materials used in [5], are present in the thin-film deposition lab. The following materials will be used: Gold for the highly reflecting mirror and the thin metal layer on top and silicon dioxide will be used as the dielectric spacer layer.

2.4 Fourier optics

In this report a so-called Fourier microscope has been used to measure radiation patterns as a function of angle. This method makes use of a normal microscope, an optical system designed for imaging. The object is placed a focal distance away from the objective lens. Fourier microscopy uses the fact that the back-focal plane of the microscope objective contains angular information. How exactly this angle is mapped, is dictated by a convention in microscopy called the ‘Abbe sin condition.’ If an objective is used with a high numerical aperture it must fulfill the Abbe sine condition. It states that the sine of the input angle should be equivalent to the sine of the output angle, represented in Eq. 33.

\[
\frac{\sin(\alpha)}{\sin(\alpha')} = \frac{\sin(\beta)}{\sin(\beta')} \quad \text{Eq. 33}
\]

where \(\alpha\) en \(\beta\) are the angles of two beams leaving the sample and \(\alpha'\) and \(\beta'\) the angles of how they reach the image plane. Now, for a system that fulfills the Abbe sine condition, the light beams coming from or going to the focal point of the objective will intersect the focal sphere, see Figure 13. [19] The focal sphere is a sphere with a radius that corresponds with the focal length of the lens, \(f\) in meters. Now, the light will end up at a certain height above the optical axis, given by Eq. 34.

\[
y = f \sin(\theta) \quad \text{Eq. 34}
\]
where $y$ is the vertical distance from the optical axis to the intersection of the focal sphere and $\theta$ is the angle a light beam makes that goes to or comes from the focal point with the respect to the optical axis.

![Figure 13: Light beams intersecting the focal sphere. [16] ](image)

The distance from the center of the sphere to the edge can also be expressed in terms of the angular wavenumber $k$. Figure 13 depicts the direction of the x-, y- and z-axis, with this information Eq. 35 - Eq. 37 can be retrieved.

\[
k_x = \frac{2\pi}{\lambda} \sin(\theta) \cos(\phi) \quad \text{Eq. 35}
\]
\[
k_y = \frac{2\pi}{\lambda} \sin(\theta) \sin(\phi) \quad \text{Eq. 36}
\]
\[
k_z = \frac{2\pi}{\lambda} \cos(\theta) \quad \text{Eq. 37}
\]

where $k_x$ is the angular wavenumber in the x-direction, $k_y$ is the angular wavenumber in the y-direction, $k_z$ is the angular wavenumber in the z-direction and $\phi$ is the azimuthal angle in degrees. The angular wavenumbers for the x- and y-direction depend of the sine of the angle, this means that $k_x$ and $k_y$ obey the Abbe sine condition.

As an example, consider that a Fourier image of grating is being created by an optical system that fulfils the Abbe sine condition. It is known, that the grating formula for normal incident light is given by Eq. 38.

\[
m\lambda = d \sin(\theta) \quad \text{Eq. 38}
\]
Where \( m \) is an integer representing the mode and \( d \) is the periodicity of the grating in meters. Eq. 38 can be rewritten in a different form by using Eq. 35 for the \( x \)-direction and Eq. 36 for the \( y \)-direction, resulting in Eq. 39 and Eq. 40.

\[
\begin{align*}
  k_x &= \frac{2\pi}{d_x} m \\
  k_y &= \frac{2\pi}{d_y} m
\end{align*}
\]  

Eq. 39  Eq. 40

Where \( d_x \) is the periodicity in the \( x \)-direction and \( d_y \) is the periodicity in the \( y \)-direction. These two equations show that when an image is made in Fourier space of a grating, then the distinct grating orders appear as isolated dots on the back focal plane. These grating orders appear as a regular array at fixed spacing equal to \( 2\pi/d \), due to the Abbe sine condition. In addition, Eq. 39 and Eq. 40 can also be rewritten by substituting them in Eq. 35 and Eq. 36, giving Eq. 41 and Eq. 42.

\[
\begin{align*}
  \frac{2\pi}{\lambda} \sin(\theta) &= \frac{2\pi}{d_x} m \\
  \frac{2\pi}{\lambda} \sin(\theta) &= \frac{2\pi}{d_y} m
\end{align*}
\]  

Eq. 41  Eq. 42

This property of gratings is used to calibrate the Fourier microscope. A grating of known periodicity \( d \) provides grating orders that in the back focal plane are spaced by \( 2\pi/d \). In an actual set-up, the Fourier image will be a disk of size \( 2 \cdot NA \cdot f_{obj} \) where \( f_{obj} \) the objective focal length in meter. Calibration of this effective magnification of k-space by a grating allows to calibrate the angle. So, if the distance between two consecutive modes and the length of \( k \) are known, the number of modes can be calculated, shown in Eq. 43.

\[
m = \frac{2\pi}{\frac{2\pi}{d_x}}
\]  

Eq. 43

From Eq. 43 follows that, when a Fourier image is obtained where the length of \( k \) is given in pixels, the distance between the modes in pixels is given by Eq. 44.

\[
d_p = \frac{k_p}{m}
\]  

Eq. 44

where \( d_p \) is the distance between the modes in pixels and \( k_p \) the length of \( k \) in pixels. With reference to Eq. 44, the distance measured between the modes can be checked by some calculations, resulting in trustworthy measurements.

