# Topology Optimization of Nearfield Enhancing Nanostructures

PART A Ph.d. Thesis

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## Introduction

Global climate changes motivates the transition from conventional fossil fuels towards renewable and  $CO_2$  neutral energy sources. Within the European Union (EU) the quantity of energy produced from renewable sources has increased by approximately 73% from 2004 to 2014. In 2014 alone 25% of EUs total energy production came from renewable energy sources mainly produced via biomass and waste,  $(63\%)^i$ , hydro power (17%), wind (11%) and solar (photovoltaic and thermal) 6% [1]. However, from 2010 to 2014 EU energy production from solar alone has increased fourfold, a trend that is also evident on a global scale where solar and wind currently are the fastest growing sources of electricity [2].

Between 2008 and 2015 the cost of solar photovoltaic has decreased by roughly 80% mainly driven by reductions in the cost of (poly)silicon and optimisation of large scale manufacturing costs as well as improvements in the overall module efficiency when converting sunlight into electricity [3]. This trend is expected to continue as long as advancements in production and material efficiency are maintained. The majority of solar cells produced today are based on crystalline silicon. This has several reasons. Earth's crust consist of 27% silicon making it the second most abundant element (after oxygen with 46%), silicon is non-toxic and from the microelectronic industry a broad processing experience is available. Despite these good properties the theoretical maximum efficiency for a "simpel" solar cell<sup>ii</sup> is defined by the the Shockley-Queisser limit [4] to only 30%, and about 15-20% for commercial available silicon based devices. This low efficiency is mainly caused by a mismatch between the band gap energy of silicon and the wide spectrum of photon energies in sunlight, thus reducing the amount of photons available for current generation.



*Figure 1.1:* AM1.5G solar spectral irradiance as a function of wavelength, $\lambda$ . The electromagnetic spectrum is colour coded: Ultra violet (blue), visible (green), near-infrared (yellow) and infarred (red). The vertical dashed line indicates the band gab energy for silicon.

<sup>i</sup>Rounded numbers.

<sup>&</sup>lt;sup>ii</sup>Single PN junction cell.

Figure 1.1 shows the standard AM1.5G<sup>iii</sup> [5] solar spectral irradiance as a function wavelength,  $\lambda$ , and photon energy. The vertical line indicates the band gap energy of silicon ( $\approx$ 1.1eV) corresponding to a wavelength of approximately 1100nm. All photon energies to the right of this line are not utilized for current generation. They represent a loss. Silicon based solar cells mainly generate current by absorbing photons within the visible and near infrared spectrum ( $400nm \le \lambda \le 1100nm$ ). Thus a majority of the solar spectrum does not contribute to the current generation, as the low energy photons ( $\lambda \ge 1100nm$ ) simply pass through the cell in what is called sub-band gap losses. The very high energy photons (ultra violet and visible spectrum) does contribute to the current generation. These losses are called ohmic or thermalilization losses.

### **1.1 SunTune project**

This work is a part of the SunTune project [6] which addresses efficiency improvements of solar modules by "tuning" the spectrum of sunlight to better match the range of efficient current generation. Figure 1.2 exemplifies the main concept which the SunTune project builds on.



*Figure 1.2:* Sketch of the SunTune concept. Additional material layers placed on the rear side of a silicon solar cell. Downshifter) Near-infrared photons are downshifted to photons in infreared spectrum (yellow to red). Upconverter) Infrared photos are upconverted to the visible range (red to green). Ultra violet photos are mainly lost as thermalilization losses (blue). A mirror reflects light upwards.

By application of additional material layers to the rear side of the silicon-cell, it is possible to convert two low energy photons to one photon with a enough energy to bridge the band gap energy. Thus, such a layer absorbs the long wavelength photons (infrared spectrum) that otherwise are transmitted through the silicon and converts them to photons with shorter wavelengths (near-infrared spectrum). These photons are reflected back towards the silicon layer where they are absorbed and thereby contribute to the current production. This process is named *upconversion* and can take place in some active rare earth ions, such as  $Er^{+3}$ , doped into a host material like  $TiO_2$ . As with silicon such ions have an intrinsic absorption band, which for  $Er^{+3}$  ions are wavelengths between 1460nm-1560nm. In order to also utilize wavelengths from 1100nm to 1460nm a downshifter is applied. This is essentially a reversed upconversion process which enables photon energies with shorter wavelengths than the absorption band of  $Er^{+3}$  to be absorbed and then upconverted to wavelengths absorbable to silicon.

<sup>&</sup>lt;sup>iii</sup>AM1.5G is the solar spectrum revived by the Earth (terrestrial) under a defined angle of 48.2° relative to normal incidence. This way the radiation passes 1.5 times the air mass compared to normal incidence. The "G" stands for global.

#### 1.1. SUNTUNE PROJECT

Hence by using a downshifting and a upconverting layer otherwise unavailable wavelengths (1100nm to 1560nm) become available and hereby increases the efficiency of silicon based solar cells. The potential increase in overall efficiency for silicon based devices is in the range of 4 percentage points. Furthermore, solar cells spend a considerable part of their lifetime illuminated at less than ideal conditions such as in partly cloudy weather, where the solar spectrum is shifted towards longer wavelengths. For such cases the advantages of upconversion yields an additional and significant energy gain when averaged over a year.

