Investigation of the optical properties of nanostructured interfaces

PhD thesis

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Chapter 1

Introduction

In the past two decades optoelectronic devices became a "natural" part of our everyday life. We use colorful and smart devices without noticing the countless technological solutions involved. Energy saving and environment protection lead us towards sophisticated new technologies, with solutions pointing towards structures of characteristic length scale of nanometers, i.e. below the wavelength of the visible light. Among others such structures are used in light emitting and light harvesting applications, in plasmonic materials, photonic crystals and metamaterials.

The common property of these devices is that light must pass one or more interfaces. During the propagation the incoming electromagnetic radiation is transmitted, reflected, absorbed or scattered. In a major part of these devices waveguide and propagating plasmonic modes can be excited, too. There is a huge potential in optimizing the interfaces in order to achieve better performance depending on the type of the application. One can achieve improved energy efficiency of solar cells, higher field enhancement for plasmonic devices, tailored permittivity for metamaterials, controlled luminescence of nanodots etc. These interfaces can be used to add entirely new functionality to the device, for example enhancing polarized light emission in LED sources.

The aim of my thesis is to understand the most important factors affecting the propagation of electromagnetic waves in multilayer systems and to use this knowledge to develop energy efficient and functional devices. The work focuses on the electromagnetic modeling of such systems, including nanoparticles on the surfaces and surface corrugations. The results of this work can be used to help the experimental realization and fine tuning of such devices with prior knowledge about the parameters affecting the behavior of the device.

The first half of this thesis deals with polarized light extraction from light emitting diodes (LEDs). Two possible solutions will be shown for
the realization of polarized light output from light emitting diodes. First, array of metallic nanoparticles of ellipsoidal shape are investigated on a semiconductor-packaging interface. Second, the polarizing property of metallic nanoslit array is exploited at different interfaces. Finally the nanoslit array is combined with a complete LED model in order to estimate the performance of the complete setup.

In the second part of this thesis I will investigate the angle and wavelength dependent mode structure of metallic nanovoid and metallic film on nanoparticle arrays. These structures support both propagating as well as localized plasmon modes. The strong electromagnetic fields of the plasmon modes can be utilized for solar cells, surface enhanced Raman scattering or for sensing applications.

The electromagnetic models used in this thesis are rather diverse. Although all models are based on Maxwell's equations, the theoretical and numerical implementation varies with the actual problem in consideration. Throughout the thesis well established methods like finite difference time domain (FDTD) method, rigorous coupled wave analysis (RCWA) and geometrical optical ray tracing are used in some cases. However these methods are sometimes inappropriate to use: they might provide a rigorous solution of Maxwell's equations, but practically we often do not have enough computation power and time to calculate the corresponding problem correctly due to large computational size. Moreover optimization or investigation of the effect of different parameters is also limited due to performance limitations.

For example in the case when nanoparticles in the size range of 10 nm-s are considered it is more straightforward to implement a method adapted to this actual problem. Here I use the periodic layered Green's tensor method. This saves running time and computational power, while increases the precision of the calculation in the given range of applicability. The flexibility in choosing the appropriate model for a given problem is a great advantage if one wants to solve the problem efficiently and precisely.

The layout of the thesis is as follows. In Chapter 2 I review the most important literature connected to the subjects of this thesis and lay emphasis on the various modeling methods used throughout this thesis. In Chapter 3 the polarizing properties of silver nanoellipsoid arrays on a single interface are investigated. Chapter 4 contains the results obtained in the field of polarizing properties of wire grid polarizers. In Chapter 5 a complete polarized LED model is investigated theoretically with the help of the combination of ray tracing and rigorous coupled wave analysis. The plasmonic modes of nanovoid and particle on film arrays are discussed in Chapter 6. The dissertation is finished with the Summary in Chapter 7, my publications are listed in Chapter 8 and finally the Bibliography closes my thesis.
Chapter 2

Summary of literature

2.1 Introduction

This chapter serves two aims. First, it provides an introduction to the physics of the investigated systems in this thesis: light emitting diodes, polarizers and plasmons. These topics are discussed in detail in Section 2.2 and 2.3.

Second, it gives an insight into the applied electromagnetic models used in this thesis. In this work I use different methods in order to calculate electromagnetic scattering on surface structures. Different types of physical problems have specific properties which can be exploited in order to construct an efficient and flexible computational method. This means in some cases that we can choose a rigorous method which is well adapted to the geometry (e.g. using periodic boundary conditions for periodic structures), whereas in other cases suitable approximations of the exact solution can be used (e.g. using quasistatic calculations in small structures compared to the wavelength). It is very important to find the most efficient and most appropriate method, because this enlarges our possibilities and allows us to optimize the system, or sometimes it is the only chance to model the physical problem with the available computational power.

First the periodic layered Green’s tensor method with quasistatic approximation is reviewed in more details in Section 2.5. In this method the Maxwell’s equations are solved with the Green’s tensor method in a multilayer system. The nanoparticles are approximated with quasistatic approximation which can be used for small particle sizes compared to the wavelength. This method is used in Chapter 3.

Second the rigorous coupled wave analysis can be used to calculate the diffraction on two or three dimensional periodic structures. In Section 2.6 I only review the two-dimensional case with binary refractive index modula-
Finally in Section 2.7 I review shortly three additional methods: finite difference time domain method (FDTD, used in Chapter 6), finite element method (FEM, used in Chapter 3) and nonsequential raytracing method (used in Chapter 5).

2.2 Introduction to light emitting diodes

In general a light emitting diode (LED) is a light source built up from semiconductor material. In its basic operation a photon is emitted from a forward biased $p$-$n$ junction where an electron-hole pair recombines spontaneously [1]. The main advantages of LEDs are the high efficiency, long lifetime, portability and flexibility [1, 2].

Gallium-arsenide was the material of the first practical light emitting diodes [3]. The two main types of LEDs presently used for lighting systems are aluminum gallium indium phosphide (AlGaInP) alloys for red, orange and yellow LEDs [4] and indium gallium nitride (InGaN) alloys for green, blue and white LEDs [5].

Figure 2.1 (a) shows a double heterostructure LED chip. The chip consists of anode and cathode electrodes, a substrate and the semiconductor layers. Figure 2.1 (b) shows the cross section of a complete LED device.

![Diagrams of LED chip and cross section](image)

**Figure 2.1:** Double heterostructure LED chip geometry (a) and cross section of a complete LED device including reflectors and packaging (b). Usually the die has a high refractive index ($n \approx 3.3$), and the encapsulation has a lower one ($n=1.5$).

The photon generated inside the active layer reaches the air-semiconductor interface and in optimal cases will be transmitted. However due to the usually large refractive index of the semiconductor ($n=3.4$ for GaAs), total internal
reflection occurs at large incident angles and the extraction efficiency of the device is reduced. For example in the case of GaAs the extraction efficiency is only 2-3% at planar semiconductor-air interface. The extraction efficiency can be enhanced by using a packaging material with refractive index between the air and the semiconductor. Usually some kind of polymer is used with refractive index around 1.5 [1]. For planar semiconductor-polymer interface the extraction efficiency is in the range of 5%.

The efficiency of the LED can be enhanced if the reflection losses are diminished at the chip-packaging interface. This can be done by shaping the LED die, for example truncated inverted pyramid structures are used with success [6]. Another solution is to use textured semiconductor surfaces where light can escape through diffraction or scattering [7]. For both cases the extraction efficiency increased to about 50%.

2.2.1 Polarized light emitting diodes

Native LED radiation is unpolarized, but polarized LED would be very useful in several applications like high-contrast imaging [8, 9], optical communications [10] and LCD backlighting [11]. Several papers deal with LEDs having polarized light output. Schubert et al. demonstrated two possible solutions for this problem: a polarization selective encapsulation based on Brewster’s effect for unpolarized white light source [12] and a polarization-enhancing reflector for a partially polarized GaInN LED dies [13].

The idea of inherently polarizing the emitted radiation of an LED is not new. Wheatley et al. patented a solution [14], in which the emitting surface of the LED die is coated with a wire grid polarizer (WGP), and on the opposite surface a polarization recycler takes place. Figure 2.2 shows the working principle of the structure.

The WGP transmits one component of the wave, with polarization perpendicular to the grating lines, and reflects the component of parallel polarization. The light reflected by the WGP has another chance to get out of the LED if its polarization changes inside for example using a polarization recycler. This concept is practically useful if the total polarized output of the LED is significantly higher than the output of a standard LED combined with a standard polarizer. The key component of the previous investigation is an appropriate polarizing structure on the top of the LED die.

Polarizing nanostructures

A widely used solution to produce polarized light with metallic nanostructures is the application of a linear grating having subwavelength grating
Figure 2.2: Schematic view of a polarized LED chip. Emitted light from the active region reaches the upper surface where a polarizer transmits the proper polarization component and reflects the improper one. The polarization of the reflected wave will be changed by a polarization recycler at the bottom. From [14].

period. This device is called a wire grid polarizer (WGP) [15]. The electron-microscope image of the wire grid polarizer from [16] can be seen in Fig. 2.3. In the case the polarization of the incident light is parallel with the wires the electric field induces oscillation of the electrons similarly to continuous metallic interfaces. Thus in this case the wires act as a mirror, reflecting the incident wave. If the polarization is perpendicular to the lines, the displacement of the electrons is confined to the linewidth of the wires, thus the material behaves as a dielectric and the incoming light is transmitted.

Optimal WGPs for visible light have a period of 100-150 nm and grid height of 150-200 nm. The reported optimal line/period ratio for normal incidence is about 0.4-0.5 [17, 18]. Experimental demonstration of such a WGP was reported by Wang et al. with a period of 146 nm, and a line width of 30 nm [16] as well as another one with a period of 118 nm and line width of 40 nm [19].

Light polarization can also be altered using array of nanoparticles with anisotropic shapes. In this case the response of such particles will depend on the polarization of the exciting light [20]. Metallic prolate spheroidal nanoparticles were used as ultrathin polarizers at normal incidence [21].
2.3. Plasmonics Introduction

Figure 2.3: Wire grid polarizer made from aluminum. Image taken from [16].

2.3 Introduction to surface plasmon polaritons and localized surface plasmons

Most of the work performed in this dissertation deals with metallic-dielectric interfaces. On such interfaces the solution of the "classical" Maxwell’s equations can lead to exotic electromagnetic modes, called surface plasmon polaritons. Surface plasmons are quasiparticles consisting of photons coupled to collective excitations of conduction electrons near a metal surface. These modes exhibit a propagation along a planar metal-dielectric interface and decays exponentially perpendicular to the interface [22]. These modes appear at interfaces of free-electron-metals, where the relative dielectric permittivity is

\[
\varepsilon_m(\omega) = 1 - \left( \frac{\omega_p}{\omega} \right)^2,
\]

where \( \omega_p \) is the plasma frequency of the metal.

The basis for the mathematical description of these modes is well known 150 years ago already, still the field started to develop when in 1968 Otto and independently Kretschmann and Raether reported the realization of surface
waves at metallic interfaces [23, 24]. The field developed in the past three decades substantially. The increased interest of plasmonics can be attributed to several factors. First, the fabrication methods of metallic nanostructures developed in the last decades so that we are capable of preparing structured surfaces in the range of nanometers as well as particles of various shapes in the same size range. Second, the measurement methods make possible only nowadays to investigate these structures in more detail, i.e. the local field enhancement can be measured already with near field optical microscopy. Third the possibility to develop new structures requires huge computing power, which is available again nowadays. Fourth, the demand for nanoplasmonics in potential applications, like sensing, solar cells, light emitting diodes increased too in the past years.

The electric field of a surface plasmon polariton can be expressed with

\[ E_{SP} = E_0 e^{ik_{sp}x - k_z|z|}, \]  

(2.2)

where \( E_0 \) is the electric field amplitude, \( k_{sp} \) is the wavenumber of the plasmon mode and \( k_z \) is the factor determining the field decay along the \( z \) direction.

The \( k_{sp} \) dispersion relation for a plane metal-dielectric interface can be calculated using Maxwell’s equations and has the form of

\[ k_{sp} = \frac{\omega}{c} \sqrt{\frac{\varepsilon_d\varepsilon_m}{\varepsilon_d + \varepsilon_m}}, \]  

(2.3)

where \( \omega \) is the angular frequency of light, \( c \) is the speed of light, \( \varepsilon_m \) is the relative permittivity of the metal and \( \varepsilon_d \) is the relative permittivity of the dielectric material. The decay constant \( k_z \) is

\[ k_z^2 = k_{sp}^2 - \left( \frac{\omega}{c} \right)^2 \varepsilon_i, \]  

(2.4)

where \( \varepsilon_i \) is the relative permittivity of either the dielectric or the metal [25].

Figure 2.4 shows the plasmon dispersion relation derived using eq. (2.3) and (2.1). In addition the dispersion relation of the light propagating in the dielectric material is also plotted. The lower branch of the surface plasmon polariton dispersion relation corresponds to the plasmon mode propagating along the metal-dielectric interface. The upper limit of this mode is at \( \omega_p/\sqrt{2} \). The upper branch corresponds to bulk plasmon modes which exist only for \( \omega > \omega_p \). In this case the permittivity is positive and longitudinal mode can propagate in the metal.

It is visible that the wavenumber of the light is always smaller than that of the surface plasmon at a given frequency. This indicates that in order to excite plasmonic modes at interfaces the wavenumber of the light needs to
be complemented. This can be done for example in the Kretschmann geometry, using a coupling prism with a refractive index larger than the dielectric interface where plasmonic propagation is desired [23]. Another possible solution is to use the quasi-impulse of a diffraction grating. Diffraction on a two-dimensional periodic structure provides wavevector conservation if the wavevector of the surface plasmon polariton matches that of the diffracted order:

\[ k_{sp}(\omega) = |\vec{k}_{in,\parallel}(\omega, \theta, \varphi) + n\vec{K}_1 + m\vec{K}_2|, \quad (2.5) \]

where \( k_{in,\parallel} \) is the projection of the incident wavevector parallel with the surface, \( \vec{K}_1 \) and \( \vec{K}_2 \) are the primitive vectors of the reciprocal lattice [26]. In the parentheses \( \theta \) and \( \varphi \) are the incident polar and azimuth angles respectively. For a given \( \omega \) frequency, azimuth angle \( \varphi \) and order denoted by \( n \) and \( m \) the \( \omega-\theta \) dispersion relation can be determined.