3. Fabrication process

In this chapter both the equipment used and the fabrication process of the samples will be explained. The goal is to fabricate a multilayer structure, consisting of different materials and layer thicknesses, on top of a glass substrate. It is important to manufacture these layers in a controlled manner. In addition, layers with increasing thicknesses need to be manufactured, thus making a layer in the form of a wedge. On this wedge, fluorophores are spincoated, so that the emitters are embedded at different heights within the
sample. In that case, different radiation patterns can be obtained with only using one sample, rather than making samples were the height of the fluorophores are fixed, which is time-consuming. Also, comparing radiation patterns from different samples is hard, since the layer thicknesses of the different sample will never match with each other but slightly differ. Therefore, wedges will be used to embed the fluorophores at different heights in the sample.

3.1. Equipment

3.1.1. E-beam evaporator

The E-beam evaporator (Polyteknik flextura M508 E) is the first equipment used in the fabrication process. E-beam evaporator stands for electron-beam evaporator and it is used for thin-film deposition. [20] The electron beam is generated by sending a current through a specific material under high vacuum, base pressure is around $10^{-8}$ mbar. The electrons are accelerated by an electric field and directed at a socket containing the evaporation material by a magnetic field. The electrons will be bombarded onto the material, converting their kinetic energy into thermal energy. The evaporation material heats up and starts to melt, leading to vaporization. The material in gas phase will travel upwards in the vacuum chamber, towards the sample, leaving a thin layer of material where the atoms hit the surface. In this way, thin layers can be made in a controlled manner. The thickness of the layer is measured by a quartz crystal microbalance, better known as QCM. Quartz is a material that exhibits piezoelectricity. Therefore, quartz can be made to oscillate at a defined frequency by applying an alternating current. Obviously, when the layer thickens on the quartz crystal, the overall mass will increase. This causes the frequency to shift so that the layer thickness can be calculated for which evaporation material is used. Another feature on the E-beam is the slow shutter. The slow shutter is a plate that can move in the x-direction and in this way block a part of the material in the gaseous phase. This allows to create a wedge of the material that needs to be evaporated.

3.1.2. Spin coater

The spin coater will be used next in the fabrication process. The spin coater is a machine that makes uniform thin films across flat substrates. [21] This is done by rotating the substrate while drop casting the solution on top. The centrifugal force will spread the solution, resulting in an uniform thin film. If the angular velocity increases, the film will get thinner. Spin coaters usually apply photoresist films to the substrate, but it can also be used to apply quantum dots on the substrate. There are two spin coaters available, one is in the clean room (Suss Microtec Delta 80) and the other one is in the chemistry lab of the Resonant Nanophotonics group (Laurell Model WS-650MZ-23NPPB).

3.1.3. Profilometer

The profilometer (KLA-Tencor alpha-step 500) is the last equipment used. It is an instrument used for measuring the profile of a surface but can also be used to measure step heights. [22] It is a mechanical,
A stylus-based step profiler that measures step heights, independent of the material properties. A camera shows where the stylus is located on the sample. Figure 14 indicates what the camera of the profilometer shows. The stylus is lowered until the tip hits the surface of the sample. After this, the stylus will move in one direction measuring the profile while maintaining contact with the sample, this is done by applying a small force onto the stylus.

![Image: The tip of the stylus from the profilometer. The blue arrow indicates the scan length.](image)

**Figure 14:** The tip of the stylus from the profilometer. The blue arrow indicates the scan length.

### 3.2. Sample fabrication

All cover slides are treated with a base Piranha solution, in order to remove contaminations. The cover slides are made from borosilicate glass with a thickness of 0.17 mm and a size of 24 x 24 mm. The standard process starts with the sonication of the cover slides in H₂O to remove any large contaminations. After this, a beaker is filled with 5 parts H₂O and 1 part NH₄OH with a concentration of 30% in H₂O and heated to 75 °C. One part of H₂O₂ with a concentration of 30% in H₂O is added and the beaker is reheated to 75 °C. Then the cover slides are added and left there for 10 to 15 minutes. Afterwards the cover slides are removed from the beaker, rinsed in distilled H₂O and 2-propanol and finally blown dry with N₂.

After this treatment, thin films are thermally deposited with the E-beam. Firstly, a chrome layer with a thickness of 3 nm is deposited. This layer will serve as an adhesion layer for the gold, which is deposited next. The gold layer has a thickness of 100 nm, this will result in an optical thick gold layer. After this, a wedge of silicon dioxide is deposited using the slow shutter. On one side, it reaches a thickness of 160 nm. On the side where the slow shutter covered the sample, no silicon dioxide will precipitate.

When the deposition of these two layers is finished, the thicknesses are measured with the profilometer. On one side, where the slow shutter covered the sample, only a layer of gold is present. On the other side, where the slow shutter did not cover the sample, there should be two layers present, one layer with a thickness of 100 nm, the gold layer, and one layer with a thickness of 160 nm, the silicon dioxide wedge.
The wedge has a length of 10 mm. However, if the evaporation rate is not constant during the process, the length of the wedge will change. These thicknesses are measured for every sample made.