The upconversion process is naturally very inefficient and without any additional light enhancement the process is negligible in regards to increasing the efficiency of the solar cell. This enhancement can be obtained by exploiting the resonant nature of metal nanoparticles when interacting with light in which the local near (electrical) field is enhanced. This resonance phenomena is called a plasmos and as with resonance of macro scaled objects the resonance frequency is influenced by parameters such as size, shape and material.

The main concept of the SunTune project is thus to utilize plasmonic enhanced upconversion for increasing the solar cell efficiency with a best case scenario of approximately 4 percentage points for silicon based solar cells.

Innovation Fund Denmark (formerly the Danish Council for Strategic Research) is investing DKK 23 million in the SunTune project which is a collaborative project between the following partners, each contributing with different areas of expertise.











Technical University of Denmark



International Solar Energy Research Center Konstanz

# **Project objectives**

This work is a part of the SunTune project, and focuses on how to diverse a topology optimization algorithm capable of solving a given optimization problem in a efficient manner while keeping production constraints in mind. The main project objective is to create a numerical framework able to obtain topologies for nanoparticles enabling enhancement of the electric field in the vicinity of particles. Also known as local near field enhancement. The nanoparticles shall be optimized towards enhancement for incoming light, at around 1500nm wavelength The numerical methods applied are the finite element method in combination with the topology optimisation strategy. This work is included as a part of the SunTune workpackage 3 contaning the following objectives:

- i) Establishing a numerical modelling setup that calculates field enhancement combined with local density of optical states for a given geometry.
- ii) Develop a topology optimization framework based on the numerical model.
- iii) Single-layer optimization metals and other materials.
- iv) Optimizing three-dimensional near-field-enhancing nano materials.

Until now the focus of this work has been on i) to iii) with main emphasis on the 2D case. Currently 2D topology optimization is possible for both single- and multi layered cases using a COMSOL Multiphysics model controlled and manipulated by custom made Matlab scripts via COMSOL's LiveLink feature. It is believed that these scripts can be adopted to control a 3D model with relative ease, hence addressing part iv). However, as the main framework is somewhat established a multitude of optimization specific parameters are still to be defined in order to obtain consistent and stable topologies producible via the Electro Beam Lithography or nanoimprint process available in the SunTune project. Defining these parameters are currently the main objective of my work. Examples of such parameters are,

- Defining a generic guideline/rule of thumb for initializing a optimization study, such as projection threshold, initial design.
- Topology optimization specific parameters such as:
  - $\beta$ -update strategy.
  - Filter radius and update strategy.
  - Robust formulation parameters to take production tolerances into account.

The following status report will focus mainly upon definition of a generic guide line, as this is a prerequisite for definition of the remaining parameters. A brief introduction to Maxwells equations and the wave equation is given in chapter 3, plasmonics and upconversion in chapter 4 and the overall methodology of topology optimization in chapter 5.1. Preliminary results are presented in section 5.2 through section 5.4.

**Submitted paper**: Co-author to a submitted paper to the journal of Applied Physics Letters. *"Plasmonically enhanced upconversion of 1500 nm light via trivalent Er in a TiO2 matrix"*, [7] Date of submission 31. October 2016.

## Time-harmonic electromagnetic waves

This chapter briefly describes the governing equation for the electromagnetic field problems solved in this work together with choice for excitation of these problems.

The governing equations for all electromagnetic problems are Maxwell's equations [8],

$$\nabla \cdot \mathbf{D} = \rho, \qquad \nabla \cdot \mathbf{B} = 0,$$
  

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \qquad \nabla \times \mathbf{H} = \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t}.$$
(3.1)

E = E(r, t) and H = H(r, t) are the electric and magnetic field respectively, with r = r(x, y, z) and  $\rho$  being the charge density. The three auxiliary fields, D, B and J are related to the electric and magnetic fields through the three constitutive equitations,

$$D = \epsilon E, \quad B = \mu H \text{ and } J = \sigma E.$$
 (3.2)

Where *D* is the displacement field, *B* is the magnetic flux density and *J* is the current density. The electric permittivity,  $\epsilon = \epsilon_r \epsilon_0$ , the magnetic permeability,  $\mu = \mu_r \mu_0$ , and the electric conductivity,  $\sigma$ , are intrinsic material properties and are in general functions of (angular)frequency,  $\omega$ . Determining the response of any object subject to an applied electromagnetic wave is the matter of solving the the wave equation subject the interface conditions stating that the components of *B* and *D* normal to a to an interface and tangential components of *E* and *H* to an interface must be continuous.