The most often used plasmonic materials are gold, silver and aluminum. For these materials the propagation length of the surface plasmon is in the order of 10-20 \( \mu m \), the decay length in metal is several 10 nm, and in air is several 100 nm in the visible regime (\( \approx 500 \) nm).

In addition to surface plasmon polariton modes in particle like geometries exist localized surface plasmon excitations. These modes are confined to bounded geometries like nanoparticles or nanovoids. In contrast to the surface plasmon polaritons the dispersion relation of such modes does not depend on the incident wave vector. The eigenfrequencies depend only on
the size and shape of the particle and its dielectric permittivity. Localized surface plasmons are also bounded to the surface similarly to the propagating ones. In general both propagating and localized surface plasmons provide high electromagnetic field enhancement and tight spatial confinement of length in the range of few 10 nm.

Metallic nanoparticles supporting localized surface plasmons are utilized in many applications like biolabeling, bioimaging, sensors, catalysis, nanodevices and nanoelectronics [27, 28]. The field enhancement of such modes can be exploited by many applications from various optical microscopic techniques such as dark-field optical and two-photon luminescence microscopy [29], to surface enhanced Raman spectroscopy [30, 31] and five-dimensional optical data storage [32]. Other photonic applications include plasmon mode propagation through chains of closely spaced metal nanoparticles [33].

### 2.3.1 Plasmonic nanovoid and nanodome arrays

The main advantage of nanovoid and nanodome arrays is that due to the periodic nature of the structure, light can be directly coupled to localized (‘Mie’ or ‘cavity’ modes) and delocalized (‘Bragg’ modes) surface plasmons [34]. While the localized modes are restricted to the protrusions of the gold layer, the propagating ones extend over the surface of the structure. The theoretical framework for the description and deeper understanding of the light-matter interaction in these quasi 2D ‘plasmonic crystals’ [35, 36] paved the way towards their use in many different research areas and suggests a strong incident angle dependence especially for the Bragg modes at different wavelengths. The strong local electromagnetic fields of the localized plasmon modes were successfully utilized for light management in solar cell applications, where enhanced electromagnetic fields and increased optical path due to scattering were shown to increase the efficiency of inorganic as well as bulk-heterojunction organic and dye sensitized solar cells [37, 38, 39]. Surface enhanced Raman scattering experiment performed on the nanovoid and nanodome arrays indicate that these structures can be very useful in label free analytical approaches [40, 41, 42, 43]. A recent demonstration has also shown that despite the inherently low efficiency of the process, these plasmonic nanostructures can allow the generation and extraction of hot charge carriers [44].

### 2.4 Maxwell’s equations in homogeneous space

The basis of each computational method are Maxwell’s equations.
2.4. MAXWELL'S EQUATIONS IN HOMOGENEOUS SPACE

\[ \nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \quad (2.6) \]
\[ \nabla \times \vec{H} = \vec{J} + \frac{\partial \vec{D}}{\partial t} \quad (2.7) \]
\[ \nabla \vec{B} = 0 \quad (2.8) \]
\[ \nabla \vec{D} = \rho, \quad (2.9) \]

where \( \vec{E} \) is the electric field, \( \vec{D} \) is the dielectric displacement field, \( \vec{H} \) is the magnetic field and \( \vec{B} \) is the magnetic induction. \( \vec{J} \) and \( \rho \) are the free current density and free charge density respectively.

In order to introduce the material parameters into the equations we need the constitutive relations between the fields. Assuming linear and isotropic materials we have the following relations:

\[ \vec{B} = \mu_0 \mu_r \vec{H} \quad (2.10) \]
\[ \vec{D} = \varepsilon_0 \varepsilon_r \vec{E}, \quad (2.11) \]

where \( \varepsilon_0 \) and \( \mu_0 \) is the vacuum permittivity and permeability respectively, \( \varepsilon_r \) and \( \mu_r \) is the relative permittivity and permeability.

Assuming time dependence of \( e^{-i\omega t} \) and using eqs. (2.6) and (2.7) the following wave equation can be derived [45]:

\[ \nabla \times \nabla \times \vec{E} - \frac{\omega^2}{c^2} \varepsilon_r \mu_r \vec{E} = i\omega \mu_0 \vec{J}, \quad (2.12) \]

where \( c \) is the speed of light in vacuum.

The wave equation (2.12) can be solved with Green’s tensor method, where the Green’s tensor (\( \tilde{G}(\vec{r},\vec{r}') \)) is defined by the response at point \( \vec{r} \) given to the point like excitation in position \( \vec{r}' \):

\[ \nabla \times \nabla \times \tilde{G}(\vec{r},\vec{r}') - \frac{\omega^2}{c^2} \varepsilon \mu \tilde{G}(\vec{r},\vec{r}') = \tilde{I}\delta(\vec{r} - \vec{r}'), \quad (2.13) \]

where \( \tilde{I} \) is the unit tensor and \( \delta(\vec{r} - \vec{r}') \) is the Dirac delta.

Then the electric field of an arbitrary current distribution (\( \vec{J}(\vec{r}) \)) can be calculated using the following integral:

\[ \vec{E}(\vec{r}) = i\omega \mu_0 \iiint \tilde{G}(\vec{r},\vec{r}') \vec{J}(\vec{r}') d^3 r' \quad (2.14) \]
2.5 Periodic-layered Green’s tensor method with quasistatic approximation

In this section I review a method, which can approximately describe the electromagnetic scattering on periodic arrangement of small scatterers in a multilayer structure. The method consists of calculating the Green’s tensor in a stratified medium [46, 47] and using this Green’s tensor for periodic structures [48]. The Green’s tensor is then used for the calculation of the light scattering on small particles.

First I describe the light scattering on small particles in the case of homogeneous and layered environment for single and periodic scatterers in the quasistatic approximation. Then the calculation of the corresponding Green’s tensors is described in detail first for homogeneous medium, then for multilayer medium.

2.5.1 Light scattering in homogeneous medium

In the quasistatic approximation the incident electric field induces a dipole moment in the small particle [20]. This induced dipole moment is assumed to be proportional to the driving electric field:

\[ \vec{p} = \varepsilon_0 \varepsilon_{m,r} \bar{\alpha} \vec{E}_{dr}(\vec{r}_0), \]  

where \( \varepsilon_0 \) is the vacuum permittivity, \( \varepsilon_{m,r} \) is the relative permittivity of the medium surrounding the particle, \( \vec{E}_{dr}(\vec{r}_0) \) is the driving electric field defined at the center \( (\vec{r}_0) \) of the particle and \( \bar{\alpha} \) is the polarizability tensor of the particle, which depends on the geometry and material of the particle and the material of the environment.

This approximation is valid if the particle is small compared to the wavelength [20], i.e.

\[ \frac{2\pi}{\lambda} \varepsilon_{m,r}^2 l << 1, \quad \frac{2\pi}{\lambda} \varepsilon_{m,r}^2 |\varepsilon_{p,r}| l << 1, \]  

where \( \lambda \) is the vacuum wavelength, \( l \) is the characteristic linear size of the particle, and \( \varepsilon_{p,r} \) is the relative permittivity of the particle.

Using the fact that in homogeneous medium the driving field is equivalent to the incident field, the scattered field can be calculated with the help of the Green’s tensor \( \bar{G}_h(\vec{r}, \vec{r}') \) of homogeneous space:

\[ \vec{E}_{sc}(\vec{r}) = \omega^2 \mu_0 \bar{G}_h(\vec{r}, \vec{r}_0) \vec{p} = \frac{\omega^2}{c^2} \varepsilon_{m,r} \bar{G}_h(\vec{r}, \vec{r}_0) \bar{\alpha} \vec{E}_{inc}(\vec{r}_0), \]
2.5. PERIODIC-LAYERED GREEN’S TENSOR METHOD ...

The total field outside the particle can be expressed as the sum of the incident and scattered field.

\[ \vec{E}_{\text{tot}}(\vec{r}) = \vec{E}_{\text{inc}}(\vec{r}) + \vec{E}_{\text{sc}}(\vec{r}) \]  \hspace{1cm} (2.18)

2.5.2 Light scattering in layered medium

In the case of layered medium there are two differences compared to the homogeneous case. First, the Green’s tensor corresponds to the layered environment instead of the homogeneous one. Second, the driving field consists not only of the incident field, but it contains the field scattered by the particle and reflected by the layers back to the particle position. Thus a self-driving mechanism appears in this case.

The induced dipole moment can be expressed again with the driving field, defined by the sum of the incident and reflected waves.

\[ \vec{p} = \varepsilon_0 \varepsilon_{m,r} \vec{\alpha} \vec{E}_{\text{dir}}(\vec{r}_0) = \varepsilon_0 \varepsilon_{m,r} \vec{\alpha} \left( \vec{E}_{\text{inc}}(\vec{r}_0) + \vec{E}_{\text{refl}}(\vec{r}_0) \right) \]  \hspace{1cm} (2.19)

The reflected part of the scattered field can be calculated using the Green’s tensor \((\tilde{G}_l)\) of the layered medium:

\[ \vec{E}_{\text{refl}}(\vec{r}_0) = \omega^2 \mu_0 \tilde{G}_l(\vec{r}_0, \vec{r}_0) \vec{p} - \vec{E}_{\text{dir}}(\vec{r}_0) = \omega^2 \mu_0 \tilde{G}_l(\vec{r}_0, \vec{r}_0) \vec{p} \]  \hspace{1cm} (2.20)

where \(\vec{E}_{\text{dir}}\) is the directly emitted part of the electric field and \(\tilde{G}_l(\vec{r}_0, \vec{r}_0)\) is the Green’s tensor containing only the reflected part of the electric field at the source position. The direct part of the electric field is actually equivalent to the field emitted in homogeneous space. Thus the Green’s tensor of the reflected field \((\tilde{G}_l'(\vec{r}_0, \vec{r}_0))\) is defined as the difference of the layered and the homogeneous Green’s tensor.

Combining eqs. (2.19) and (2.20):

\[ \vec{p} = \varepsilon_0 \varepsilon_{m,r} \vec{\alpha'} \vec{E}_{\text{inc}}(\vec{r}_0) \]  \hspace{1cm} (2.21)

where \(\vec{\alpha'}\) is defined by

\[ \vec{\alpha'} = \left( \vec{\alpha}^{-1} - \frac{\omega^2}{c^2} \varepsilon_{m,r} \tilde{G}_l'(\vec{r}_0, \vec{r}_0) \right)^{-1} \]  \hspace{1cm} (2.22)
Thus a similar equation to eq. (2.15) was found, but the polarizability tensor now contains the self driving term in itself. With this the scattered field at any desired position outside the scatterer can be calculated:

\[
\vec{E}_{sc}(\vec{r}) = \omega^2 \mu_0 \vec{G}_l(\vec{r}, \vec{r}_0) \vec{p} = \frac{\omega^2}{c^2} \vec{G}_l(\vec{r}, \vec{r}_0) \varepsilon_m r \vec{\alpha} \vec{E}_{inc}(\vec{r}_0)
\]  

(2.23)

### 2.5.3 Periodic array of scatterers in layered medium

In the case of multiple particles the scattered electric field can be calculated by the sum of the contribution of each particle. However the driving field of a single particle in this case also consists of the incident field, the scattered field of the other particles and the reflected field of the particle itself. Accordingly we get a coupled problem, where the particles influence each other, i.e. there is electromagnetic interaction between the particles.

Let us assume that an incident plane wave ($\vec{E}_{inc}$) is scattered by a periodic array of small identical scatterers in a multilayer background. The multiple layers are stacked on each other in the z direction, and the particles have the same $z_0$ coordinate. If we know the polarizability of each particle we can formally express the driving electric field at any particle position:

\[
\vec{E}_{dr}(\vec{r}_l) = \vec{E}_{inc}(\vec{r}_l) + \omega^2 \mu_0 \left( \sum_{j \neq l} \vec{G}_l(\vec{r}_l, \vec{r}_j) \vec{p}_j + \vec{G}_l'(\vec{r}_l, \vec{r}_l) \vec{p}_l \right) = \\
= \vec{E}_{inc}(\vec{r}_l) + \frac{\omega^2}{c^2} \varepsilon_m r (\sum_{j \neq l} \vec{G}_l(\vec{r}_l, \vec{r}_j) \vec{\alpha} \vec{E}_{dr}(\vec{r}_j) + \vec{G}_l'(\vec{r}_l, \vec{r}_l) \vec{\alpha} \vec{E}_{dr}(\vec{r}_l))
\]  

(2.24)

If the angle of incidence of the incident plane wave is oblique, i.e. the parallel components of the incident wave vector ($\vec{k}_\parallel$) is nonzero, the resulting electric field is Bloch-periodic with a Bloch-vector of $\vec{k}_\parallel$. In this case we can express the driving electric field with an amplitude ($\vec{E}$) and a phase term:

\[
\vec{E}_{dr}(\vec{r}_l) = e^{i\vec{k}_\parallel \cdot \vec{r}_l} \vec{E}.
\]  

(2.25)

Substituting eq. (2.25) into eq. (2.24):

\[
\vec{E} e^{i\vec{k}_\parallel \cdot \vec{r}_l} = \vec{E}_{inc}(\vec{r}_l) + \frac{\omega^2}{c^2} \varepsilon_m r (\sum_{j \neq l} \vec{G}_l(\vec{r}_l, \vec{r}_j) e^{i\vec{k}_\parallel \cdot \vec{r}_j} + \vec{G}_l'(\vec{r}_l, \vec{r}_l) e^{i\vec{k}_\parallel \cdot \vec{r}_l} ) \vec{\alpha} \vec{E} = \\
= \vec{E}_{inc}(\vec{r}_l) + \frac{\omega^2}{c^2} \varepsilon_m r \vec{G}_k(\vec{r}_l) \vec{\alpha} \vec{E},
\]  

(2.26)
where
\[
\tilde{G}_k(\vec{r}) := \sum_{j \neq l} \tilde{G}_l(\vec{r}_l, \vec{r}_j) e^{i\vec{k}_j \cdot \vec{r}_l} + \tilde{G}_l^*(\vec{r}_l, \vec{r}_l) e^{i\vec{k}_l \cdot \vec{r}_l}
\] (2.27)

This expression is called the lattice sum and is a very important quantity in all field of physics of periodic structures [49]. The amplitude of the driving electric field can be calculated by solving eq. (2.26):
\[
\vec{E} = \left( \frac{\pi e^{i\vec{k} \cdot \vec{r}_l}}{c^2} \varepsilon_{m,r} \tilde{G}_k(\vec{r}_l) \tilde{\alpha} \right)^{-1} \vec{E}_{inc}(\vec{r}_l)
\] (2.28)

After the calculation of \( \vec{E} \), the electric field in the whole space can be calculated by the sum of the contribution of each scatterer.