When these thicknesses are measured, the emitters need to be spincoated on top of the silicon dioxide wedge. The emitters used are Qdot 655 and Qdot 800 ITK organic quantum dots from ThermoFisher dissolved in decane, giving solutions with a concentration of 1 µM. The solutions with Qdots are diluted a thousand and a million times, in toluene. Resulting in two solution for each Qdot, one with a concentration of 1 nM and one with a concentration of 1 pM. The samples are rotated with an angular velocity of 3000 rpm for 30 seconds, while drop casting 50 ml of one of the solutions at the start. This is done with the spin coater in the chemistry lab.

After spincoating the quantum dots, another wedge of silicon dioxide is deposited, again with a thickness of 160 nm, but rotated horizontally with respect to the first wedge. This results in a rectangle, allowing the Qdots to be stationed at different heights within the silicon dioxide. Since the Qdots are at different heights within one sample, different radiation patterns can be measured. Then the final layer is deposited, a layer of gold with a thickness of 20 nm.

The final step is to measure the thicknesses of the last two layers deposited, again with the profilometer. The silicon dioxide layer should now have a thickness of 160 nm on both sides and on top a 20 nm thick layer. Note that this fabricated structure is optimal for Qdot655, as will be later shown in 5.1 Design of the planar antenna structure. The layer thickness for an optimal structure of Qdot800, are slightly different. But all steps in the fabrication process are the same. After this, the fabrication process is completed.

Later, it was discovered that the Qdots will not survive the evaporation process, shown in chapter 5 Results. This was fixed by adding an additional layer of PMMA. It is spincoated on top of the quantum dots, by using the spin coater stationed in the cleanroom. The PMMA is dissolved in a 1:2 solution of unisole. The samples are rotated with an angular velocity of 4000 rpm for 45 seconds, while drop casting 50 ml of the PMMA solution at the start. After spinning, the sample is heated to 180 °C for 2 minutes, this will harden the layer of PMMA. This should result in a thickness of 50 nm of PMMA. Again, the thickness is checked by the profilometer.

4. Methods

This chapter will describe the method of measuring the radiation pattern of single photon sources in a planer antenna structure. First, the set-up used will be described. Fourier microscopy is used to measure the angular response rather than to obtain a spatial image of the sample. Second, the calibration method of the setup will be explained. Finally, measurements on the single photon sources can be conducted.

4.1. Set-up

4.1.1. Fourier microscopy

All measurements are carried out in the optical set-up, shown in Figure 16, which has the capabilities of real space imaging and Fourier imaging. For this reason, it is called a Fourier microscope. A ray diagram is depicted in Figure 15, this shows what the effect is when measuring with the Fourier lens. The blue lines are rays originating from a single point on the source. An objective collimates these rays and the tube lens
focusses it on the camera. But, this does not give any information under which angle the light is emitted from the sample. This information can be retrieved by adding an extra lens in the set-up, the Fourier lens. The red lines are collimated light rays which enter the microscope objective with different angles of incidence. Rays with the same angles of incidences, will be focused at a certain height on the back focal plane of the objective, since the system obeys the Abbe-sin condition. The Fourier lens is placed in such a way, that these rays will also be focused on the same spot on the CMOS. This happens for all light rays with the same angle of incidence. Therefore, an image is obtained that comprehends the radiated power as function of the incidence angle. The objective is in this case the collection device of the system. Therefore, the maximum collection angle is limited by the numerical aperture of the objective. The numerical aperture is described by the maximum angle with respect to the optical axis under which light can be collected multiplied by the refractive index of the medium between the emitter and the objective.

![Ray diagram](image)

*Figure 15: Ray diagram for image formation with and without the Fourier lens. The blue lines represent the projection of a real space image on the CMOS, obviously this is without the Fourier lens. The red lines represent the projection of a Fourier image on the CMOS when the Fourier lens is present. [16]*

4.1.2. Experimental set-up

In this section, the experimental set-up will be described. The schematics of the experiment is depicted in Figure 16. Light from a supercontinuum laser (NKT, SuperK EXTREME EXR-15, 490 nm ~ 2100 nm) is used to illuminate the sample. A supercontinuum laser uses nonlinear mechanisms to induce spectral broadening of the short laser pulses. [23] Therefore, a broad white laser output is provided. This output can be filtered by using an Acousto-Optic Tunable Filter (Crystal Technology, PCAOM VIS, 450 nm ~ 750 nm, spectral bandwidth 1 nm), which transmits one wavelength with a spectral bandwidth of 1 nm. Three AOTFs can operate at same time and thus increase the output power. The light is collected by an optical fiber (1) and guided into the set-up (2). Firstly, the light beam passes through a bandpass filter (3) to clean it from other frequencies transmitted by the AOTFs. An achromatic doublet (Thorlabs, AC254-200-B, focal length = 200 mm) (4) focuses the light on the back focal plane of a microscope objective (Olympus UPlanSApo, 100x magnification, NA=1.40; infinity corrected or Nikon, 100x magnification, NA=0.85; infinity corrected) (5) by making use of a pellicle beamsplitter (Thorlabs, BP145B2, coated for Reflection =
45% Transmission = 55% for 700 nm ~ 900 nm. If the microscope objective with a numerical aperture of 1.40 is used, immersion oil (n=1.52; Sigma Aldrich, fluka) will be applied between the objective and the sample. Because the light is focused on the back focal plane of the microscope objective, the sample will be illuminated by a collimated beam. The fluorescence will be collected by the same microscope objective and a part of it will travel through the pellicle beamsplitter. It passes through a 1x magnification telescope, both achromatic doublets (Thorlabs, AC254-50-B, focal length = 50 mm) (8) (9), which allows to spatially select an area on the sample using a pinhole in the middle of the telescope. An achromatic doublet (Thorlabs, AC254-200-B, focal length = 200 mm) will be used as tube lens (11), that projects the image on a camera (PCO, panda 4.2 sCMOS, 2048 x 2048 pixels) (12). Under these circumstances, the projected image will be in real space. Now, it is possible to flip in an extra achromatic doublet (Thorlabs, AC254-200-B, focal length = 200 mm) (10), the so-called Fourier lens. This lens projects the back focal plane of the microscope objective on the camera. A key thing in the set-up is, that it is possible to have both the pinhole and the Fourier lens ‘active’. As a result, back focal plane images of a single emitter can be made. When measuring fluorescence of molecules, a longpass filter (7) can be placed in front of the camera, to filter out the excitation light.