#### 3.1 Wave equation

Light is electromagnetic waves propagating through air or any other medium. As opposed to acoustic waves which propagate by compressing the medium in which they travel, electromagnetic waves can propagate through empty space it self. The two curl equations of (3.1) are two coupled first order differential equations which can be de-coupled at the expense of increasing the order of differentiation. Taking the curl of the two curl equations in (3.1) and using the constitutive equations and the relations  $\epsilon = \epsilon_r \epsilon_0$ ,  $\mu = \mu_r \mu_0$  with  $c_0 = (\mu_0 \epsilon_0)^{-1/2}$  yields the two vector wave equation for the electric and the magnetic fields in homogeneous and isotropic matter.

$$\nabla \times (\mu_r^{-1} \nabla \times E) + \mu_0 \frac{\partial J}{\partial t} + \frac{\epsilon_r}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \mathbf{0} \quad \text{and} \quad \nabla \times (\epsilon_r^{-1} \nabla \times H) + \frac{\mu_r}{c_0^2} \frac{\partial^2 H}{\partial t^2} - \frac{1}{\epsilon_r} \nabla \times J = \mathbf{0}.$$
(3.3)

In this work the wave propagation is time harmonic allowing the fields to be re-written in the form  $A(\mathbf{r}, t) = \hat{A}(\mathbf{r})cos(\omega t + \phi)$  where  $\omega$  and  $\phi$  are the frequency and phase of the wave. Using exponential functions  $A(\mathbf{r}, t)$  can be expressed using the complex phasor  $\tilde{A}(\mathbf{r})$ ,

$$A(\mathbf{r},t) = \hat{A}(\mathbf{r})\cos(\omega t + \phi) = Re[\hat{A}(\mathbf{r})e^{i\phi}e^{i\omega}] = Re[\tilde{A}(\mathbf{r})e^{i\omega t}], \qquad (3.4)$$

with  $Re[\cdot]$  denoting the real part. Phasor notation simplifies the mathematics as  $\partial/\partial t$  corresponds to multiplication by  $i\omega$ . Applying phasor notation (3.3) is rewritten from the time domain to the

frequency domain. The tilde i omitted.

$$\nabla \times (\mu_r^{-1} \nabla \times E) + i\omega \mu_0 J - k_0^2 \epsilon_r E = \mathbf{0} \quad \text{and} \quad \nabla \times (\epsilon_r^{-1} \nabla \times H) - \frac{1}{\epsilon_r} \nabla \times J - k_0^2 \mu_r H = \mathbf{0}$$
(3.5)

Where,  $k_0 = \frac{\omega}{c_0}$ . The physical fields of the phasor fields solved for in (3.5) are obtained by taking the real part as stated in (3.4). In dispersive matter the electric permittivity  $\epsilon_r$  is complex and is denoted by

$$\hat{\epsilon_r} = \epsilon_r - \frac{i\sigma}{\omega} = \epsilon' - i\epsilon''. \tag{3.6}$$

In this work the values of  $\epsilon_r$  and  $\sigma$  are not know explicitly hence the permittivity is derived from the complex refractive index via  $\hat{n} = \sqrt{\hat{\epsilon}_r \mu_r}$  where  $\hat{n} = n + i\kappa$  and  $\mu_r \approx 1$  as the materials are non-magnetic. This explicitly yields

$$\epsilon' = n^2 - \kappa^2 \quad \text{and} \quad \epsilon'' = 2n\kappa.$$
 (3.7)

*n* and  $\kappa$  are the refractive index and extinction coefficient respectively, determined via reflectivity studies. Both are functions of frequency.  $\kappa$  determines the absorption of electromagnetic wave propagating through a given medium and is related to exponential attenuation of the field intensity,  $I(\mathbf{r})$  as the wave propagates through the medium, [9],

$$I(\mathbf{r}) = I_0 e^{-\alpha \mathbf{r}}$$
 with  $\alpha(\omega) = \frac{2\kappa\omega}{c}$ . (3.8)

With  $\alpha$  being the attenuation constant. The "2" in the nominator originates from the fact that  $I \propto E^2$  with  $E \propto e^{ikx}$  hence  $I \propto e^{i2kx}$ , with *k* being the wave number which through the dispersion relation can be related to  $\kappa$ . Thus the imaginary part of  $\hat{\epsilon}$  is related to absorption loses in materials. The penetration depth is defined via the attenuation constant as,

$$\delta_p(\omega) = \frac{1}{\alpha(\omega)},\tag{3.9}$$

defining a characteristic length for which the intensity of the electric wave has decreased to 1/e or approximately 37% relative to the surface intensity. Often, also the skin depth is used and is defined as  $\delta_s(\omega) = 2\delta_p(\omega) = 2\alpha(\omega)^{-1}$  and defines a length/depth where the surface intensity is attenuated to  $1/e^2$  or  $\approx 13\%$ .

#### 3.2 Background excitation method

The inhomogeneous wave equations in (3.5) governs the electromagnetic field problems. In this work COMSOL Multiphysics is used to numerically solve the electric wave equation All numerical models are excited using the background field excitation method where the background field  $E_b$  is taken as the analytical solution without any scatterer (nanoparticles) present leaving only the scattered field  $E_s$  as the unknown. The total field E is formulated as  $E = E_b + E_s$  in which the electric wave equation in (3.5) is rewritten using the scattered field formulation

$$\nabla \times (\mu_r^{-1} \nabla \times \mathbf{E}_s) - k_0^2 \epsilon_r \mathbf{E}_s = -\nabla \times (\mu_r^{-1} \nabla \times \mathbf{E}_b) + k_0^2 \epsilon_r \mathbf{E}_b.$$
(3.10)

The analytical solution to  $E_b$  is obtained using the transfer matrix method for multi layered systems. The transfer matrix method is briefly explained in the following.