We get a similar expression to eq. (2.21) if we express the induced dipole moment of the \( l \)-th particle:
\[
\tilde{p}_l = \varepsilon_{m,r} \varepsilon_0 \tilde{\alpha} \vec{E} e^{i\vec{k} \cdot \vec{r}_l} = \varepsilon_{m,r} \varepsilon_0 \left( \tilde{\alpha}^{-1} - \frac{\omega^2}{c^2} \varepsilon_{m,r} \tilde{G}_k(\vec{r}_l) e^{-i\vec{k} \cdot \vec{r}_l} \right)^{-1} \vec{E}_{inc}(\vec{r}_l)
\] (2.29)

From this result we can obtain again an effective polarizability which accounts for the effect of the environment and the other particles:
\[
\tilde{\alpha}' = \left( \tilde{\alpha}^{-1} - \frac{\omega^2}{c^2} \varepsilon_{m,r} \tilde{G}_k(\vec{r}_l) e^{-i\vec{k} \cdot \vec{r}_l} \right)^{-1}
\] (2.30)

The scattered field in the whole space can be calculated with the following sum:
\[
\vec{E}_{sc}(\vec{r}) = \frac{\omega^2}{c^2} \varepsilon_{m,r} \sum_j \tilde{G}_l(\vec{r}, \vec{r}_j) e^{i\vec{k} \cdot \vec{r}_j} \tilde{\alpha} \vec{E}
\] (2.31)

If we are interested only in the farfield, i.e. the transmitted and reflected power needs to be calculated, it is straightforward to express the lattice sum in eq. (2.31) using two dimensional spectral expansion of the single particle Green’s tensor:
\[
\sum_j \tilde{G}(\vec{r}_j) e^{i\vec{k} \cdot \vec{r}_j} =
\]
\[
= \frac{1}{4\pi^2} \sum_j \int \int \tilde{G}(K_x, K_y, z, z_0) e^{iK_{\parallel} \cdot (\vec{r}_j - \vec{r}_j)} dK_x dK_y e^{i\vec{k} \cdot \vec{r}_j} =
\]
\[
= \frac{1}{4\pi^2} \int \int \tilde{G}(K_x, K_y, z, z_0) e^{iK_{\parallel} \cdot \vec{r}_j} \left( \sum_j e^{i(K_{\parallel} \cdot \vec{r}_j)} \right) dK_x dK_y,
\] (2.32)
where $\tilde{G}(K_x, K_y, z, z')$ is the two dimensional Fourier-transform of the single particle Green’s tensor.

The formula between the parentheses can be expressed with the Dirac-delta comb and the reciprocal lattice vectors ($\vec{K}_j$):

$$
\sum_j e^{i(\vec{k}_\parallel - \vec{K}_j) \vec{r}_j,\parallel} = \frac{1}{A} \sum_j \delta \left( \vec{k}_\parallel - \vec{K} - \vec{K}_j \right), \quad (2.33)
$$

where $A$ is the area of the primitive cell. Using this identity the lattice sum can be expressed with the discrete sum of the Fourier-transform of the Green’s tensor:

$$
\sum_j \tilde{G}(\vec{r}, \vec{r}_j) e^{i\vec{k}_\parallel \vec{r}_j,\parallel} = \frac{1}{A} \sum_j \tilde{G}(k_x - K_{j,x}, k_y - K_{j,y}, z, z_0) e^{i(\vec{k}_\parallel - \vec{K}_j) \vec{r}_j,\parallel}. \quad (2.34)
$$

This expression speeds up the calculations because in layered systems first the two dimensional Fourier transform of the Green’s tensor is calculated anyway. From this expression we can also observe the well known physical principle, that the scattering on periodic structures occurs in discrete diffractive orders with parallel component of the wave vector of $\vec{k}_\parallel - \vec{K}_j$.

In the numerical implementation the only difficulty lies in the calculation of the $\tilde{G}_k(\vec{r})$ lattice sum, which is used for the calculation of the effective polarizability in eq. (2.30). The convergence of the summation is quite slow, which can be sped up by Ewald summation and summation in the spectral domain [49].

### 2.5.4 Green’s tensor in homogeneous environment

Until now I have dealt with the formalism of the electromagnetic scattering of a single particle and multiple particles. This was done without the exact definition of the Green’s tensor corresponding to the actual background. In the following subsections I review the Green’s tensor for homogeneous environment and also for layered background. Note however that the previous subsections are valid in different environments too, if we know the corresponding Green’s tensor.
2.5. **PERIODIC-LAYERED GREEN’S TENSOR METHOD** ...

The Green’s tensor solution of eq. (2.13) for homogeneous space [50, 47]:

\[
\bar{G}(\vec{r}, \vec{r}') = \left( I - \frac{\nabla \circ \nabla}{k^2} \right) \frac{e^{ik|r-r'|}}{4\pi |r-r'|} = \\
\left( I + \frac{i k |r-r'|}{k^2 |r-r'|^2} \hat{I} + \\
\frac{3 - 3ik |r-r'| - k^2 |r-r'|^2}{k^2 |r-r'|^4} \left( \vec{r} - \vec{r}' \right) \circ \left( \vec{r} - \vec{r}' \right) \right) \frac{e^{ik|r-r'|}}{4\pi |r-r'|} 
\]

(2.35)

where \( k = \omega/c\sqrt{\epsilon_r} \) and \( \circ \) denotes the dyadic product.

### 2.5.5 Two-dimensional spectral expansion of the Green’s tensor

In multilayer system instead of the Green’s tensor of the homogeneous environment the Green’s tensor of the particular multilayer geometry needs to be calculated, which can be only done by numerical calculations in general. The calculation of the multilayer Green’s tensor consists of the plane wave expansion of the homogeneous Green’s tensor, then the plane waves are propagated one by one through the system. Finally at the output position the contributions of the particular plane waves are summed.

The two dimensional plane wave expansion of the homogeneous Green’s tensor can be calculated in the following way [50, 47]:

\[
\bar{G}(\vec{r}, \vec{r}') = -\hat{z} \circ \hat{z} \frac{\delta(\vec{r} - \vec{r}')}{k^2} + i \frac{1}{8\pi^2} \int \int \frac{1}{k_z} \left[ \hat{I} - \frac{\vec{k}^+ \circ \vec{k}^+}{k^2} \right] e^{ik^+(r-r')} dk_x dk_y, 
\]

(2.36)

where \( \vec{k}^\pm = k_x \hat{x} + k_y \hat{y} \pm k_z \hat{z} \), and the \(+/-\) sign is valid above/below \( z' \).

In the \( k \)-space instead of Descartes-coordinate system we can use local coordinate system for each \( k \)-vector. Basis vectors are \( \hat{k}^\pm, \hat{e}^\pm \) and \( \hat{h}^\pm \).

\[
\hat{k}^\pm = \frac{1}{k} (k_x \hat{x} + k_y \hat{y} \pm k_z \hat{z}) 
\]

(2.37)

\[
\hat{e}^\pm = \frac{\hat{k}^\pm \times \hat{z}}{|\hat{k}^\pm \times \hat{z}|} = \frac{1}{\sqrt{k_x^2 + k_y^2}} \left( k_y \hat{x} - k_x \hat{y} \right) 
\]

(2.38)

\[
\hat{h}^\pm = \hat{e}^\pm \times \frac{\hat{k}^\pm}{k} = \frac{1}{k \sqrt{k_x^2 + k_y^2}} \left( \mp k_x k_z \hat{x} \mp k_y k_z \hat{y} + (k_x^2 + k_y^2) \hat{z} \right) 
\]

(2.39)
We can think about $\hat{e}^\pm$ and $\hat{h}^\pm$ as the transverse electric (TE) and transverse magnetic (TM) direction unit vectors. Using these basis functions and omitting the Dirac-delta term we get the following expression for the homogeneous space Green’s tensor:

$$ \tilde{G}(\vec{r},\vec{r}') = \frac{i}{8\pi^2} \int \int \frac{1}{k_z} \left[ \hat{e}^\pm \circ \hat{e}^\pm + \hat{h}^\pm \circ \hat{h}^\pm \right] e^{ik_z(r-r')} dk_x dk_y, $$

where both $\hat{e}^\pm$ and $\hat{h}^\pm$ depend on $k_x$, $k_y$, and $k_z$.

From here we can get the two-dimensional Fourier-transform of the Green’s tensor:

$$ \tilde{G}(k_x, k_y, z, z') = \frac{i}{2k_z} \left[ \hat{e}^\pm \circ \hat{e}^\pm + \hat{h}^\pm \circ \hat{h}^\pm \right] e^{\pm ik_z(z-z')} $$

The $ij$-th element of the Green’s tensor is calculated using the following relation:

$$ G_{ij}(k_x, k_y, z, z') = \hat{i} \left( \frac{i}{2k_z} \left[ \hat{e}^\pm \circ \hat{e}^\pm + \hat{h}^\pm \circ \hat{h}^\pm \right] e^{\pm ik_z(z-z')} \right) = \hat{i} \left( A_{e,j}^\pm(z, z') \hat{e}^\pm + A_{h,j}^\pm(z, z') \hat{h}^\pm \right) = \hat{i} \tilde{E}_j, $$

where $\hat{i}$ and $\hat{j}$ are the unit vectors in the $i$ and $j$ direction respectively and $A_{e,\beta}^\pm(z, z') = i/(2k_z)\hat{e}^\pm \beta e^{\pm ik_z(z-z')}$ and $A_{h,\beta}^\pm(z, z') = i/(2k_z)\hat{h}^\pm \beta e^{\pm ik_z(z-z')}$. Thus $G_{ij}(k_x, k_y)$ can be calculated as a scalar product of a basis vector ($\hat{i}$) and a quantity which is proportional to the electric field produced by a source of direction $\hat{j}$. This electric field is decomposed into TE and TM part and contains propagation either to the positive or the negative $z$-direction.

### 2.5.6 Green’s tensor in multilayer medium

To calculate a single spectral component of the Green’s tensor in a multilayer medium corresponding to the wave vector $(k_x, k_y)$, the previously introduced $\tilde{E}$ field is written in each layer as a sum of upward and downward propagating waves. The resulting field for a single plane wave component is calculated by propagating the field through the layered system and is realized numerically with a standard transfer matrix calculation method [47].

To get the multilayer Green’s tensor in the coordinate space we need to sum up the contributing plane waves at the desired location. This is actually equivalent to finding the two dimensional Fourier-transform of $G_{ij}(k_x, k_y)$. However instead of calculating the two dimensional Fourier transform in
Descartes coordinates we can change to cylindrical coordinate system both in the $k$-space and the coordinate space:

\[
G_{ij}(\vec{r}, \vec{r}') = \frac{1}{4\pi^2} \int_0^{2\pi} \int_0^{2\pi} G_{ij}(k_\rho, k_\phi, z, z') e^{i k_\rho (\cos k_\phi \cos \phi + \sin k_\phi \sin \phi) k_\rho} dk_\rho dk_\phi d k_\rho
\]

(2.43)

The integration over $k_\phi$ can be evaluated analytically. Then we get an integral of the following form:

\[
G_{ij}(\vec{r}, \vec{r}') = \frac{1}{4\pi^2} \int_0^{2\pi} \left( G^e_{ij}(k_\rho) + G^h_{ij}(k_\rho) \right) dk_\rho,
\]

(2.44)

where $G^e_{ij}(k_\rho)$ and $G^h_{ij}(k_\rho)$ are defined in Appendix B of [46] (in that article $G^e$ is denoted by $f^s$ and $G^h$ is denoted by $f^p$). Finally only a one-dimensional integral needs to be evaluated numerically in order to get the multilayer Green’s tensor at the desired location [46].

The previously presented method to calculate electromagnetic scattering on dipolar scatterers in multilayer background involves only $3 \times 3$ matrix products and inversions, one dimensional numerical integrals, thus it is very fast and uses very small computer power. The presented method is appropriate to be used for parameter sweeps and optimization.

2.6 Rigorous Coupled Wave Analysis

Rigorous coupled wave analysis is used to solve Maxwell’s equations with Floquet-periodic boundary conditions in one or two dimensions [51, 52]. The method can be explained most easily in the one-dimensional binary case, but the method can extended to two dimensional periodicity [53] and multilayer structures [51].

Let us consider the two-dimensional computational geometry in the $xz$ plane shown in Fig. 2.5. The figure shows two periods of a binary grating. The refractive index of the substrate and superstrate are $n_I$ and $n_{II}$ respectively, the incident angle of the exciting wave is $\theta$. The grating consists of two alternating materials of refractive index of $n_1$ and $n_2$ and length of $f \Lambda$ and $(1-f)\Lambda$ respectively, where $\Lambda$ is the grating period and $f$ is the fill factor. The thickness of the layer is $d$.