![Figure 16: Schematic of the Fourier microscope. The numbered items are explained in the text above.](image)

4.2. Characterizations

The set-up is characterized for real space measurements and for Fourier space measurements. This is done to ensure reliable imaging of the samples. For the characterizations, the microscope objective with a NA of 1.40 is used. In both cases, 1 mW output power is used.

4.2.1. Real space

To characterize the real space, a sample with a diffraction grating is used. The diffraction grating has a periodicity which is known. The grating consists of a square grid, composed of horizontal and vertical lines with a periodicity of 7.7 micron. The characterization is performed by measuring the grating with the Fourier microscope and a fluorescence imaging microscope as reference. First, the characterization process of the Fourier microscope is explained. The diffraction grating will be illuminated by focusing a collimated laser beam, with a wavelength of 520 nm, on the back focal plane of the microscope objective. This will result in a collimating beam at the output of the objective. The reflected light is collected by the CMOS camera. After this, the diffraction grating will be measured with the fluorescence imaging microscope as reference. A white light source will be used for illuminating the sample. Again, the reflected light will be collected by the camera. Since two different microscopes are used, two independent measurements for the periodicity are obtained, resulting in a reliable characterization. However, a periodicity of 7.7 micron is expected from both measurements.
4.2.2. Fourier space

To characterize the Fourier space i.e. the angular response, the same diffraction grating is used and also a fluorescing compound, in this case Rhodamine 6G. The Rhodamine 6G will act as a dipole, emitting light under certain angles. Both measurements for the characterization of the angular response are carried out at the Fourier microscope. Both samples are again illuminated by focusing a collimated laser beam, with a wavelength of 520 nm, on the back focal plane of the microscope objective. First, a focused image is obtained in real space by adjusting the distance between the sample and the objective. After this, the Fourier lens is flipped in. Now, a focused image needs to be obtained in Fourier space. This is done by moving the Fourier lens. When both samples are measured, theory explained in 2.4 Fourier optics will be used to show the consistency of both samples in Fourier space. For example, if the y-direction is used for calibration, Eq. 44 will show that the theory and the measured value agree with each other. First, the amount of modes are calculated by Eq. 42:

\[ m = \frac{2\pi}{520 \cdot 10^{-9}} \cdot \frac{7,7 \cdot 10^{-6}}{2\pi} \]
\[ m = 14,81 \text{ modes.} \]

The distance in pixels between the modes is thus given by:

\[ d_p = \frac{k_p}{14,81} \]

Since the length of \( k \) in pixels is measured due to the Fourier image of the Rhodamine 6G, the measured distance between the modes can be compared to the calculated value \( d_p \). It is expected that the calculated value and the measured value agree with each other.

4.3. Measuring quantum dots in a multilayer structure

After the characterization of the Fourier microscope, radiation patterns of the fabricated samples are measured and compared with the radiation patterns following from the MATLAB code. For all measurements, the microscope objective with a NA of 0.85 is used. For Qdots655, the wavelength was set at 520 nm by the AOTF, resulting in an output power of 2 mW. For Qdots800, the wavelength was set at 20 nm by the AOTF. Both laser beams were cleaned from other frequencies by the bandpass filter. In both cases will the sample be illuminated by a collimated laser beam. The reflected laser light coming from the sample was filtered out by longpass filters, meaning that only the fluorescent light of the Qdots is transmitted and reaches the camera. First, a focused image is obtained in real space by adjusting the distance between the sample and the objective. After this, the Fourier lens is flipped in. Now, a focused image needs to be obtained in Fourier space. This is done by moving the Fourier lens.

There are several samples made with Qdots. One sample is a glass substrate with Qdots spincoated on top, depicted in Figure 17. Another sample is a glass substrate with a layer of 100 nm gold, a wedge of 160 nm silicon dioxide and Qdots spincoated on top, depicted in Figure 18. The last sample is the sample shown in Figure 22. It is expected that the structure of Figure 22 radiates most of the light into the microscope objective. The glass substrate with Qdots is expected to give the lowest signal and the sample with the layer of gold and one wedge is expected to be somewhere in between. Obviously, it is expected that the simulations agree with the obtained results from the measurements.
5. Results

In this chapter, the design of the planar antenna structure is presented. A design is shown for Qdot655 and Qdot800. Also, a layer thickness measurement is shown for one of the sample. Afterwards, results for the characterization of real space and Fourier space, of the Fourier microscopy are presented. Afterwards measurements on simplified structures are shown. First off, the influence of the excitation wavelength is shown. Second, the results for the protection layer of the Qdots are depicted. Finally, radiation patterns are shown for planar antenna structures.