Figure 3.1 shows a multi layered system with a plane wave incident from the left and propagating in the direction of k. Due to different refractive indices of each layer some light is reflected and



Figure 3.1: External reflection and transmission at a multi layered system.

*Figure 3.2:* Refractive indices of a multi layered system consisting of N layers.

some transmitted through the layers. The ratio of externally reflected and transmitted light can be related to a total transfer matrix M which takes into account the internal reflected and transmitted light for each layer. Figure 3.2 shows the axis orientation together with the interface planes,  $x_i$ . Where,  $\hat{n}_i$  is the complex refractive index of layer i of N layers and  $x_i$  marks the interface between two layers, i and (i + 1).

The electric field in layer *i* is given by

$$E_i(x, y, z) = E_F(x)e^{-ik_{z,i}z} + E_B(x)e^{-ik_{z,i}z}.$$
(3.11)

Where  $E_F$  and  $E_B$  are the amplitudes of the forward (transmitted) and backward (reflected) propagating waves, respectively. The components of the wave vector is related to the ray angle  $\theta_i$ , such that  $k_{x,i} = |\mathbf{k}_i| \cos(\theta_i)$ , with  $|\mathbf{k}_i| = \frac{\omega}{c} n_i = \frac{2\pi}{\lambda_0} n_i$  where  $\lambda_0$  is the wavelength of the incident wave.

Figure 3.3 shows the forward and backward wave amplitudes at, a interface and within a single layer. The wave propagation through the *N* layered system is expressed by the total transfer matrix



*Figure 3.3:* Amplitudes of incident, reflected and transmitted waves. a) At a interface  $x_i$  between two layers with different refractive indices,  $n_i \neq n_j$ . b) Within a layer. Super indices  $(^-)$  and  $(^+)$  denotes before and after an interface,  $x_i$ .

*M*, relating the amplitudes of the reflected and transmitted waves at the first and last layer. Super indices  $(^{-})$  and  $(^{+})$  denotes before and after an interface,  $x_i$ .

$$\left\{ \begin{array}{c} E_F(x_1^{-}) \\ E_B(x_1^{-}) \end{array} \right\} = M \left\{ \begin{array}{c} E_F(x_{N-1}^{+}) \\ E_B(x_{N-1}^{+}) \end{array} \right\} = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \left\{ \begin{array}{c} E_F(x_{N-1}^{+}) \\ E_B(x_{N-1}^{+}) \end{array} \right\}$$
(3.12)

*M* is given by the cyclic product of propagation through a layer and over a interface

$$M = T_1 I_{12} T_2 I_{23} \dots T_N I_{(N-1)N},$$
(3.13)

where *T* and *I* are the transfer matrices for propagation through a layer and over a interface respectively. The transfer matrices  $T_i$  are obtained by propagating the forward and backward going waves from a interface through the thickness of a given layer,  $d_i = x_{i+1} - x_i$  and relating amplitudes just after a interface  $x_{i-1}$  and just before the next interface  $x_i$  as

$$\left\{ \begin{array}{c} E_F(x_i^-) \\ E_B(x_{i-1}^+) \end{array} \right\} = T_i \left\{ \begin{array}{c} E_F(x_{i-1}^+) \\ E_B(x_i^-) \end{array} \right\}, \quad T_i = \begin{bmatrix} e^{j\Phi_i} & 0 \\ 0 & e^{-j\Phi_i} \end{bmatrix}, \text{ for } i = 1, 2, \dots, N$$
(3.14)

with  $\Phi_i = k_{x,i}d_i = \frac{2\pi}{\lambda_0}n_id_icos(\theta_i)$ . The interface matrices,  $I_i$  are obtained by using the polarization dependent Fresnel coefficients [8]  $r_{ij}$  and  $t_{ij}$  and relating the amplitudes on each side of an interface as

$$\left\{ \begin{array}{c} E_F(x_i^-) \\ E_B(x_i^-) \end{array} \right\} = \mathbf{I}_{ij} \left\{ \begin{array}{c} E_F(x_j^+) \\ E_B(x_j^+) \end{array} \right\}, \quad \mathbf{I}_{ij} = \frac{1}{t_{ij}} \begin{bmatrix} 1 & r_{ij} \\ r_{ij} & 1 \end{bmatrix},$$
(3.15)

When all "local" matrices known, the total transfer matrix is obtained through equation (3.13) substituted into equation (3.16). Setting  $E_B(x_{N-1}^+) = 0$  as no light is reflected within the last layer, hence,

$$\left\{ \begin{array}{c} E_F(x_1^-) \\ E_B(x_1^-) \end{array} \right\} = M \left\{ \begin{array}{c} E_F(x_{N-1}^+) \\ E_B(x_{N-1}^+) \end{array} \right\} = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \left\{ \begin{array}{c} E_F(x_{N-1}^+) \\ 0 \end{array} \right\}$$
(3.16)

Equating into two equations with two unknowns,  $E_B(x_0^-)$  and  $E_F(x_{N-1}^+)$ , which is solved for and hereafter are the internal wave amplitudes obtained using each of the individual matrices. One work through layers from "left to right" using the  $E_F(x_0^-)$  and  $E_B(x_0^-)$  amplitudes or from "right to left" using  $E_F(x_{N-1}^+)$  and  $E_B(x_{N-1}^+)$ .