For such periodic structures, it seems straightforward to look for the electric and magnetic field in terms of Fourier series during the solution of the
Maxwell’s equations. First we can divide the problem to two independent polarization states. In the case of transverse electric (TE) polarization the electric field is perpendicular to the plane of incidence and the magnetic field lies in the plane of incidence. In the transverse magnetic (TM) case the magnetic field is perpendicular to the plane of incidence and the electric field lies in the plane of incidence. We will see that in case of nonmagnetic materials the transverse magnetic (TM) polarization is more difficult to handle, thus we use this polarization to illustrate the method. The electric and magnetic field has the form of

\[ \vec{E}(x, z) = E_x(x, z)\hat{x} + E_z(x, z)\hat{z}, \quad \vec{H}(x, z) = H_y(x, z)\hat{y}, \]  

where \( \hat{x}, \hat{y} \) and \( \hat{z} \) are the unit vectors in the corresponding direction.

Assuming \( e^{-i\omega t} \) time dependence and looking for solutions without current sources, eqs. (2.6) and (2.7) become:

\[ \nabla \times \vec{E} = i\omega \vec{B}, \quad \nabla \times \vec{H} = -i\omega \vec{D} \]  

Substituting eqs. (2.45) into eqs. (2.46) we get the following three coupled differential equations:

\[ \frac{\partial H_y}{\partial z} = i\omega \varepsilon_0 \varepsilon_r E_x \]  
\[ \frac{\partial H_y}{\partial x} = -i\omega \varepsilon_0 \varepsilon_r E_z \]  
\[ \frac{\partial E_x}{\partial z} - \frac{\partial E_z}{\partial x} = i\omega \mu_0 H_y \]
The tangential field components in domain $I$ and domain $II$ are defined as:

$$H_{I,y}(x, z) = H_{0,y} \exp \{ -i(k_{x,0}x + k_{z,0,I}z) \} + \sum_{m} R_{m} \exp \{ -i(k_{x,m}x - k_{z,m,I}z) \}$$

$$E_{I,x}(x, z) = E_{0,x} \exp \{ -i(k_{x,0}x + k_{z,0,I}z) \} + \frac{1}{\omega \varepsilon_{0}\varepsilon_{r,I}} \sum_{m} k_{z,m,I} R_{m} \exp \{ -i(k_{x,m}x - k_{z,m,I}z) \}$$

$$H_{II,y}(x, z) = \sum_{m} T_{m} \exp \{ -i(k_{x,m}x + k_{z,m,II}(z - d)) \}$$

$$E_{II,x}(x, z) = -\frac{1}{\omega \varepsilon_{0}\varepsilon_{r,II}} \sum_{m} k_{z,m,II} T_{m} \exp \{ -i(k_{x,m}x + k_{z,m,II}(z - d)) \}$$

where $k_{z,m,1/II}^2 = k_{0}^2 n_{1/II}^2 - k_{x,m}^2$, $R_{m}$ and $T_{m}$ are the field amplitudes of the reflected and transmitted orders respectively and $H_{0,y}$ and $E_{0,x}$ are the complex amplitudes of the incident planewave.
Differentiation of the field components corresponding to the \( z \) coordinate is equivalent with the differentiation of the \( z \)-dependent Fourier coefficient, whereas differentiation corresponding to the \( x \) coordinate is equivalent with multiplying the Fourier coefficients by \( ik_{x,m} \).

The Fourier-series of the product of two functions can be calculated using Laurent’s rule:

\[
    f(x)g(x) = \left( \sum_m f_m e^{i(k_{x,m} - k_{x,0})x} \right) \left( \sum_n g_n e^{ik_{x,n}x} \right) = \sum_j \left( \sum_k f_{j-k} g_k \right) e^{ik_{x,j}x}
\]  

(2.60)

Define the Toeplitz matrix by \( \|f\| \), where \( \|f\|_{i,j} = f_{i-j} \). With this notation the new Fourier coefficients are calculated as

\[
    [f \times g] = \|f\| [g],
\]  

(2.61)

In numerical calculations the original function is always approximated with a finite number of Fourier-coefficients. It has been shown, that the finite Fourier-expansion of the product of two functions does not converge uniformly, if the two functions have concurrent jumps [54]. For example in eq. (2.47) the left hand side is a continuous function, so shall be the right hand side. However the right hand side is a product of two discontinuous functions \( E_x \) and \( \varepsilon_r \), with concurrent jumps. Thus Laurent’s rule cannot be applied in this case to obtain the Fourier coefficients of the product. Instead in this case according to [54] the inverse rule should be used. Taking this into consideration eqs. (2.47)-(2.49) can be rewritten for the Fourier coefficients:

\[
    \frac{\partial [H_y]}{\partial z} = i\omega \varepsilon_0 \| \frac{1}{\varepsilon_r} \|^{-1} [E_x] 
\]  

(2.62)

\[
    i\overline{k}_x [H_y] = -i\omega \varepsilon_0 \| \varepsilon_r \| [E_x] 
\]  

(2.63)

\[
    \frac{\partial [E_x]}{\partial z} - \overline{k}_x [E_x] = i\omega \mu_0 [H_y]
\]  

(2.64)

where \( \overline{k}_x \) is the diagonal matrix composed of the \( k_{x,m} \) elements. Combining these equations to express the Fourier coefficients of \( H_y \), we get a differential equation similar to (2.50):

\[
    \frac{\partial^2 [H_y]}{\partial z^2} = \| \frac{1}{\varepsilon_r} \|^{-1} \left( \overline{k}_x \| \varepsilon_r \|^{-1} \overline{k}_x - \mu_0 \| \mu_r \| \right) [H_y]
\]  

(2.65)

The solution of the set of differential equation can be found by finding the eigenvectors and eigenvalues of the system matrix on the left side of eq.
(2.65):
\[
\bar{A} = \left( \frac{1}{\mu_{xx}} \right)^{-1} \left( \frac{1}{k_0^2} \kappa_x \mu_{zz} \| \kappa_x - \| \epsilon_{yy} \| \right) = \bar{V} \bar{D} \bar{V}^{-1},
\]
where \( \bar{V} \) is the matrix composed by the column eigenvectors and \( \bar{D} \) is the diagonal matrix of the eigenvalues of \( \bar{A} \).

The solution of eq. (2.65) has the following form using the eigenvectors and eigenvalues:
\[
[H_y] = \bar{V} \left( \exp \left\{ k_0 \sqrt{D} (z - d) \right\} [c^+] + \exp \left\{ -k_0 \sqrt{D} z \right\} [c^-] \right) \tag{2.67}
\]
\[
[E_x] = \frac{k_0}{i \omega \varepsilon_0} \left\| \bar{V} \sqrt{D} \left( \exp \left\{ k_0 \sqrt{D} (z - d) \right\} [c^+] - \exp \left\{ -k_0 \sqrt{D} z \right\} [c^-] \right) \right\| \tag{2.68}
\]
where \([c^+]\) is the coefficient vector to satisfy the boundary conditions, i.e. \( H_y \) and \( E_x \) are continuous at \( z = 0 \) and \( z = d \).

Combining eqs. (2.67-2.68) with eqs. (2.56-2.59) the unknown coefficients \([c^+], [c^-], R_m, T_m\) can be calculated.

The equations can be generalized to oblique incidence to two dimensional gratings and to stack of gratings. The method is very fast for one dimensional gratings but becomes more demanding for two dimensional gratings since the number of the unknowns is the product of the number of orders in two dimensions.

2.7 Other numerical methods

2.7.1 Finite difference time domain method

Finite difference time domain (FDTD) method is a widely used tool for electromagnetic modeling [55]. Its application areas range from industrial products (solar cells [56], OLEDs [57]) to fundamental research problems (surface plasmons [58], metamaterials [59]). The main advantage of the method is the flexibility and scalability. It is very easy to use the method in different geometries and the method can handle even large computational sizes compared to other numerical solutions of the Maxwell’s equations.

The method solves the time dependent Maxwell’s equations with finite differences both in the time and the spatial domain on a rectangular grid using a special computational unit called the Yee-cell [60]. During the calculation the time evolution of the electromagnetic fields is calculated step by step in the time domain: the fields corresponding to a given timestep are derived from that of the previous one.
The FDTD grid is inherently a tetragonal grid because of the geometry of the Yee-cell. The grid geometry makes it rather hard to mesh curved surfaces. This drawback however can be diminished by subpixel averaging techniques [61], and local mesh refinement [55]. Even with subpixel averaging this method is not really suitable for geometries where the resolution needs to be very fine at a particular point like in nanoparticle scattering, where other methods e.g. finite element method are more appropriate.

At the side of the computational volume different boundary conditions can be set up. These are perfect electric conductor (PEC), perfect magnetic conductor (PMC), periodic and Bloch-periodic boundary conditions. Although not a boundary condition in the mathematical sense, a very useful kind of termination is the perfectly matched layer (PML) [62]. This is rather a specific material, which absorbs all incoming waves without reflecting them. In practice some minor reflection always occurs, but reflection on PML is in the order of -10 dB even with only 10 PML cells. Thus scattering problems can be realized with this type of boundary condition fairly precisely.

Wave sources can be set up by defining current sources inside the computational volume. Scattering problems can be set up using the total field-scattered field formalism [55].

Materials in FDTD are set up either with wavelength independent material parameters, or with defining auxiliary equations for the polarizability of the material, which are also solved simultaneously with the Maxwell’s equations [55]. With this technique only specific time and frequency dependent material models can be described. In general the electric susceptibility can be written as a sum of individual resonance terms:

$$\chi(\omega) = \chi_0 + \chi_D(\omega) + \sum_j \chi_{L,j}(\omega),$$  \hspace{1cm} (2.69)

where $\chi_D(\omega)$ describes the Drude model of the susceptibility:

$$\chi_D(\omega) = -\frac{\omega_D^2}{\omega^2 + i\omega \Gamma_D},$$  \hspace{1cm} (2.70)

and $\chi_{L,j}(\omega)$ corresponds to the Drude-Lorentz model:

$$\chi_{L,j}(\omega) = -\frac{f_{L,j} \omega_{L,j}^2}{\omega_{L,j}^2 - \omega^2 - i\omega \Gamma_{L,j}}.$$

(2.71)

A wavelength dependent material then can be represented by defining the $\chi_0, \chi_D, \Gamma_D, f_{L,j}, \omega_{L,j}$ and $\Gamma_{L,j}$ parameters. These parameters are obtained by fitting on the measured frequency dependent complex refractive index.

Available commercial and free FDTD programs:
2.7. OTHER NUMERICAL METHODS

- Lumerical FDTD Solutions (commercial)
  (https://www.lumerical.com/tcad-products/fdtd/)
- Fullwave (commercial)
- CST Microwave Studio (commercial)
  (https://www.cst.com/products/cstmws/)
- Meep (open source)
  (http://ab-initio.mit.edu/wiki/index.php/Meep)
- Angora (open source)
  (http://angorafdtd.org/)
- Electromagnetic Template Library (free)
  (http://fdtd.kintechlab.com/en/start)

2.7.2 Finite element method

The finite element method (FEM) is another numerical method that can be used to solve Maxwell’s equations with appropriate boundary conditions [63, 64, 65]. The main idea behind the method the representation of the computational volume with smaller sub-domains, called finite elements. The differential equation is applied to each single element using an integro-differential form, called "weak" form. In a single finite element the unknown variable is approximated with a sum of polynomials (called "shape" functions) of unknown coefficients. The accuracy of the solution depends, among other factors, on the order of these polynomials, which may be linear, quadratic, or higher order. After a set of linear equations is obtained for each element in the discretized domain, a global matrix system is formed.

The shape of the elements is polyhedron in three dimension. Usually tetrahedron and cuboid are the most often used shapes. Since the size and shape of the elements is variable, the mesh can be adapted to the actual geometry with great precision. Thus the method is especially suited for problems where the geometry varies at a fine scale.

For this method Maxwell’s equations are formulated most often in the time independent form. That means that a single calculation corresponds to a single frequency. Finite spectral width can be modeled using a frequency sweep. Frequency dependent materials can be modeled using the complex refractive index.

The computational volume is terminated with the same boundary conditions as that of the FDTD: PEC, PMC, periodic, Bloch and PML.
Wave sources can be set up either by current sources or by defining excitation ports.

Available finite element software:

- Comsol Multiphysics (commercial)  
  (https://www.comsol.com/)

- Ansys HFSS (commercial)  
  (http://www.ansys.com/products/electronics/ansys-hfss)

- CST Microwave Studio (commercial)  
  (https://www.cst.com/products/cstmws/)

- deal.II (open source)  
  (http://www.dealii.org/)

2.7.3 Ray tracing method

In this thesis I use nonsequential ray tracing to model a complete LED structure. Geometrical optical ray tracing method is used for the modeling of large scale optical systems [66, 67]. The method works within the validity range of geometrical optics, where light is represented by directional rays propagating in straight direction between two interfaces. The properties of a single ray are the intensity, direction and wavelength. Interfaces are treated as planes locally, where a ray is divided into transmitting and reflecting rays, propagating further with the intensity corresponding to the transmittance and reflectance respectively.

Usually nonsequential ray tracing softwares utilize Monte-Carlo method. That means a large number of rays are launched from the light source with random states. For example in the case of an LED chip rays are launched from the active region with random direction, polarization and position. One-by-one each ray is propagated through the system, and at each interface the ray is transmitted, reflected or absorbed with the probability equal to the transmittance, reflectance and absorbance of the interface. Finally the ray is terminated if it escapes the system or reaches an absorbing detector. In the case of launching many rays the method approximates the real operation of the optical system.