5.1 Design of the planar antenna structure

So, as mentioned before in 2.3 Matlab calculation benchmarks, the structure consists of a gold layer for the highly reflecting mirror and the thin metal layer on top and silicon dioxide will be used as the dielectric spacer layer. The refractive indices of these materials are implemented in the code. The layer thicknesses used in [5] are also implemented. Since a different emitter wavelength will be used, these thicknesses need to be modified for an optimal collection efficiency. Two designs will be realized, one for an emitter with an emission wavelength of 655 nm and one for an emitter with an emission wavelength of 800 nm. The design for the emitter with an emission wavelength of 655 nm is shown in Figure 19. This design will give an optimal collection efficiency of 0.9376 for a numerical aperture of 0.85 and a height of 70 nm for the emitters in the silicon dioxide is. This collection efficiency is obtained by tweaking the thickness of the
dielectric spacer layer and the height of the emitters. However, a wedge is made to have one sample with several emitters at different heights. This grants a sample where the emitted radiation pattern has a height dependency, as mentioned earlier. The refractive indices for gold and silicon dioxide are obtained from [24]. In Figure 20, the angular distribution of the radiated power for an emitter with an emission wavelength peak at 655 nm is shown. This plot indicates the beaming effect, since most of the power is emitted under small angles. In addition, the angular distribution of the radiated power for an emitter with the same emission wavelength peak but a different height is shown. The height is chosen at 0 nm and is depicted in . If these two images are compared, it gets clear that the angular distribution of the power for a dipole at a height of 70 nm is more efficient than for a dipole at a height of 0 nm.

Figure 19: The structure design for an emitter with the emission peak at 655 nm. The optimal collection efficiency is obtained when the emitters are embedded at a height of 70 nm.
Figure 20: The angular distribution of the radiated power emitted by a dipole at a height of 70 nm with an emission wavelength peak at 655 nm. This plot shows that most power is emitted at small angles which indicates the beaming effect.

Figure 21: The angular distribution of the radiated power emitted by a dipole at a height of 0 nm with an emission wavelength peak at 655 nm. At this height, the dipole emits less power at small angles than the dipole at a height of 70 nm.

Besides the structure for an emitter with an emission wavelength peak at 655 nm, a structure for the emitters with an emission wavelength peak at 800 nm is also designed, depicted in Figure 22. Figure 22 will give an optimal collection efficiency of 0.9282 for a numerical aperture of 0.85 and the height of the emitter in the silicon dioxide is 70 nm. Equally, this collection efficiency is obtained by tweaking the thickness of the dielectric spacer layer and the height of the emitters. Again, a wedge is made to have one
sample with several emitters at different heights. In Figure 23, the angular distribution of the radiated power for an emitter with an emission wavelength peak at 800 nm at a height of 70 nm is shown. This plot similarly indicates the beaming effect, since most of the power is emitted under small angles. In addition, the angular distribution of the radiated power for an emitter with the same emission wavelength peak but a different height is shown. The height is chosen at 0 nm and is depicted in Figure 24. If these two images are compared, it gets clear that the angular distribution of the power for a dipole at a height of 70 nm is more efficient than for a dipole at a height of 0 nm.

![Diagram of structure design for an emitter with the emission peak at 800 nm.](image)

*Figure 22: The structure design for an emitter with the emission peak at 800 nm. The optimal collection efficiency is obtained when the emitters are embedded at a height of 70 nm.*
Figure 23: The angular distribution of the radiated power emitted by a dipole at a height of 70 nm with an emission wavelength peak at 800 nm. This plot shows that most power is emitted at small angles which indicates the beaming effect.

Figure 24: The angular distribution of the radiated power emitted by a dipole at a height of 0 nm with an emission wavelength peak at 800 nm. At this height, the dipole emits less power at small angles than the dipole at a height of 70 nm.
5.2 Characterization of the sample that include the optimal design.

As mentioned before, the thickness of the layers on the fabricated samples, are measured by using the profilometer. To give an indication where the stylus will be placed if a measurement is carried out, a schematic of a sample with a layer of gold is shown in Figure 25. The stylus will move in the y-direction at a certain speed, measuring the profile of the sample.

![Stylus Diagram](image)

*Figure 25: A sketch of a sample with a layer of gold on top of a glass substrate. The position of the stylus is also shown. When a measurement is started, it will start moving in the y-direction with a certain speed.*

The thickness results are shown for one sample as example. The results for the other samples are obtained in the same manner. The stylus is placed at the edge of the sample to measure a step height. In Figure 26, an example is shown when only a layer of gold is present on the sample, note that this also includes the chromium layer that functions as adhesion layer. The measured thickness of this layer is 114 ± 1 nm, so the gold has a thickness of 111 ± 1 nm.
Figure 26: A thickness measurement on a layer of gold, carried out with the profilometer. The measured thickness is $114 \pm 1$ nm, this includes the $3 \pm 1$ nm chromium layer.