Using equation (3.11) a single expression for the systems total E-field can be obtained as a function of each of the layers. This is the background field expression implemented into COMSOL.

## Plasmonic enhanced upconversion

This chapter introduces main concepts of plasmonic enhanced upconversion as the overall nature of these phenomena serves, as input the topology optimization conducted in this work and explained chapter 5.

### 4.1 Upconversion

In 1913 Bohr purposed a quantized shell model for the Hydrogen atom [10] which required that electrons move in orbits of fixed size and energy. He derived an expression for the energy as a function of these shell-orbits,

$$E = \frac{-13.6Z^2}{n^2} \text{eV},$$
 (4.1)

where Z is the atom number,  $n^i$  is the quantum state number and -13.6eV is the energy of the lowest quantum state, n = 1, called the ground state. Figure 4.1 shows a sketch of Bohr's shell model with indicated energy levels (qualitative ) associated with these quantum states. Equation (4.1) states that the bonding energy between the electron and the nuclei decreases for increasing quantum state, and at some point the atom is ionised as,  $E \ge 0$  eV. Thus an electron can get excited from



Figure 4.1: Sketch of Bohr's shell model for a Hydrogen atom.

any quantum state to a higher one, e.g. from  $|1\rangle$  to  $|2\rangle$ . Conservation of energy, however still apply, hence excitation of a electron to a higher quantum state requires addition of energy and de-excitation from a higher state to a lower state requires that excess energy must be emitted out of the system. Upconversion is, very simplified, the process where a number of incident low energy photons are converted into one higher energy photon by excitation of electrons followed by radiative de-excitation. These processes can occur in some rare earth ions, e.g. in Er<sup>+3</sup>. Figure 4.2 shows possible upconversion mechanisms involving a single ion (left) and between two ions (right). Single iron upconversion takes place when a free electron is first excited from the ground state  $|1\rangle$ by absorption of one photon with energy  $hv_{e1}$  to the first extricated state  $|2\rangle$ . This process is called ground state absorption (GSA). Hereafter the ion absorbs an additional photon,  $hv_{e2}$ , and is exited from  $|2\rangle$  to  $|3\rangle$ . This is called excited state absorption (ESA). h is Plancks constant, and  $v_{e1}$  is the frequency of photon 1. After some time the exited ion relaxes to back to the ground state, and this

<sup>&</sup>lt;sup>i</sup>Not to be related to the refractive index

relaxation process can occur radativly, with photon emission where in general,  $hv_{e1} < hv_r$ . Two ion upconversion process takes place when both ions are excited by ground state absorption, then one ion relaxes to a lover energy state by radiative energy transfer to the second ion. The second ion is then excited from  $|2\rangle$  to  $|3\rangle$  with the higher energy level,  $E_3$ . This process involving energy transfer is called energy transfer upconversion (ETU).





**Figure 4.2:** Possible excitation processes.(Left) One ion 2-step absorption (GSA) followed by (ESA). (Right) Two ion absorption. (GSA) followed by (ETU).  $hv_e$  the photon energy of the absorbed photons enabling the excitation, and  $hv_r$  is the energy of the radiated photon  $E_i$  indicates the energy level of the different quantum states.

**Figure 4.3:** 4f energy levels of  $Er^{+3}$ . Black arrows corresponds to excitation paths at 1500nm excitation. Coloured solid arrows shows radiative decay/relaxation the corresponding emission wavelength indicated. Gray dashed arrows correspond to possible non-radiative decays.

Relaxation can in general occur through two competing processes, the aforementioned radiative process and a non-radiative de-excitation process. Only the radiative relaxation process is of interest in relation to upconversion, as this involves photon emisiion. A third excitation process is also possible, however with less probability, where one or more phonons assist the excitation of the second ion by non-radiative relaxation of the first ion. Figure 4.3 shows the energy levels in the partly field 4f orbital in Er<sup>+3</sup> ions and possible wavelengths of the emitted/upconverted photons.

The upconversion process is highly dependent of the intensity of the incoming light, and [11] showed that the intensity of the emitted/upconverted photon is proportional to the intensity of the incoming light,

$$I_{UC} \propto I_{in}^{n_p}. \tag{4.2}$$

Where  $I_{in}$  and,  $I_{UC}$  are the intensities of the incoming light and of the emitted light from the upconversion process with radiative relaxation, respectively.  $n_p$  is a number associated with the number of photons required for excitation of a electron. Experiments with ion excitation by focused laser beams have shown that  $n_p = 2$  for low beam intensity and n = 1 for high beam intensity as the upconversion saturates. A average value of  $n_p = 1.5$ , has also been observed, [7].

The upconversion process shown in figure 4.2 is very simplified and in general a multitude of extrication and relaxation process can occur, these are however out of the scope of this work. Emphasis is laid on the intensity relation of (4.2) and the fact that Er<sup>+3</sup> ions can absorb low energy photons with wavelengths between 1480nm to 1560nm and emit (when radiative relaxation occur) photons with a wavelength in the visible and near-infrared spectrum, as shown in figure 4.3.