The method is valid if the length scales of the objects are much larger than the wavelength ($> 10 – 100\mu m$). Nano and microstructures can thus inherently not represented in ray tracing calculations. A workaround is to calculate the response of the nanostructured interface in a separate electromagnetic model and then use the farfield results in the ray tracing code.
2.7. OTHER NUMERICAL METHODS

Ray tracing programs:

- Zemax Optic Studio (commercial)
  (http://www.zemax.com/)

- Code V (commercial)
  (https://optics.synopsys.com/codev/)
CHAPTER 2. SUMMARY OF LITERATURE
Chapter 3

Polarizing properties of silver nanoellipsoid arrays

3.1 Introduction

In this chapter I am investigating arrays of silver nanoellipsoids for polarized light extraction in conventional LED applications. The working principle is based on exploiting different surface plasmon resonances belonging to incident wave polarization parallel with the major or the minor axis of the ellipsoids. The array should transmit the waves with allowed polarization and block the ones with perpendicular polarization.

The device is described with a simple model consisting of the interface between the chip and the packaging. The whole LED chip is described with a homogeneous refractive index, but the model provided can be easily extended to a multilayer structure. The polarizing structure consists of silver prolate spheroidal particles having two equal minor axes and a single major axis. The particles are placed on top of the LED chip in a two-dimensional square lattice in order to eliminate lattice anisotropy. The nanoparticles are assumed to be oriented in the same direction. The schematic of the investigated geometry is shown in Fig. 3.1 in a cross section view.

In a conventional LED the sources of the radiation can be described by dipole emitters [1]. If we assume that the polarization of the emitters is random, the radiation is unpolarized in average. In order to model this kind of unpolarized radiation the LED source can be modeled as a single unpolarized dipole emitter by the incoherent summation of three dipole sources with orthogonal polarizations.

The polarizer should work in "reflective mode", i.e. the waves with the wrong polarization should be reflected rather than absorbed. This makes
CHAPTER 3. SILVER NANOELLIPSOID ARRAYS

Figure 3.1: Side view of the schematics of the investigated planar LED structure with silver nanoellipsoids on top. Substrate corresponds to InGaN (grey, $n_1 = 2.45$), superstrate corresponds to polymer encapsulation ($n_2 = 1.5$). Prolate spheroid silver nanoparticles in two-dimensional square lattice are represented with turquoise color (observed from the side). Dipole source is represented with black star in the middle.

The nanostructure a potential candidate for using with polarization recycling layers in combination. These layers transform the reflected waves having inappropriate polarization to waves with proper polarization, thus they have another chance to escape from the system and are not lost. With this solution the efficiency of the polarized LED can be highly enhanced.

The investigation of the polarized LED source tackles us with the question how we measure the quality of such a device. The problem is that the source is not collimated, therefore the polarization is not adequately defined. Throughout this section I define the polarization state by placing a linear polarizer above the source and measure the power transmitted through the polarizer. This definition is not general, but it is a good starting point to characterize the polarizing interface. In the case the polarizing array is developed for a particular application other definitions might be more useful.

The most important quantities which describe the characteristic of the device are the degree of polarization (DOP), transmittance, reflectance and absorbance of the interface shown in Fig. 3.1 as the function of the geometrical parameters of the nanoparticle lattice. The aim of this chapter is to optimize the geometry to reach high enough DOP and very low absorbance.

3.2 Electromagnetic model

Electromagnetic calculations are carried out in order to characterize the device quantitatively. The description of the particles is performed through the polarizability tensor of an ellipsoid [20]:
3.2. ELECTROMAGNETIC MODEL

\[ \alpha_{ij} = \frac{4\pi l_x l_y l_z}{3} \frac{\varepsilon_p - \varepsilon_h}{\varepsilon_h + L_i(\varepsilon_p - \varepsilon_h)} \delta_{ij}, \]  

(3.1)

where \( l_x, l_y \) and \( l_z \) are the length of the semi-principal axes, \( \varepsilon_p \) is the electric permittivity of the particle, \( \varepsilon_h \) is the electric permittivity of the host material and \( \delta_{ij} \) is the Kronecker delta. The \( L_i \) parameters are called shape factors and their calculation can be found in [20].

With increasing nanoparticle size the quasistatic approximation becomes more and more inaccurate. Radiation damping and dynamic depolarization begin to play an important role even in the case when the scattered fields are still of dipolar nature. Introducing the modified long wavelength approximation (MLWA) into the dipolar polarizability the validity of the dipolar approximation can be ensured for particles as large as the tenth of the wavelength:

\[ \tilde{\alpha}_{ij} = \alpha_{ij} \left( 1 - \frac{k_h^2}{4\pi l_i} \alpha_{ij} - i \frac{1}{6\pi} k_i^3 \alpha_{ij} \right)^{-1}, \]  

(3.2)

where \( k_h \) is the wavenumber in the host material [30].

For nanoparticles of size smaller than mean free path of the conductive electrons (\( \approx 50 \) nm in metals like gold and silver), the dielectric function of the particles is modified to take the intrinsic size effect into account [68, 69]. Figure 3.2 shows the relative permittivity with intrinsic size effect corrections for silver nanospheres of different sizes. The bulk material parameters of silver are taken from [70].

![Figure 3.2](image-url)
The real part of the dielectric function is mainly unaffected by the size of
the nanoparticles. The imaginary part however seems to be heavily influenced
by the finite size correction. At decreasing particle sizes the imaginary part
of the relative permittivity increases, causing broadened and damped plasma
resonance.

Transmittance, reflectance and polarizing properties of the structure is
calculated using the total electric field below and above the structure. I
implemented the periodic layered Green’s tensor model described in section
2.5 in a python code. Technically the spectral representation of the Green’s
tensor is used as shown in subsection 2.5.3 and only the zeroth order is taken
into consideration because of the subwavelength period of the nanoparticle
lattice.

Using the developed model a single calculation runs for several seconds on
an Intel quad-core computer for a single wavelength depending on the actual
parameters. In contrast a rigorous method like FDTD or finite element takes
in the range of 1-10 minutes to calculate the response of a single geometry.
The computational cost of this model is much lower than that of the rigorous
methods like FDTD or finite elements, thus a suitable optimization can be
carried out in order to find the optimal parameters for the desired application.

3.3 Scattering cross section of single nanoparticles: spheres and ellipsoids

The first important step in the investigation of the response of the system
is the understanding of the single particle response since this serves as the
starting point in the many particle system. In this section I will calculate
the response of a single nanosphere and nanoellipsoid with different numerical
methods. The quantity investigated will be the scattering cross section, defined as

\[
C_{sca} = \frac{W_s}{I_i},
\]

where \( W_s \) is the power scattered by the particle, \( I_i \) is the intensity of the in-
cident plane wave [20]. In the case of a spherical particle the Mie-expansion
can be used to calculate the scattered field. Using the Mie-expansion the
scattering cross section can be expressed with the \( a_n \) and \( b_n \) scattering coef-
ficients:

\[
C_{sca} = \frac{2\pi}{k^2} \sum_n (2n + 1)(|a_n|^2 + |b_n|^2)
\]
If the particle is small compared to the wavelength the main characteristics of the scattering is of dipolar nature. The scattering cross section in this case can be expressed with the dipolar polarizability $\alpha$:

$$C_{sca} = \frac{k^4}{6\pi} |\alpha|^2$$  \hfill (3.5)

Figure 3.3 shows the scattering cross section of a silver sphere in a background of refractive index of 2.45 as a function of the sphere radius. The wavelength is 620 nm. The scattering cross section is calculated using different methods. The numerically exact solution can be obtained using the Mie-expansion using the whole expansion (denoted as mie-full). In case of small particles one can truncate the expansion to the electric dipole part, i.e. instead of the summation in eq. (3.4) only the $a_1$ term is used (denoted as mie). Furthermore the quasistatic approximation (qs) and the modified long wavelength approximation (MLWA) is used to calculate the dipolar polarizability and the scattering cross section using eq. (3.5).

Above 30 nm particle size the full expansion of the Mie solution and dipolar Mie solution start to differ meaning that the particle supports multipole radiating modes. The quasistatic approximation seems to be applicable only for particle radius of up to 10 nm, while we can see a much better agreement between the MLWA and the Mie solution.

The optical response of the nanoparticles having anisotropic shapes depends on the polarization of the incident light. To illustrate this I compare the scattering cross section of a single ellipsoidal nanoparticle calculated by
the quasistatic model with the MLWA and a finite element model. I chose the finite element method, because it is a rigorous solution of Maxwell's equation and even if it is not analytic it serves as a comparison to the approximate methods. The Comsol Multiphysics software was used for the finite element calculations with PML "boundary conditions" on all sides of the computational volume. Total field-scattered field formalism was used as the excitation inside the computational volume.

Figure 3.4 shows the extinction cross section of prolate spheroids excited with an electromagnetic plane wave of polarization parallel with (left column) and perpendicular to the major axis of the ellipsoid (right column). The direction vector of the incident wave is perpendicular to the major axis of the prolate spheroid. The figure shows the standard quasistatic model without MLWA correction, the MLWA correction and a finite element method for reference. The calculation is performed with particle sizes of 10 nm (top), 20 nm (middle) and 50 nm (bottom) major semi axis, and an axial ratio of 2. The background material is vacuum in this case.

The scattering cross section shows a resonant peak around 400 nm for polarization parallel with the major axis and around 350 nm for polarization parallel with the minor axis. The resonant wavelength depends on the size of the particle as well as on the polarization of the incident wave. The calculations show that the smaller the particle, the smaller the difference between the FEM, MLWA and quasistatic results. With increasing particle size the quasistatic scattering cross section spectrum changes only by a multiplication factor whereas the FEM and MLWA calculations are redshifted with respect to the quasistatic case. It can also be seen that the quasistatic calculation overshoots the magnitude of the peak at larger particle sizes. Using MLWA correction, the spectrum of the dipole scattering is closer to the FEM calculations both in maximum cross section and in spectral position even if there still remains some difference between the two.

### 3.4 Absorption of array of silver nanospheres.

#### Comparison of dipole model with finite element method

In order to validate the two-dimensional periodic-layered-Green’s tensor model developed in Section 3.2 a direct comparison with finite element method is presented for a two-dimensional periodic array of nanospheres. The finite element calculations are performed again in Comsol Multiphysics, in this case with periodic boundary conditions in the two lateral dimensions and PML
Figure 3.4: Extinction cross section at different sizes and polarizations. Left column: polarization parallel with the major axis, right column: polarization perpendicular to major axis. Length of major semiaxis: 10 nm (top), 20 nm (middle), 50 nm (bottom). Incident wave direction perpendicular to major axis.
boundary condition in the vertical one. Plane wave excitation is used. In Fig. 3.5 I show the absorbance of two-dimensional array of silver spheres with different interparticle distances as a function of wavelength. The radius of the particles is 5 nm, the background material is again vacuum.

![Figure 3.5: Absorption of silver nanosphere array. Particle radius is 5 nm, period of array is 12 nm (left) and 50 nm (right)](image)

The results were calculated using the finite element method using periodic boundary conditions as well as the periodic layered Green’s tensor method, the sphere polarizability is calculated using the MLWA approximation. The results agree very well for both small and large particle separations, a small difference appears at small separations between the finite element method and the dipolar approximation. There appears a small peak around 350 nm in the finite element results indicating the appearance of a higher order mode of the nanoparticles. However at larger separations this mode disappears.

### 3.5 Investigation of the polarized LED structure using silver nanoellipsoid array

The two-dimensional periodic-layered-Green’s tensor model developed in Section 3.2 is used to investigate the polarizing properties of two-dimensional array of silver prolate spheroids on an LED chip substrate. The investigated structure consists of a single interface with silver nanoparticles placed on the top, immersed in the encapsulation material. The structure is excited with an “unpolarized” and uncollimated dipole source placed into the sample structure depicted by a black star in Fig. 3.1. The refractive index of the substrate and that of the superstrate is 2.45 and 1.5 respectively. These
values correspond to an InGaN [71] LED structure. The ellipsoidal scatterers are placed into two-dimensional square lattice in order to eliminate the effect of lattice anisotropy. To avoid possible near field interaction with the interface the distance between the surface of the particles and the interface was set to 5 nm.

The numerical calculation was done with the periodic layered Green’s tensor method. The single particle polarizability is expressed with the MLWA approximation. This method is efficient enough to run parameter sweeps and to optimize the structure.

### 3.5.1 Dependence of the spectra on the parameters of the nanoparticle layer

I have investigated the effect of the parameters of the structure on four quantities: the degree of polarization (DOP), transmittance, reflectance and absorbance. The DOP is defined by

\[
DOP = \frac{I_\perp - I_\parallel}{I_\perp + I_\parallel},
\]

where \(I_\perp\) and \(I_\parallel\) are the intensities transmitted through an imaginary linear polarizer placed perpendicular to and parallel with the major axis of the ellipsoids respectively. The length of the semi-major axis (\(l_x\)) of the ellipsoid varies between 2.5 nm and 40 nm. The axial ratio (\(\text{AR} = l_x/l_y\)) varies between 1.1 and 5.0. The lattice constant (\(p\)) is chosen so that the separation between the particles is at least 1 nm and at most 15 nm.

Figure 3.6 shows the investigated four quantities for different particle sizes at fixed axial ratio value of 2.0 while the separation between the particles is kept at constant 5 nm.

The DOP curves show that the nanoellipsoid lattice achieves polarization selection as expected. With increasing particle size the maximum of the DOP increases and as we reach larger particle sizes the increase slows down and the maximum value approaches 50%. For the smallest investigated particle size the peak maximum is around 1.5%. The width of the peak first narrows with increasing particle size then broadens above 15 nm semi-major axis size. For small particle sizes the DOP curve is broad due to the reduced mean free path of the conducting electrons. With increasing particle sizes the size effect diminishes thus the width of the DOP peak decreases. However, above 20 nm the radiation damping becomes important and causes again broadening in the DOP peak. Moreover a redshift is visible in the DOP spectra which can be attributed to the dynamic depolarization at larger particle sizes.
We can see negative DOP values at wavelengths below 450 nm. As it can be seen in Fig. 3.4 the extinction cross sections have resonance peaks at different wavelengths for different incident polarizations. This indicates, that the nanoellipsoids support resonant modes even when the incident polarization is parallel with the minor axis. Thus the nanoellipsoid array resonates for both polarization component at different wavelengths.