Figure 27: A thickness measurement on a layer of gold and a layer of silicon dioxide, carried out with the profilometer. The measured thickness of the silicon dioxide is $143 \pm 1$ nm.

These measurements can be done along the whole side of the wedge, resulting in the profile. The x- and y-coordinate of the stylus can be set, so that the profile can be measured along the x-direction. As mentioned before, the wedge has a length of 10 mm along the x-direction. The beginning of the wedge is determined, so that the profile can be characterized. The stylus will move in steps of 1 mm in the x-direction, until the maximum thickness of the wedge is measured. In Figure 28, a measurement is shown when the stylus is set at a x-coordinate of 2 mm, the start of the wedge is considered at 0 mm. The height of the wedge at this position is $31 \pm 1$ nm. If more coordinates along the x-axis are measured, the slope of the wedge can be determined. Then, the height of the Qdots is known for every x-coordinate. The slope for this sample was determined at $15$ nm/mm, meaning that the wedge is longer than originally thought.
Figure 28: A thickness measurement on the wedge of silicon dioxide, carried out with the profilometer. The x-coordinate of the stylus is set at 2 mm, the start of the wedge is considered at 0 mm. The measured thickness of the silicon dioxide is 31 ± 1 nm.

5.2 Characterization results

The Fourier microscope is characterized for both real space and Fourier space.

5.2.1. Real space

Results for the characterization of real space are shown in Figure 29 and Figure 30. Figure 29 is an image obtained with the Fourier microscope and Figure 30 is an image obtained with the fluorescence lifetime microscope. Both images are measured by the Olympus oil objective. In both figures, the grating lines give either a maxima or a minima in counts. Therefore, by measuring the distance between the two peaks, the periodicity is obtained in pixels. The pixel values are converted to microns by multiplying it with the size of a pixel and then dividing it by the magnification of the set-up. From the specifications follows that the size of one pixel is 6.5 micron. The magnification of the set-up is given by the ratio of the focal length of the tube lens and the focal length of the microscope objective. The measured periodicity is determined on 136 pixels for Figure 29. The magnification of the set-up is 111 times for Figure 29. The objective has a magnification of 100 times for a tube lens with a focal distance of 18 cm. However, the focal length of the tube lens in the set-up is 20 cm. Meaning that the magnification of the set-up is not 100 times but given by $100 \cdot 20/18 = 111$. so that the pixel value can be converted to microns by doing the following:

$$d = \frac{136}{111} \cdot 6.5 = 7.9 \mu m$$

For Figure 30, the following periodicity is measured, 119 pixels. On both set-ups, the same camera is present, so that the pixel size again gives 6.5 micron. The magnification is 100 times for Figure 30. The same procedure is maintained and this results in the following periodicity in microns:

$$d = \frac{119}{100} \cdot 6.5 = 7.7 \mu m$$

The difference between the two values is 2.7%, this is acceptable. Therefore, measurements in real space are trustworthy.
Figure 29: A real space image of the diffraction grating with a periodicity of 7.7 micron. This image is obtained with the Fourier microscope.

Figure 30: A real space image of the diffraction grating with a periodicity of 7.7 micron. This image is obtained from the fluorescence lifetime microscope.
5.2.2. Fourier space

Results for the characterization of Fourier space are shown in Figure 31 and Figure 32. Both images are obtained with the Fourier microscope and again, the Olympus oil objective is used. Figure 31 is the Fourier space image of the diffraction grating and Figure 32 is the Fourier space image of the Rhodamine 6G. The real space image of the Rhodamine 6G is also shown, in Figure 33. In Figure 31, a very intense reflection covers the inner orders. Beside this spot, the diffraction orders are present and evenly spaced. A total of 17 orders are observed over a distance of 303 pixels. Thus, the orders have an even spacing of 18 pixels.

Furthermore, from Figure 32 the radius between the center of the ring and the black ring can be extrapolated. The onset at the black ring originates from the critical angle between glass and air which corresponds to a numerical aperture of 1. This follows from the figures originating from [18], also calculated with the Matlab code, see Figure 12. Therefore, the distance between the black and red ring is defined by the numerical aperture of the microscope objective. For instance, if a microscope objective with a numerical aperture of 1.4 is used, the radius corresponds to the NA of 1.4. By fitting this black ring, the center of the ring and the radius can be obtained. So, for Figure 32 the numerical aperture of 1.0 relates to 263 pixels.

From Eq. 44, the distance between the orders can be calculated because the radius from the black circle is known. Therefore, the theoretical value for the distance between the modes in pixels can be calculated, resulting in the following:

\[ d_p = \frac{263}{14.81} = 18 \text{ pixels} \]

Both values agree with each other. Therefore, measurements in Fourier space are trustworthy.

Figure 31: A Fourier space image of the diffraction grating with a periodicity of 7.7 micron. As expected, all orders have an even spacing.
Figure 32: A Fourier space image of Rhodamine 6G. The black line corresponds to a NA of 1 and the red line corresponds to a NA of 1.4.

Figure 33: A real space image of Rhodamine 6G. The image is obtained with the Fourier microscope.

5.3. Background fluorescence

The radiation patterns of the Qdots655 embedded in the planar antenna structure were measured with the Fourier microscope. Measurements are first done in real space obviously, to obtain a focused image of the Qdots. The real space image of the Qdots655 embedded in the planar antenna structure is shown.
in Figure 34. However, the image does not agree with the expected results. No Qdots appear in the image, only a bright spot appears. Of course, no Fourier image can be made if this bright spot appears.