The efficiency of the upconversion process in natural very weak, and in order to enhance the efficiency, one can either increase the probability of radiative de-excitation, or increase the number

of photons available for ion excitation. The latter option can be obtained, by utilizing the interaction between light and nano scaled objects in which the incoming light is enhanced due to material polarization trough plasmonic processes.

### 4.2 Plasmonics

The constitutive relation  $D = \epsilon E$ , from (3.2) relates a materials response of the displacement field to a applied electric field. As mentioned in chapter 3 the permittivity, is a function of frequency,  $\epsilon(\omega)$ , meaning that a given material has a finite response time. By modelling the frequency dependent permittivity classically a simple model for the plasmonic behaviour of noble metals can be obtained. The Drude model takes this classical approach in which the true quantum mechanical behaviour of the free conduction electrons is simplified. The conduction electrons are assumed non-interacting moving freely around in the crystal lattice with a periodic potential (Bloch periodicity) created by the immobile positive ion cores, [12]. One can thus model the oscillating motion of the free electrons in response to an applied electromagnetic field via the classical equation of motion  $m\ddot{z}(t) + c\dot{z}(t) + kz(t) = F(t)$  which for a free-electron gas is given by,

$$m\ddot{z}(t) + \frac{m}{\tau}\dot{z}(t) = -qE_{tot}(t).$$
(4.3)

Where *m* is the electron effective mass<sup>ii</sup>, *z* is the relative displacement of the electron wrt. the atom nuclei,  $\tau$  is the decay/relaxation time taking into account that the electronic motion is damped by collisions with other scatters in this free electron gas<sup>iii</sup>. *q* is the electron charge and  $E_{tot}(t)$  is the total electric field acting on the electron. Figure 4.4 shows the one-dimensional charge distribution



*Figure 4.4:* One dimensional bulk plasmon response to external field. The external field  $E_{ext}$  polarizes the material and alters the charge density  $\rho$  which creates bulk plasmons.

induced in bulk material by the external field,  $E_{ext}(t)$ . In equilibrium the charge distribution,  $\rho$ , is uniformly distributed but as the electrons react to the external field the charge distribution is changed. Due to accumulated positive charges one side and negative charges accumulated on the other. As the external field oscillates a non-uniform charge distribution is created as the electrons oscillate with a distance of z(t). This motion of moving charges creates bulk oscillations called plasma oscillations. Given that the external field is time harmonic and oscillating with the frequency  $\omega$  the total field and the electronic charge distribution will oscillate with the same frequency,

$$E_{tot}(t) = Re[E_{tot}(\omega)e^{-i\omega t}] \quad \text{and} \quad z(t) = Re[z(\omega)e^{-i\omega t}].$$
(4.4)

Substituting (4.4) in (4.3) equates an expression for the electronic motion relative to the fixed atomic nuclei,

$$z(\omega) = \frac{qE_{tot}(\omega)}{m(\omega^2 + i\omega/\tau)}.$$
(4.5)

<sup>&</sup>lt;sup>ii</sup>Often denoted  $m^*$ .

<sup>&</sup>lt;sup>iii</sup>Often also the free collision times  $\gamma = \frac{1}{\tau}$  is used to describe damping effects

The collective motion of moving charges/electrons polarizes the material with the polarization density given by  $P = -qzn_e$ , with  $n_e$  denoting the electron density. The displacement field is then given by

$$D(\omega) = \epsilon_0 E_{tot}(\omega) + P(\omega). \tag{4.6}$$

Substitution of equation (4.5) into equation (4.6) equates the complex Drude permittivity/dielectric function [8],

$$\frac{\tilde{\epsilon}(\omega)}{\epsilon_0} = \left[1 - \frac{\omega_p^2 \tau}{1 + \omega^2 \tau}\right] + i \left[\frac{\omega_p^2 \tau}{\omega} \frac{1}{1 + \omega^2 \tau^2}\right] \quad \text{with} \quad \omega_p = \sqrt{\frac{q^2 n_e}{\epsilon_0 m}},\tag{4.7}$$

 $\omega_p$  is the bulk plasma/plasmon frequency. The total field  $E_{tot}$  is the sum of the applied external field  $E_{ext}$  and the field induced due to the polarization, *P*, thus  $E_{tot}(\omega) = E_{ext}(\omega) + \epsilon_0^{-1}P(\omega)$ . Substitution of z(t) from equation (4.4) and the explicit expression of  $E_{tot}(\omega)$  into the equation of motion of (4.3) and collecting terms of electronic displacement  $z(\omega)$  equates [12],

$$z(\omega)\left(\omega^2 + \frac{i\omega}{\tau} - \frac{q^2 n_e}{\epsilon_0 m}\right) = \frac{q}{m} E_{ext}(\omega) \rightarrow \lim_{\tau \to \infty} i\omega_p z(\omega_p) = \tau \frac{q}{m} E_{ext}(\omega_p).$$
(4.8)

At the bulk plasmon frequency  $\omega = \omega_p$  and in the limit with no damping when  $\epsilon' \to 0$  ( $\tau \to \infty$ ), the electron motion  $z(\omega)$  remains finite for an arbitrary small applied field. Meaning that at  $\omega = \omega_p$  the electrons are excited for even a infinite small applied external field. This is a resonance behaviour of the conduction electrons and the phenomenon is called *bulk plasmons*. Thus, a plasmon is an excitation of a electron, and bulk plasmons are the collective oscillations (waves) of the excited conduction electrons. In metals  $\tau \neq \infty$  due to absorption, as indicated through the imaginary part of complex permittivity. In general is the bulk plasmon resonance frequency well defined/sharp for low loss materials whereas it is broaden with an increased imaginary part of the permittivity, due to absorption losses.