The transmittance of the interface decreases as the particle size increases. Obviously waves with not allowed polarization will be either reflected or absorbed by the nanoparticles. It is more important however that the waves that are not transmitted should not be absorbed by the particles but rather reflected. Using a proper polarization recycler the reflected waves have another chance to escape from the device. It can be seen that with increasing size the absorbance decreases at the peak position and the reflectance in-
creases to about 80%.

In case of the smallest particle size (2.5 nm) the nanostructure has only a minor effect and the plotted data approaches the response of the bare chip-encapsulation interface. Thus without nanoparticles we would see about 18-20% transmittance and approximately 80% of reflectance.

Figure 3.7 shows the DOP and the absorbance for different lattice constants at $l_x = 20$ nm and $AR = 2.0$.

By decreasing the distance between the particles redshift and broadening of the DOP peak can be observed. This redshift and broadening is a sign of near field coupling between the particles [72]. By reducing the distance between the particles the absorbance of the nanoparticle array decreases. This leads to higher reflection due to the strong dipolar interaction of the nanoparticles.

I have investigated the effect of the axial ratio variation on the DOP and absorbance spectra. I have found that AR values between 1.5 and 2.5 yield the best DOP (about 50%) for the visible wavelengths where the axial ratio influences only the peak position.

### 3.5.2 Optimal parameters for single wavelength

In order to obtain the optimal parameters I have carried out another parameter sweep with finer resolution at a single wavelength of 620 nm. Figure 3.8 shows the axial ratio and period dependence of the degree of polarization as well as the absorbance. These quantities are calculated for particle sizes
Figure 3.8: Degree of polarization (left) and absorbance (right) maps as a function of period and axial ratio (AR). The major semi-axis was $l_x = 5$ nm (top), $25$ nm (middle) and $50$ nm (bottom). The wavelength is $620$ nm.
between 5 nm and 50 nm in steps of 5 nm. For illustration purposes I include only the results obtained for particle sizes of 5 nm, 25 nm and 50 nm in the dissertation.

The parameter sweep indicates that for each particle size the arrangement calculated with the smallest period (i.e. when the particles are the closest to each other) and with $\text{AR} = 2.2$ yields the highest DOP and lowest absorbance values. Figure 3.9 shows the DOP, absorbance, transmittance and reflectance curves as a function of the particle semi-major axis at 620 nm wavelength, $\text{AR} = 2.2$ and $p/l_x = 2.1$. The DOP values increase while the absorbance values decrease with increasing particle size. The curves approach a specific limit but do not reach the extreme values in this size regime. Note however that at the margin of the investigated interval the change of the curves is minor. The investigation of larger particle sizes would require more sophisticated calculation methods and can be the subject of future investigation.

![Figure 3.9: DOP, absorbance, transmittance and reflectance at 620 nm as function of particle semi-major axis. The axial ratio is equal to 2.2, the period is $p = 2.1l_x$.](image)

The best configuration yields about 50% DOP, and less than 15% absorbance. The transmittance of the interface is about 10% while the reflectance reaches 75%. In contrast the bare interface would yield a transmittance of about 20% and reflectance of about 80%. Comparing the transmittances only shows a factor of two ratio between the bare and nanostructured interface, however a polarization recycler would increase the extraction efficiency of the structure.
3.6 Thesis

I have proposed the application of an array of ellipsoidal nanoparticles as integrated polarizer to achieve inherently polarized LED technology. I have carried out an analysis and optimization of the polarizing nanoparticle array using a periodic Green’s tensor model. I have shown that particles in the size range of 100 nm are optimal to reach 50% degree of polarization and less than 15% absorbance for an uncollimated and unpolarized dipole source.

3.7 Related publications


Chapter 4

Polarizing properties of wire grid polarizers

4.1 Introduction

In this chapter the performance of wire grid polarizers (WGPs) is examined for the purpose of polarized LED applications. Two different constructions are investigated. The first construction is Grating-on-die (GD), which matches the structure described in [14], i.e. the WGP is placed on the plane surface of the LED chip and is embedded in the encapsulation (Fig. 4.1 (a)). The second construction is Grating-on-encapsulation (GE), a new construction in which the grating is placed on the hemispherical curved surface of the encapsulation (Fig. 4.1 (b)). The results of this chapter are used in the model assembly of a complete polarized LED in the next chapter. In Fig. 4.1 parallel black lines indicate a possible placement of the wire grid polarizer, hemispherical object represents the encapsulation.

Figure 4.1: (a) Schematic diagram of the polarized LED model with WGP placed on the LED die (GD). (b) Schematic design of the WGP on the encapsulation LED model (GE). Parallel lines indicate a possible placement of the grating lines, hemispherical object represents encapsulation.
In this chapter I use the well-established rigorous coupled wave analysis (RCWA) introduced in Section 2.6. I have implemented the one-dimensional version with oblique incidence in a Matlab code to compute transmittance, absorption and extinction ratio of WGPs with different grating parameters. In contrast to Fig. 4.1 the curved interfaces are taken into account locally as plane interfaces.

Note that the results of this chapter will be summarized in the thesis at the end of the next chapter, thus at the end of this chapter independent thesis cannot be found.

4.2 Modeling the wire grid polarizer

The optical properties of the WGP are investigated using the RCWA method \[51, 54\]. This method has been proven to be efficient in the simulation of periodic gratings of a wide range of materials and geometries. During the investigation I use the code developed by me in Matlab to calculate the results. I emphasize that the simulations in this chapter are performed in both cases for planar interfaces, thus the curved surface of the encapsulation is not taken into account. This assumption can be confirmed with the fact that the radius of curvature is always much larger than the dimensions of the grating.

The calculations are performed at a single wavelength of 620 nm taken from a typical AlGaInP red LED specification, but the same calculations can be implemented for any other wavelengths and the finite spectral width of the LED can also be taken into account. The samples consist of a superstrate, a single layer one-dimensional binary metallic grating and a substrate. The cross section of both configurations is the same, simple rectangular profile on flat surface (Fig. 2.5). The structure consists of a one-dimensional aluminum grating (refractive index: \( n_{AL} = 1.32 + 7.52i \)) between a substrate and superstrate which are semi-infinite. In case of GD the substrate is the die, the superstrate is the encapsulation material. In case of GE the substrate is the encapsulation and the superstrate is air. The refractive index of the die is approximated with \( n_{die} = 3.3 \) (according to AlGaInP LED chip) and the encapsulation refractive index is approximated with \( n_{enc} = 1.5 \).

Figure 4.2 shows an example of the characteristics of wire grid polarizers illuminated by polarizations parallel with and perpendicular to the grating lines respectively. Each single subplot represents a single period of the grating. The series visualize the electric field at consecutive time frames through a whole time period. The upper image shows the case when the polarization of the incident electric field is parallel with the grating lines. In this
4.3 Parameter dependence at normal incidence

The extinction ratio, average transmittance and the average absorption is studied as the function of grating period, height and line/period ratio. Preliminary investigation showed that the parameter dependence of the GE and GD geometries is very similar, however the GE geometry yields much better performance (see Section 4.4). Therefore in this section the "grating-on-encapsulation" geometry is investigated in details.

In accordance to previous publications [17, 18, 16, 19] good performances (transmittance above 90%, extinction ratio in the range of 200 [16]) are available at grating period below 150 nm, line/period ratio about 0.4 and grating height of the range 100 - 200 nm. It is clear that in the design of the WGP there is a trade-off between extinction ratio and the average transmission.

In order to find the most appropriate polarizing structure a parameter sweep was carried out. The variables were the period of the structure (between 100 and 200 nm), the line/period ratio (between 0.4 and 0.6) and the
height of the grating (between 100 nm and 200 nm). The refractive index of the substrate was 1.5 and that of the superstrate was 1.0. The performance was investigated at normal incidence. The investigated quantities are the extinction ratio and the average absorption. The extinction ratio is defined by

$$ER = \frac{\langle I_\perp \rangle}{\langle I_\parallel \rangle},$$

where $I$ denotes the transmitted intensities, the subscript denotes the parallel or perpendicular polarization component at the detector corresponding to the grating. $\langle \cdot \rangle$ denotes the averaging over the incident polarization states. The average absorption is defined by the absorbance averaged over the incident polarization states. Figure 4.3 to Fig. 4.5 show the extinction ratio and the average absorption for line/period ratio of 0.2, 0.4 and 0.6 respectively. The height of the grating is denoted in the legend of the figures.

For all configurations the polarizing effect of the wire grid polarizer is visible. In the investigated parameter range the average extinction ratio varies between 10 and $10^8$, meaning that even at the worst case the portion of the perpendicular polarization is 10 times larger than that of the parallel polarization.

The minimal absorption in the investigated parameter range is as low as 10.5%, and the maximum reaches 18%.

The extinction ratio decreases monotonically for all heights and line/period ratios with increasing period, whereas the absorption slightly decreases or remains constant. With increasing height of the grating both the extinction ratio and the absorption increases. Increasing the line/period ratio increases both the extinction ratio and the absorption.

The optimal parameters depend on the trade off between the highest extinction ratio and the lowest absorption. Another important aspect is the manufacturing limitations of the structure.

For further investigations I have chosen the height and period to be 150 nm, and the line/period ratio 0.4. These parameters provide low enough absorption and high enough extinction ratio of the investigated geometry.

### 4.4 Comparison of Grating-on-die (GD) and Grating-on-encapsulation (GE)

The performance of the polarizers are investigated at general angle of incidence. Figure 4.6(a) shows the extinction ratio of the gratings defined by
4.4. GRATING-ON-DIE VS GRATING-ON-ENCAPSULATION

Figure 4.3: Extinction ratio and average absorption for aluminum grating. The line/period ratio is 0.2, height of the lines is shown in the legend.

Figure 4.4: Extinction ratio and average absorption for aluminum grating. The line/period ratio is 0.4, height of the lines is shown in the legend.

Figure 4.5: Extinction ratio and average absorption for aluminum grating. The line/period ratio is 0.6, height of the lines is shown in the legend.
the ratio of the power that can be observed by an external analyzer aligned in the x-direction and the power with the external analyzer aligned in the y-direction. Figure 4.6(b) displays the absorption of an unpolarized incident wave by the gratings. In spherical coordinates, the curves depend on the polar angle and are drawn at a few azimuth angles.

Figure 4.6: (a) Extinction ratio and (b) average absorption for unpolarized incident wave. The WGP is placed onto the encapsulation (Grating on encapsulation: line + symbol) or onto the LED die (Grating on die: solid line).

It can be seen from the figures, that GE achieves better performances than GD both in extinction ratio and in extraction efficiency. On the one hand, the extinction ratio of GE is an order of magnitude higher than that of GD for each azimuth angle [Fig. 3(a)]. On the other hand, the average absorption of GE – when an unpolarized wave hits the surface – is much lower than that of GD for each azimuth angle. Besides that, there is another aspect in which GE is superior. From the geometry of the LED source it is clear, that the wave coming from the chip and hitting the encapsulation surface falls nearly at normal incidence onto the surface. On the contrary inside the chip, the waves fall on the interface of the chip and the encapsulation in polar angles uniformly distributed. Consequently a large amount of light is trapped inside the LED die by total internal reflection. Thus one can conclude, that if these conditions hold, it is reasonable to put the WGP on the encapsulation rather than on the chip surface. In this case the large absorption arising from the larger polar angle region in GD can be avoided and the high extinction ratio of GE at near normal incidence is insured.
4.5 Comparison with silver nanoellipsoid arrays

In the previous chapter I showed that the silver nanoellipsoid array structure can produce polarized output from dipolar emitters. Direct comparison of the nanoellipsoid array with the wire grid polarizer structure can be done only if the same quantities are calculated for both structures. Therefore I have calculated the degree of polarization, average transmittance, average reflectance and average absorbance as defined in the previous section for the wire grid polarizer too. The optimal wire grid structure is used for the comparison (period = 150 nm, line/period = 0.4, height = 150 nm).

The degree of polarization is 65.8%, the average transmittance is 9.8%, the average reflectance is 70.5% and the average absorption is 19.7% for the wire grid polarizer structure. In comparison the DOP was less than 50%, the average transmittance was 10%, the average reflectance was about 75% and the average absorbance was 15% for the best nanoellipsoid array.

The wire grid polarizer yields much better DOP than the nanoellipsoid array, but at the same time the average absorption is by 5 percentage point higher. The average transmittance is almost the same for both cases. By summing up these results we can conclude, that the wire grid polarizer is more suited for the polarized LED application than the nanoellipsoid array.
Chapter 5

Complete polarized LED model

5.1 Introduction

In the previous two chapters I have investigated two possible polarizing structures which are both capable of transforming the unpolarized output of the LED chip into polarized light emission. Both of them provided good enough optical responses in order to use them in polarized LED application, however the wire grid polarizer seemed to achieve slightly better. To bring the idea one step further towards the realization of such a device in this section I build up a complete optical model of an AlGaInP LED device using Zemax OpticStudio, a commercial ray tracing software in nonsequential mode. During this work the idea of Wheatley et al. [14] is reviewed and modified in order to achieve better performance.

The polarizing structure consists of a wire grid polarizer placed either on the LED chip surface or onto the encapsulation. The response of the wire grid polarizer is calculated by the RCWA module introduced in the previous section and is handled by a custom implemented dynamic linked library (DLL) which is linked to the main ray tracing program.

The polarized LED device would work optimally if the reflected waves could go through a polarization rotator upon reflection by the WGP. However in this study such structure was not used, it can be a subject of future research. Even in this case the reflected waves could be perturbed by the geometric structure of the device, thus a slight polarization recycling property is awaited.