Therefore, the sample was taken to the fluorescence imaging microscope, which has been used before for the characterizations. A spectrometer is present on this set-up and for this reason, the sample was taken there for further investigation. The same real space image was obtained from the microscope, indicating that the bright spot is not set-up dependent. From this real space image, a spectrum is taken, shown in Figure 35. The sharp peak at 530 nm originates from the laser beam. Also, a broad spectrum appears with the shape of a fluorescence emission spectrum, with a peak around 570 nm. This background fluorescence has an intensity higher than the intensity of the Qdots fluorescence. This result is attributed to the fact that the deposited silicon dioxide can fluoresce, especially if the deposited glass is not stochiometric. This fluorescence is hypothesized to be stronger the higher the pump photon energy i.e. bluer the laser light.

Figure 34: A real space image of the planar antenna structure, taken with the Fourier microscope. A bright spot appears and no quantum dots are visible.
Figure 35: A spectrum taken of the planar antenna structure with the fluorescence imaging microscope. The same bright spot appeared at this set-up and this is the retrieved spectrum. The sharp peak around 530 nm originates from the laser beam. The broad spectra with the peak around 570 nm has the shape of an fluorescence emission spectrum.

5.4 Measurements on quantum dots 800

Two adjustments have been made to the design, in order to effectively measure the Fourier image of the Qdots. The first adjustment was replacing quantum dots with an emission peak at 655 nm to quantum dots with an emission peak at 800 nm. Therefore, the fluorescence peak of the quantum dot shifts to the near infrared, avoiding the relative big bump of background fluorescence. This shift also allows to shift the pump laser wavelength to the red, further reducing background fluorescence. The results are shown in Figure 36. In this case, the fluorescence from the quantum dot is visible in the on state. However, when the quantum dot is in the off state, the signal coming from the background fluorescence dictates the spectrum. In order to get a better idea of the intensity coming from the Qdots compared to the background fluorescence, a normalized spectrum is also shown. From Figure 36 C, it is clear that another adjustment has to be made in order to effectively measure the signal emitted by the quantum dots.
Figure 36: Spectrum of the structures embedded with Qdots800. A) The spectrum when a quantum dot is in the on state, a bump appears. B) The spectrum when a quantum dot is in the off state, only background fluorescence visible. C) Normalized fluorescence spectrum for an on and off state of a quantum dot.

As mentioned before, the change in emission wavelength from the quantum dots, allows to shift the excitation wavelength of the pump laser. If the excitation wavelength is shifted to a higher wavelength, the background fluorescence could be eliminated. The spectrum from Figure 37 is obtained by using a laser with a wavelength of 640 nm. As can be seen, no background fluorescence is present, since the intensity of 495 counts is the offset of the spectrometer. Therefore, measurements on the Fourier microscope will be carried out with a wavelength of 620 nm. Note that the emission peak of the Qdot800 is not at the wavelength expected, the maxima is found at 725 nm. However, this is not an issue, since this measurement showed that the background fluorescence is no longer present. Later was found that the calibration of the spectrometer was slightly off.
Thus, a structure for Qdot800 described in 5.1 Design of the planar antenna structure is fabricated. Three AOTFs were set in usage at 620 nm, 622 nm and 625 nm, resulting in a output power of 6 mW. However, no fluorescence was measured from the Qdots800 with the Fourier microscope. Afterwards, the sample was taken to the lifetime fluorescence microscope to make sure that it was not due to the set-up. Again, no fluorescence was measured, indicating that the sample is not performing as expected.

5.5 PMMA measurement

So, either the Qdots800 died while fabricating the sample or the design does not provide the beaming effect. Since the beaming effect for these structures has been shown in recent studies, the assumption is made that fabrication process destroys the Qdots800. In particular, thermal evaporation of silicon dioxide on quantum dots is suspect, since the sample is effectively exposed to high temperature. Therefore, it is examined whether a thin layer could protect the quantum dots. To examine this, two glass substrates are spincoated with 1 nM of Qdots800. One of these samples, will be treated by spincoating a layer of PMMA on top, with a thickness of 50 nm. Thereafter, 160 nm of silicon dioxide is evaporated. Measurements are carried out with the Fourier microscope and the same parameters are used as described above, resulting in Figure 38. The figures indicated with 1 belong to the sample with no layer of PMMA and the figures indicated with 2 belong to the sample with a layer of PMMA. Furthermore, the A indicates that the image was taken before the evaporation of silicon dioxide and the B indicates that the image was taken after the
evaporation of silicon dioxide. Figure 38 implies that the layer of PMMA protects the Qdots800. Therefore, an extra step is added to the fabrication process.

Figure 38: Real space images of the glass substrates spincoated with Qdots800. The figures indicated with 1 belong to the sample with no PMMA and the figures indicated with 2 belong to the sample with PMMA. The indication A means that the image was taken before the evaporation of silicon dioxide and indication B means that the image was taken after the evaporation of silicon dioxide.

5.6 Radiation patterns from the structures

Now, with the set-up characterized and the right recipe for the structures, radiation patterns can be measured and compared with literature. First off, the radiation pattern for Qdots800 on a substrate of
glass are measured, shown in Figure 17. The results are depicted in Figure 39. Figure 39 A is the plot profile of Figure 39 B, which is the Fourier image taken.