The important property of plasmons originates from the material polarization which increases the electric (near)field by the aforementioned relation

$$E_{tot}(\omega) = E_{ext}(\omega) + \epsilon_0^{-1} P(\omega).$$
(4.9)

These bulk plasmon waves are longitudinal and they propagate parallel to direction of the wave propagation, meaning that bulk plasmons can not be excited by the *E*-field in an electromagnetic wave, hence light cannot excite bulk plasmons, [9, 12]. However a wide range of plasmons modes exist, some of which couple to light.

Metal nano particles can support such modes as the plasmonic behaviour is not governed by the wave vector, k but by the geometry of the particle it self. Figure 4.5 shows lower order modes for different particle geometries. The lowest order of a spherical particle is dipole-like as negative and positive charges accumulate on opposite sides of the particle. By extending the sphere in one direction only, a nano-rod is effectively obtained which support longitudinal and transverse modes depending on the axis, the external field drives the charges along. The polarization sets up an Coulomb restoring force between the displaced electrons and the ion cores and the strength of this force is dependent of the accumulative charge distribution on the surface. This restoring force determines the frequency at which the electrons oscillate. Thus to sum up, the plasmons resonance of metal nano particles depends on the shape, determining how charges accumulate at the surface, and on the electron density,  $n_e$ , that determines the amount of accumulative charge. The high electron density in noble metals like silver and gold results in plasmon resonances in the visible and near infrared spectrum.



*Figure 4.5:* Sketch of geometry dependent plasmons modes in nano particles, [12]. Left) Dipole mode of nano sphere. *Right)* Longitudinal and traverse modes of nano rods.

For simple geometries analytical expressions for the polarizability can be derived using an quasistatic approximation. For the sphere the quasistatic approximation is that the particle radius, *a* is much smaller than the wavelength of the external field<sup>iv</sup>,  $a \ll \lambda$ . The electric field, *E*, inside and outside of the sphere can be expressed using potential theory with the electric potentials expressed in terms of Legendre polynomials. From thus expression the size dependent polarizability can be obtained as [12],

$$\alpha_p = \frac{(\epsilon_{np} - \epsilon_m)\epsilon_m a^3}{\epsilon_{np} + 2\epsilon_m}.$$
(4.10)

Here  $\alpha_p$  is the polarizability<sup>v</sup>, describing the ability for a given material to be polarized/to alter the charge density,  $\rho$ , from the equilibrium state.  $\epsilon_{np}$  and  $\epsilon_m$  are dielectric functions for the metal nanoparticle and the medium in which the nanoparticle resides, respectively. The external field polarizing the sphere induces a dipole moment, p, within the sphere proportional to the magnitude of the external field  $p = \alpha_p \epsilon_0 E_{ext}$  from which the polarization density is  $P = n_e p = n_e \alpha_p \epsilon_0 E_{ext}$ .

Plasmonic modes at the interface between a dielectric and a metallic medium can also occur. Here only the metal surface is polarized and the plasmon wave propagate along the surface with oscillations decaying exponentially into the two media. Such modes are also called surface plasmon polaritons (SPP). The conditions for such interface modes to occur are [13],

$$\epsilon_d \epsilon_m < 0, \quad \text{and} \quad \epsilon_d + \epsilon_m < 0.$$

$$(4.11)$$

Where indices  $_d$  and  $_m$  denotes the dielectric and metallic media respectively. Which means that  $\epsilon_m \leq -\epsilon_d$  must be satisfied in order for a metallic surface to support interface modes.

<sup>&</sup>lt;sup>iv</sup>Ignoring retardation effects so that the variation of incident plane wave across the particle is approximated to be constant.

<sup>&</sup>lt;sup>v</sup>Index *p* is used to distinguish the polarizability  $\alpha_p$  from the attenuation  $\alpha$  constant in equation (3.8).

# **Topology** optimization

Within this chapter a brief introduction to the methodology of topology optimization is given in chapter 5.1 and prepares the reader in together with chapters 3 and 4 for the intermediate results descried in sections 5.2 through 5.5 dealing specifically with the optimization conducted in this work until now.

## 5.1 Methodology

The framework for topology optimization was originally purposed by Kikuchi and Bendsøe [14] and was suggested as an alternative to classical shape optimization in which one seeks to find a optimized shape of a initially given design represented by some basis functions describing the boundary of the initial design. In shape optimization the initial design is known a priori and therefore poses a bias to the topology (layout) of the optimized design as the iterative optimization is then effectively a restricted search within variations of the initial design. Topology optimization is a gradient based optimization method that work by varying the spatial distribution of materials within a bounded design domain. Consequently no explicit formulation of the design exists and the shape of the topology can be changed without any geometrical constrains. The original paper was introduced in 1989 to minimize the compliance of continuum structures and since the method has proved successful for optimization within areas such as fluid dynamics (e.g. [15, 16]), acoustics, (e.g. [17, 18]), micro electro mechanical systems (e.g. [19, 20]) and also within the field of plasmonics (e.g. [21]).