In this chapter I compare two different realizations of a polarized LED. The first corresponds to the Grating-on-encapsulation (GE), the second to the Grating-on-die (GD) defined in the previous chapter. Moreover an unpolarized LED with and without external polarizer is modeled for comparison.
5.2 The complete polarized LED device model

In this research a high efficiency, transparent substrate AlGaInP LED \[73, 74\] is investigated. The model consists of an LED chip (assumed to be non-absorbing), an encapsulation (spherical, with $n = 1.5$ refractive index) and a mirror at the backside (Fig. 4.1 (a)). The chip area is $500 \times 500 \, \mu m^2$, the chip height is $250 \, \mu m$. The refractive index of the chip is assumed to be 3.3, and absorption is neglected for this wavelength. Spherical encapsulation is assumed with a diameter of 5 mm.

I investigate three different theoretical LED configurations. The first one is an unpolarized LED + external polarizer combination. For the external polarizer I use a WGP with the same parameters for better comparison. The second one is a standard LED with a WGP placed onto all sides of the LED chip (Fig. 4.1 (a)). The third one is also a standard LED with a WGP placed onto the curved encapsulation surface. In this last case the polarizing WGP is designed similarly to the circles of latitude on the globe. Figure 4.1 (b) shows the schematic diagram of such an encapsulation. For both configurations the grating period as well as the grating height are 150 nm and the line/period ratio is 0.4.

In the case of the spherical surface the WGP is approximated with wire grids on a locally plane surface. The grating has a constant period and an appropriate direction. The distortion of the parallel lines due to the spherical surface as well as the variation of the line spacing are not included in the model. I use the locally flat surface approximation, because the radius of curvature of the sphere is much larger than the size (period, height) of the grating. Practically the grating can be imagined on the whole curved surface as consisting of multiple segments, where the size of one segment is much smaller than the area of the sphere, but much larger than the grating period. Similar modeling method is used by Kim et al. \[75\]. The direction of the grating lines on the curved surface can also be optimized, to reach higher extinction ratio as well as higher efficiency. In the optimized setup the direction of the lines is adjusted so that the polarization of the outcoupled wave lies in the globally defined $xz$-plane. In this case higher extinction ratio is expected.

The LED device is modeled using the Zemax ray tracing software in non-sequential mode. With this method arbitrary LED configurations can be designed with appropriate geometry and parameter set. To evaluate optical performance a Monte-Carlo simulation is performed, that is twenty thousand rays are launched from the active region of the LED die. The initial direction, polarization and phase of the rays are randomly generated. Geometrical optical ray tracing takes place until the ray hits the WGP surface. Here with
the help of an external dynamic-link library routine (DLL) written in C, the ray is transmitted, reflected or absorbed with the probability equal to the diffraction efficiency of the transmission, reflection or absorption respectively. After the surface, geometrical optical ray tracing continues until the ray escapes from the system, gets absorbed or hits again the WGP. On the detector surface the incoming rays are incoherently summed for each pixel. Practically as shown in Fig. 5.1 (a) the RCWA program is used to generate tabular diffraction data for a single WGP as a function of incident angle and polarization. This data file is then loaded by the dll for further use. The dll is communicating with the main ray tracing program, loads the current properties of the incoming ray and calculates the direction and polarization of the outgoing ray. These ray parameters are then given back to the main program.

Figure 5.1: (a) Schematics of the combination of the geometrical optical model with the diffraction optical model. (b) Schematic drawing of the simulation setup. D1 and D2 denotes the detectors, which detect the uncollimated and collimated waves respectively. L denotes a theoretical “perfect” collimating lens, and (P) denotes an optional polarizer for the LED + external polarizer configuration.

The polarization properties of the LED device can be measured properly with an external polarizer only if its radiation is collimated. In the uncollimated case most of the rays do not hit the external polarizer at normal incidence which makes it difficult to evaluate the results. However the native radiation pattern of an LED is inherently uncollimated. To overcome this difficulty I use a virtual lens with the LED chip placed in the focal point of the lens to collimate the LED radiation. For comparison I also list the uncollimated results. In the uncollimated case a virtual polarization sensitive detector is placed at 10 cm far from the source. In the collimated case behind the collimating lens there is another virtual detector. The simulation setup can be seen in Fig. 5.1 (b).
5.3 Comparison of the modeled LED sources

For a detailed examination of the polarized LED using WGPs, various structures are modeled and compared. Table 5.1 shows the results of the previously defined configurations:

- a standard unpolarized LED for comparison of the total extraction efficiency
- an unpolarized LED + external polarizer configuration
- a setup with a WGP placed onto all sides of the chip
- a configuration, where the WGP is placed onto the spherical encapsulation
- an improved WGP on the encapsulation setup - where the direction of the grating lines are optimized - is also listed in the table.

<table>
<thead>
<tr>
<th>Configuration</th>
<th>Extraction efficiency</th>
<th>Loss relative to the unpolarized LED</th>
<th>Extinction ratio</th>
<th>Extinction ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unpolarized LED</td>
<td>23%</td>
<td>-</td>
<td>0.97</td>
<td>0.96</td>
</tr>
<tr>
<td>LED + external polarizer</td>
<td>9.6%</td>
<td>58.3%</td>
<td>-</td>
<td>2063.27</td>
</tr>
<tr>
<td>WGP on all sides of the chip</td>
<td>5.6%</td>
<td>75.7%</td>
<td>2.04</td>
<td>5.38</td>
</tr>
<tr>
<td>WGP on the encapsulation</td>
<td>12.9%</td>
<td>43.9%</td>
<td>2.72</td>
<td>5.01</td>
</tr>
<tr>
<td>Optimized WGP on encapsulation</td>
<td>12.6%</td>
<td>45.2%</td>
<td>2.37</td>
<td>76.86</td>
</tr>
</tbody>
</table>

Table 5.1: Performances of the investigated LED models

Standard unpolarized LED

The extraction efficiency of the unpolarized LED is 23%. This value is typical for an LED without any extraction enhancing structure. Figure 5.2 shows the detector images of the x-polarization component (left), y-polarization
5.3. COMPARISON OF THE MODELED LED SOURCES

Figure 5.2: $x$ polarization component (left), $y$ polarization component (middle) and total intensity (right) of the uncollimated unpolarized LED source.

Figure 5.3: $x$ polarization component (left), $y$ polarization component (middle) and total intensity (right) of the collimated unpolarized LED source with external polarizer.

component (middle) and the unpolarized intensity (right) of the unpolarized and uncollimated LED source. The total intensity values of the $x$ and $y$ component are the same, only the distribution is slightly different. This shows qualitatively that the radiation is unpolarized.

In the LED + external polarizer case, with the applied external wire grid polarizer the extinction ratio of the collimated case is very high, about 2000. However the total extraction efficiency is highly reduced compared to the unpolarized LED source, i.e. more than half of the extracted light is lost (58.3% loss compared to the unpolarized case). Figure 5.3 shows the detector images of the collimated unpolarized LED source with an external polarizer applied. It can be seen, that the $y$ component of the radiation is practically completely absent.

**WGP on the chip surface**

The WGP on the chip surface setup has the worst results both in extraction efficiency and in extinction ratio. In accordance with Chapter 4, the high loss
CHAPTER 5. COMPLETE POLARIZED LED MODEL

Figure 5.4: $x$ polarization component (left), $y$ polarization component (middle) and total intensity (right) of the uncollimated grating-on-die geometry

Figure 5.5: $x$ polarization component (left), $y$ polarization component (middle) and total intensity (right) of the collimated grating-on-die geometry

on the WGP at the chip-encapsulation interface causes reduced extraction efficiency. The extraction efficiency is 5.6%, i.e. about 75% loss is realized compared to the extraction efficiency of the unpolarized LED. Even if this setup shows worse performance than that of the unpolarized LED + external polarizer setup, it shows linearly polarized extraction with an integrated solution. Figure 5.4 and 5.5 show the uncollimated and collimated detector images of the grating-on-die geometry respectively. These figures show again that the $y$ component is reduced compared to the $x$ component but the extinction is not yet perfect.

WGP on the encapsulation

Two configurations of the WGP on the encapsulation setup were also investigated. The grating lines of the first one are similar to Fig. 4.1 (b). The direction of the grating lines of the second one are optimized in order to reach higher extinction ratio in the collimated case. The extraction efficiency in both cases is about 13% (43.9% and 45.2% explicit loss), definitely higher than the extraction efficiency of the LED + external polarizer combination.
5.3. COMPARISON OF THE MODELED LED SOURCES

Figure 5.6: $x$ polarization component (left), $y$ polarization component (middle) and total intensity (right) of the uncollimated optimized grating-on-encapsulation geometry

Figure 5.7: $x$ polarization component (left), $y$ polarization component (middle) and total intensity (right) of the collimated optimized grating-on-encapsulation geometry

This fact shows that these configurations have a polarization recycling property, i.e. a part of the light reflected by the WGP undergoes reflection and polarization change and thus can escape again with proper polarization state. Note that although explicit polarization recycler grating is not included in the model, the polarization recycling property is present because the inner structure of the LED indicates phase change of the reflected light.

Figure 5.6 and 5.7 show the detector images of the uncollimated and collimated optimized grating-on-encapsulation structure respectively. In the uncollimated case the $y$ polarization component of the radiation is still present. This is the consequence of the uncollimated direction distribution of the radiation. In contrast in the collimated case the detector images also show that the $y$ component is extincted by the polarizing structure on the encapsulation.

The extinction ratio values measured in the uncollimated case, compare favorably with previous publications [12, 13]. The relatively low extinction ratio can be explained by the fact that the intensity of the light source before
collimation has a wide angle distribution. In this case the z-component of the polarization can be relatively high, which contributes to both measured polarization states and reduces the extinction ratio. In the collimated case, the angle distribution of the light source is narrow, thus the z-component of the polarization is small. The presented WGP on the encapsulation setups show strong polarized properties in the collimated case. Moreover, the extinction ratio of the optimized case is over 70. This seems much lower than the extinction ratio of the LED + external polarizer configuration, however this value is also well suited to most practical applications. On the other hand, the significantly higher extraction efficiency of this setup is a remarkable advantage in contrast to the LED + external polarizer configuration.

To illustrate that the presented solution is compatible with modern light extracting structures, the optimized WGP on the encapsulation setup is investigated with the use of a pyramidal structure placed onto the chip surface. This technique is widely used to enhance the light extraction efficiency of high power LED sources [76]. The simulations show that the total extraction efficiency is over 30% and the (collimated) extinction ratio of this setup is about 100. This result shows that the proposed setup is compatible with high extraction efficiency power LEDs at slightly improved extinction ratio. Although the feasibility of the presented polarized LED remains a challenge because of the non-planar WGP, recent studies show that fabrication of gratings with similar parameters on curved surface is possible for example with nanoinprint lithography techniques [77, 78].

5.4 Thesis

I have proved the feasibility of polarized light emitting diodes with integrated wire grid polarizers, using a model combining geometrical and physical optics. The whole LED structure was modeled with ray tracing, the effect of the wire grid polarizer was taken into account locally using rigorous coupled wave analysis. I have shown that the wire grid polarizer in this setup yields better performance than that of the nanoellipsoid array. I have shown that it is advantageous to place the WGP onto the encapsulation. In this case the efficiency of the device (12.6%) was significantly higher than that of an unpolarized LED and external polarizer combination (9.6%) while the extinction ratio was appropriate for most applications (76.86).
5.5 Related publications


Chapter 6

Characterization of two dimensional metallic nanoparticle and void films derived from a colloidal template layer

6.1 Introduction

In this chapter I investigate the plasmonic properties of nanodome and nanovoid arrays prepared by Langmuir-Blodgett template layer. I use the experimental samples prepared by the group of András Deák (MTA TTK MFA Photonics Group) to investigate the reflectance spectra experimentally of such plasmonic interfaces.

An electromagnetic model was developed to explain the measured reflectance spectra. Using this model it is possible to derive physical quantities which are difficult to measure directly e.g. the electromagnetic near fields.

An excellent agreement was found between the experimental data and electromagnetic simulation results when the inherent domain structure of the self-assembled template monolayer and hence the resulting final structure was taken into account.

6.2 Sample preparation and measurements

The samples investigated in this chapter were provided by the group of András Deák (MTA TTK MFA Photonics Group). In this section I shortly review the sample preparation and measurement process.
The sample preparation consists of three major steps, involving the template particle preparation, their monolayer formation on water/air interface and the final structure preparation by heat treatment and metal deposition. An overview of the preparation procedure is provided in Fig. 6.1. For all experiments ultrapure water with a resistivity of 18.2 MΩ cm was used.

First, the template particles have been synthesized according to Stöber’s method with some modification according to the literature [79], using Ludox®-S40 silica nanoparticles (Sigma-Aldrich) as seeds for the particle growth. This seeded-growth approach improves the size-distribution of the particles compared to the original Stöber method, which is critical for the directed self-assembly process. Second, a monolayer of the template particles were prepared in a Langmuir film balance and transferred onto a substrate by the Langmuir-Blodgett (LB) method as reported by us earlier [80]. The monolayers were transferred onto Si-wafers, coated with a 300nm thick Shipley S1805 photoresist layer in a class 10-10000 clean room facility. As a last step, the monolayer deposited on the substrate were subjected to heat treatment at 120°C for 60 seconds. During the annealing process, the monolayer submerged into the polymer film to an extent depending on the temperature and duration of the heat treatment process. After completing the annealing step, the template particles were optionally removed from the polymer film by soaking the structure in 2% HF-solution for 20 seconds before e-beam evaporation of 140 nm gold at a rate of 2Å/s in a ATC ORION 8-E UHV e-beam evaporation system.

The prepared samples showed really exotic and colorful reflectance spectra as viewing by naked eyes. The photographs at different incident angles are shown in Fig. 6.2.

The as-synthesized particles were characterized by dynamic light scattering (DLS) (Malvern Nano ZS) to verify their narrow size distribution. For the structural characterization of the samples scanning electron microscopy
6.2. SAMPLE PREPARATION AND MEASUREMENTS

Figure 6.2: Photographs of the plasmonic structures. The color of the samples changes with the observation angle and it is the result of the plasmonic properties of the structure.