![Figure 39: A radiation pattern for Qdots800 on a substrate of glass. A) is the plot profile of B) the Fourier image.](image1)

Second, the radiation pattern for Qdots800 on a structure depicted in Figure 18 are measured. The Qdot is at a height of 100 nm. The results are depicted in Figure 40. Figure 40 A is the plot profile of Figure 40 B, which is the Fourier image taken. A central peak is observed. This is a part of the pump laser light, which is transmitted by the longpass filters. In , a radiation pattern is calculated with the Matlab code. As can be seen, the calculated form agrees with the measured form shown in Figure 40 A.

![Figure 40: A radiation pattern for Qdots800 on a structure depicted in Figure 18. A) is the plot profile and B) the Fourier image. In both plots, a peak can be seen. This is a part of the original laser light, which is transmitted by the longpass filters.](image2)
Figure 41: The calculated plot profile of a quantum dot at a height of 100 nm, obtained with the Matlab code.

Finally, the radiation pattern for Qdots800 embedded in a structure displayed in Figure 22 are measured. The Qdot is embedded at a height of 70 nm. The results are depicted in Figure 42. Figure 42 A is the plot profile of Figure 42 B, which is the measured Fourier image. Besides this measurement, the radiation pattern for a Qdot embedded at a height of 0 nm is displayed. Figure 43 A is the plot profile of Figure 43 B, which is the measured Fourier image. Earlier, the angular distributions of the radiated power for both heights has been calculated and depicted in Figure 23 and Figure 24.

Figure 42: A radiation pattern for Qdots800 embedded at a height of 70 nm in a structure displayed in Figure 22. A) is the plot profile and B) the Fourier image.
It gets clear from Figure 39, Figure 40 and Figure 42 that the structure design from Figure 22 radiates the most power into the microscope objective. The structure with only one wedge has less power then the structure from Figure 22 and the glass substrate with Qdots800 radiated the least power into the microscope objective, this is expected.

However, the expected beaming effect is not measured. If the calculated radiation patterns are compared with the measured radiation patterns, it is clear that the beaming effect is not obtained. Nevertheless, the calculated radiation pattern show that if the height of the quantum dot increases in the silicon dioxide, a higher intensity is obtained at lower angles. In Figure 42 and Figure 43, the same is observed. For the Qdots at an optimal height of 70 nm, a lot more counts are measured then for the Qdots at a height 0 nm.

6. Conclusion

The goal of this work was to optimize the collection efficiency of a fluorophore in a planar antenna structure. A Matlab code was used to calculate the radiation pattern and the amount of light collected by the microscope objective for an emitted with a random dipole orientation embedded in a multilayer stack. The refractive indices and the thicknesses of the layers could be varied, as well as the height of the emitter in the dielectric spacer layer. A structure was designed, consisting of a highly reflecting mirror, in this case gold, a dielectric spacer layer, in this case silicon dioxide, embedded with quantum dots and a thin gold layer on top. A structure of this kind, resulted in the highest collection efficiency. A detailed fabrication recipe for the manufacturing of these structure has been reported. The layer thicknesses could be made with high accuracy using the e-beam evaporator.

The structures were measured with a optical set-up, that is capable of mapping the complete angular response of a sample. This is a Fourier microscope where the back focal plane of a microscope objective is measured for fluorescent emitters, which can be translated to the radiation pattern. The first experiment was characterizing the Fourier microscope for real space and Fourier space by using a grating and a sample with Rhodamine 6G. These results showed that further experiments on the Fourier microscope could be trusted for both real space and Fourier space.
When trying to measure the radiation pattern for the designed structure, it was observed that the silicon dioxide layer would heavily fluoresce in the region of the fluorescence spectra of the Qdot655. Measurements showed that the background fluorescence could only be avoided, if the pump laser wavelength was shifted towards the red, forcing the usage of Qdots with a longer emission wavelength peak, the Qdots800. Another adjustment that had to be made, was the extra fabrication step of spincoating a layer of PMMA. The Qdots did not survive the thermal evaporation of silicon dioxide, since the sample is effectively exposed to high temperature. The added layer of PMMA fixed this problem.

Subsequently, the Fourier microscope could be used to investigate the radiation patterns of emitters embedded in the planar antenna structures. Besides planar antenna structures, radiation patterns of emitters in simpler configurations are also measured. The experimentally obtained radiation patterns are compared with the calculated radiation patterns. It is shown that the calculated angular distribution of the radiated power has the same shape as the experimental result, for a structure with a silicon dioxide wedge on top of a gold layer. However, the planar antenna structure does not show an agreement between the experimental and the calculated radiation patterns. The expected beaming effect is not observed. Nevertheless, as the height of the Qdot increases in the planar antenna structure, it is expected to emit more light under a small angle, following from calculations. This is something what is observed in the experimental measurements.

To conclude, it has been shown that it is possible to fabricate a planar antenna structure embedded with quantum dots. This can play a role in other applications where the working distances are relatively long. A first attempt has been made to measure the radiation pattern of such a planar antenna structure. However, more research is necessary in order to make quantitative statements about the so called beaming effect of the quantum dots which are embedded in a planar antenna structure.
References