(LEO 1540 XB, Philips XL30 FEG) and atomic force microscopy (Bruker Dimension Icon) has been used.

The mean particle size is 500 nm. Figure 6.3 shows the characteristic SEM images of the two samples. Both for the positive (dome) and negative (void) structures, the top view images (Fig. 6.3 (a, b)) clearly indicate a domain-like structure of the film after the gold deposition. The typical size of the domains is around 10-20 µm. The domain structure of the film is an inherent property of the template monolayer and is naturally developed for self-assembled mono and multilayers in general. The cross-sectional view of the samples (Fig. 6.3 (c, d)) clearly evidence the partial submerging of the template particles into the polymer film, as well as the dome and void structures after gold deposition. For both types of samples the deposited gold film is continuous.

The angle-dependent optical reflectance spectra of the samples was measured using a Perkin Elmer Lambda 1050 spectrophotometer with Universal Reflectance Accessory (URA) in the wavelength range of 400-800 nm and angular range from 8 to 65 degrees in 13 points.
Figure 6.3: SEM top (a-b) and cross-section (c-d) images of the final dome (a,c) and void (b,d) structures after gold deposition. The insets show the schematic model setup used during the simulation of the respective samples.

6.3 Electromagnetic simulations

Finite difference time domain (FDTD) calculations were used to evaluate the measured reflectance spectra of the different samples using the Lumerical FDTD Solution software [81]. This method ensures that the complex geometry is described with all necessary details, still the computational demands of the method is low enough to be run on a personal computer with 64 GBytes of memory and 6 cores.

The optical model of both the nanovoid and the dome array consists of a semi-infinite Si substrate (refractive index from [82]), a photoresist layer of thickness of 300nm (refractive index from [83]) and a gold layer of thickness of 140nm (refractive index from [70]). The refractive indices are approximated with the Drude-Lorentz susceptibility relations (eq. (2.69)) and are automatically fitted by the program.

Figure 6.4 shows the cross section (left) and the top view (middle) of the simulated nanovoid structure as well as the parameters of the incident wavevector (right). In the simulation setup, both the photoresist and the gold layer were patterned with a hexagonal lattice of spherical voids/domes with vertical position of the spherical void/dome of 175 nm. The lattice period was 570 nm and the sphere diameter was 500nm, these values were obtained.
from the AFM and SEM measurements. Bloch boundary conditions were applied in lateral directions and perfectly matched layer (PML) [62] in the vertical one. The structure was excited with a plane wave having variable polar and azimuth angle of incidence with respect to the lattice defined by the simulated unit cell, using a wavelength range between 400nm and 800nm. The polar angle (θ) varied between 0 and 65 degree, while the azimuth angle (φ) was set between 0 and 30 degree in 5 degree steps. Calculation was only necessary up to 30 degree because of lattice symmetry.

The advantage of the time domain calculation is that the whole wavelength range can be calculated at a single simulation using the Fourier transform of the time signal. Using Bloch boundary conditions at oblique incidence requires the set up of the lateral component of the incident wave vector as a constant parameter instead of the polar angle. Hence this leads to a wavelength dependent polar angle for each calculation. Finally wavelength and angle dependent spectra can be produced in the postprocessing. Due to time limitations I have calculated the reflectance spectra only up to a certain maximum value of the Bloch vector which results in incomplete reflectance images at higher polar angles (e.g. see Fig. 6.5).

Calculating the response of a single structure for all angles, and wavelengths and both transverse electric (TE) and magnetic (TM) polarizations took about a week on a PC with 6 cores and 64 GB of RAM. Altogether $40 \times 7 \times 2 = 560$ different setup was run, taking in average 18 minutes per single run.
6.4 Results

I have first calculated the specular reflectance of the structures at discrete azimuth angles of the incident wave. As an example Fig. 6.5 (a-d) shows the reflectance spectra for the TM polarization of the void structure at azimuth angles of 0, 10, 20 and 30 degree.

A relatively strong absorption line is visible in all spectra at the same position starting at around 600 nm and can be identified as the Bragg plasmon mode of the system [36]. The apparent ‘patchiness’ of the data at this narrow reflection minimum band originates from the limited angular resolution of the simulation and is a direct consequence of data interpolation.

The appearance of sharp lines and edges in the calculated spectra correspond actually to a dispersion relation where a plasmon mode can couple into the structure causing absorption. These absorption lines appear to be quite close to the lines where the number of propagating orders changes, i.e. a propagating order becomes evanescent. However, a more precise description of the dispersion relation can be derived using eq. (2.5). Note that this equation is valid asymptotically if the interface is plane. In our case this equation is a good approximation since the height of the domes or voids is not too large compared to the diameter. For comparison I show the number of propagating orders as well as the dispersion relation calculated by eq. (2.5) in Fig. 6.6. Indeed we can observe the accordance between the dispersion curves and the calculated reflectance spectra. At lower wavelengths however the reflectance of the gold layer decreases thus below 550 nm the dispersion curves are not easily observable in the calculated spectrum.

Moreover a weak signal of localized modes is present around 650 nm, where independently of the azimuth angle a slight dip can be observed in the reflectance spectra. Similarly around 600 nm we can observe a dip in the reflectance spectra, but only at small polar angles. More spectacular localized modes could be observed if the depth/diameter ratio of the voids was larger.

The local electric field corresponding to particular azimuth angle, polar angle and wavelength can be calculated using field monitors in the model. As an example Figure 6.7 shows the magnitude of the electric field in top view right above the gold layer and in cross section view for the void geometry. The top row corresponds to a propagating plasmon mode at polar angle of 17 degree, azimuth angle of 30 degree and wavelength of 680 degree. The bottom row shows an example of a particle plasmon at polar angle of 7 degree, azimuth angle of 5 degree and wavelength of 550 nm. These field plots show the local field enhancement properties of surface plasmons.

The predicted angle-dispersive plasmon band can be also observed in the
6.4. RESULTS

Figure 6.5: Simulated specular reflectance spectra of the void structure for TM polarization at azimuth angles of 0 (a), 10 (b), 20 (c) and 30 degree (d).

Figure 6.6: Dispersion relation of the two dimensional hexagonal lattice with period of 570 nm. The azimuth angle is 0 degree (a), 10 degree (b), 20 degree (c) and 30 degree (d). Different colors mean different number of propagating orders.
optical reflection measurements (Fig. 6.8 and 6.9 top row), which is consistent with the coupling mechanism of the incident light to the propagating plasmon mode by the regularly spaced scatterer centers in a hexagonal lattice [34]. The domain structure of the film has important implication on the optical behavior of the samples. Due to the different orientation of the domains with respect to the plane of incidence and the macroscopic size of the light beam, all lattice directions are represented randomly in the reflected beam, since reflected electromagnetic waves arising from each domain are summed as they reach the detector. Since the domain size is larger than the wavelength and the propagating plasmon decay length (which is well below 20 microns for a smooth gold film in this wavelength range [84]) the reflection
for each domain can be accounted for independently. Hence despite the lateral ‘multicrystalline’ nature of the sample surface, the Bragg mode is clearly developed in such a macroscopic ensemble measurement.

In order to reproduce the experimental results the model was developed to take into account all possible lattice directions in the calculated reflectance spectra. Due to the uncorrelated domain position, the summation of calculated spectra is done incoherently, leading to an average of the reflected intensity over the lattice orientations (azimuth angles). This operation allows a direct comparison with the experimental spectra. After averaging, an excellent agreement can be seen between the simulated and the measured spectra. The lower row in Fig. 6.8 and Fig. 6.9 shows the simulated reflectance spectra for the nanovoid and the nanodome structures for both polarizations. The good agreement indicates a very good quality of the structures (i.e. high local order within the domains), and implies indirectly, that
the optical response of each domain is well described by the theoretically calculated spectrum at a particular lattice orientation. The absorption lines in Fig. 6.5 at given incident angles correspond to plasmon modes, suggesting strong near fields at the structure surface. This behavior makes the structure useful for potential applications, where strong near fields and local plasmonic modes are advantageous. Sample field maps can be seen in Fig. 6.7.

The measured reflectance values are about 10 percentage point lower than the calculated ones. This difference can be attributed to losses associated with scattering from lattice defects and domain boundaries. It is important to note, that the reflectance spectra for the void and dome structure are very similar for TE and TM polarizations respectively. This indicates that in these relatively shallow structures the spectrum depends mainly on the lattice geometry, not on the shape of the particular scatterer.
6.5 Thesis

I have explained the reflectance spectra of two-dimensional metallic nanovoid and nanodome arrays prepared by Langmuir-Blodgett template layer using finite difference time domain method. The inherent domain-like structure of the samples is modeled by incoherent averaging over all possible lattice directions. The calculated and measured reflectance spectra are in very good agreement for both nanodome and nanovoid arrays and for both polarizations. The developed model enables us to have an insight into the plasmonic properties of the particular structures.

6.6 Related publications

Chapter 7

Summary and conclusions

In this work I have investigated the interaction of light with nanostructured interfaces composed of dielectric, semiconductor and metallic materials to provide a basis for new functional photonic devices. The results provided here can be applied for the design of light emitting diodes, solar cells, or different plasmonic applications.

In Chapter 3 I have investigated the optical properties of silver nanoparticles both in single particle and in periodic array configurations. I have compared the developed dipole model with the Mie-expansion and with a numerically exact finite element model to show the boundaries of the validity of the model. The model was used to investigate the polarizing properties of silver nanoellipsoid arrays on a conventional LED surface. The model included the effect of layered background as well as the dipolar interaction of nanoparticles. The method was extended to larger particle sizes with the use of correction factors for dynamic depolarization and radiation damping in the particle polarizability. The finite size effect of the particles was taken into account in the dielectric function of the silver.

The investigated curves show strong size, shape, and lattice period dependence. I have demonstrated that optimization of the structure is possible with the available method at a single wavelength. The best results were obtained at particle major axis of 100 nm, axial ratio of 2.2 and period of 210 nm. The results indicate that particle sizes above 100 nm can be subject of future investigation in order to achieve even higher DOP values and lower absorbance.

For the best configurations the DOP values are near 50%, while the absorbance is below 15% for the unpolarized dipole emitter. The transmittance and reflectance is about 10% and 75% respectively. Using a proper polarization recycler to recover the reflected waves, the extracted power can largely
surpass the LED + external polarizer configuration.

In Chapter 4 I have investigated the performance of a wire grid polarizer for the polarized LED application. The rigorous coupled wave analysis results showed that it is more advantageous to place the wire grid polarizer onto the encapsulation rather than onto the LED die. In this case the absorption of the grating is smaller and the extinction ratio is larger for all incident angles. A parameter sweep was used over the period, linewidth and height parameters in order to understand the optical properties of the wire grid polarizer and to choose the best performing parameter set. The optimal parameters for normal incidence yield extinction ratio in the order of $10^5$ and absorption of about 14.4%. The chosen structure was compared with the nanoellipsoid array using the definitions of the previous chapter. The wire grid polarizer yields much better degree of polarization and a bit higher absorption than the optimal nanoellipsoid structure. Since the transmittance of the two structures is about the same the wire grid polarizer structure was chosen for the complete polarized LED model.

In Chapter 5 the combination of light emitting diodes and WGP structures were investigated in order to produce polarized light output. A complete AlGaInP LED structure was built up in a Monte-Carlo ray tracing software to determine the optical properties of this configuration. The model includes the main parts of a conventional LED structure as well as optional light extracting structure and the wire grid polarizer. In accordance with the previous chapter the ray tracing analysis shows that it is advantageous to place the WGP on the encapsulation surface rather than on the die. In conclusion the average extinction ratio was found to be 2.37 in the uncollimated case and 76.86 in the collimated case, while the light extraction was significantly higher than that of the LED + external polarizer combination. Moreover with additional light extracting structure on the chip surface, over 30% extraction efficiency and an extinction ratio of about 100 was estimated. The presented results were not optimized for any special application, but the proposed model can be used for the design of polarized LED applications requiring high extinction ratio, power and specific angular distribution.

In Chapter 6 I have investigated the plasmonic properties of nanodome and nanovoid arrays. Experimental samples were fabricated based on a short annealing of a polymer layer supported particle monolayer template. As a result of the annealing process the template particles partially submerge into the polymer layer and after an optional removal of the template the structured surface can be coated with a uniform gold film using electron beam evaporation. AFM and SEM measurements show that the prepared struc-
tures have a domain-like texture with a typical feature size of 10-20 µm and high local order of the two-dimensional hexagonal lattice within the domains. The measured specular reflectance spectra show a strong absorption band for TM polarizations and a weaker one for TE polarization, which can be attributed to a propagating plasmon mode.

The developed electromagnetic model shows very good agreement with the measured reflectance spectra for all configurations provided incoherent averaging of the theoretical data is performed over all lattice orientations. The good agreement indicates indirectly that the electromagnetic response of locally each domain supports the theoretically predicted plasmonic modes with the corresponding near fields.

Finally I would like to highlight some applications based on the models developed during this work. The common property of these applications is that all of them are joined to projects with industrial partners. The finite difference time domain method was used in two projects. First the extraction efficiency of organic light emitting diodes (OLEDs) was enhanced using aggregated TiO$_2$ nanoparticles of size between 100 and 300 nm. In this case the FDTD method was the only one that could handle the large number of particles in this size regime. Second, the efficiency of thin film solar cells was improved using rough surfaces. Here again the size of the computational volume made it necessary to use this method.

The periodic layered Green’s tensor model was also applied in an industrial project. The peculiar transmittance of ultrathin nano-porous silver film in complex multi-layered structure was investigated using this method, that is fast enough for the optimization procedure, and still able to describe the investigated structure.

These results have not been published due to confidentiality reasons, therefore they are not included among the results of this dissertation.
Chapter 8

My publications

8.1 Related publications


8.2 Other publications


8.2. OTHER PUBLICATIONS

Place and date of conference: San Diego, United States of America, 10/08/2008-14/08/2008. p. 111.

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