A generic formulation of a topology optimization problem could be in the form

minimize	$\Phi(u(x), x)$	$\forall \pmb{x} \in \Omega_e$	objective function	
subject to:	$g_i(\boldsymbol{u}(\boldsymbol{x}), \boldsymbol{x}) \leq 0,$	i = 1, 2, m	constraint functions	
	S(x)u(x) - f(x) = 0		state problem	(0.1)
	$0 \leq x \leq 1$ ,	$\forall x \in \Omega_d, x = 0  \forall x \notin \Omega_d$	design variable bounds.	

 $\Phi$  is the objective function one seeks to minimize<sup>i</sup> and is a measure of a given designs performance evaluated in some domain  $\Omega_e$ . x is a vector containing the design variables for all n design variables within the bounded design domain  $\Omega_d$ . The state equation is the finite element problem to be solved. An interpolation scheme is used to model the relation between the design variables and the element material, e.g. a linear interpolation in the form

$$M(x_e) = A + x_e(B - A).$$
 (5.2)

Where  $M(x_e)$  is the material at element *e* with the value of  $x_e$ . *A* and *B* are the material properties of the in this case two constituents used, which could be "void" and material or two "real" materials,.

<sup>&</sup>lt;sup>i</sup>(5.1) can be formulated as a maximization problem by e.g. minimizing the negative of the objective function,  $-\Phi$ .

e.g. (A, B) = (Air, metal) or  $(A, B) = (SiO_2, Au)$ . Bendsøe introduced the concept of the density approach [22] where a continuous scalar density,  $\rho$ , is associated with each element, in the design domain such that  $0 \le \rho_e \le 1$ , efficiently making the element wise densities the design variables.  $x_e = \rho_e$ .

The aim of topology optimization is then to obtain a clear binary (0/1) material(density) distribution that minimizes the objective function  $\Phi$  subject to the given set of constraints,  $g_i$ , as in equation (5.1). However interpreting the design variables  $\rho$  as physical densities and directly associating these densities with the material properties e.g. Young's moduli or the refractive index might result in mesh-dependent designs. Thus a refinement of the mesh would lead to completely different designs instead of only improving the finite element modelling of the same optimal design. Furthermore, utilizing linear elements density checker board patterns may occur, e.g. where an element would have  $\rho_e = 1$ , and adjacent elements  $\rho_e = 0$ , thus obtaining a non-physical discrete material distribution. A popular method to prevent mesh dependency and checker board designs is by application of a density filter. [23] purposed a localized density filter which regularizes the original problem by defining the element wise density to be a weighted average of the surrounding element densities within some circular (2D) or spherical (3D) neighbourhood defined by a set radius. The density filter method introduces a filtered density field,  $\tilde{\rho}$ . Such filtering methods introduce gray element regions with intermediate densities which for some optimization problems does not pose any issues when interpreting the final design. However, in other optimization problems, e.g. electrodynamics, designs with gray scaled regions may result in a wrong physical interpretation. Projection methods that project the filtered densities,  $\tilde{\rho}$ , to a 0/1 density field,  $\bar{\tilde{\rho}}$ , is then often applied to obtain designs with clear material boundaries.

[24] purposed an alternative to the localized density filter in [23] where the filtered densities,  $\tilde{\rho}$ , are implicit defined by solution of the Helmholtz PDE

$$-r^2 \nabla^2 \tilde{\rho} + \tilde{\rho} = \rho \tag{5.3}$$

with a homogeneous Neumann condition  $\frac{\partial \tilde{\rho}}{\partial n} = 0$  on the design domain boundary,  $\Gamma_d$ . Here *r* plays a similar role as the neighbourhood defining radius in the localised filtering method. Additionally, [25] suggested a modified threshold projection method in which the projected densities,  $\bar{\rho}$ , are defined using the tangent hyperbolic function,

$$\bar{\tilde{\rho}}_e = \frac{tanh(\beta\eta) + tanh(\beta(\tilde{\rho}_e - \eta))}{tanh(\beta\eta) + tanh(\beta(1 - \eta))}.$$
(5.4)

 $\beta \in [1, \infty[$  is a scalar parameter determining the level of projection, and  $\eta \in [0..1]$  is a projection threshold parameter. For a high  $\beta$ -value equation (5.4) approaches the Heaviside step function. The value of  $\beta$  is continuously increased during the optimization process in order obtain a high contrast design where elements contain either of the constituents used e.g. as in equation (5.2). Also by continuously increasing  $\beta$ , smoothness of the projection function is maintained which relaxes the optimization problem<sup>ii</sup>. The threshold parameter  $\eta$  determines for which the intermediate values of the filtered densities  $\tilde{\rho}_e$  is projected towards either  $\tilde{\rho}_e = 0$  or  $\tilde{\rho}_e = 1$ . All filtered densities below  $\eta$  is projected towards 0 and all above towards 1 at a degree dependent upon the value of  $\beta$ . Using the density filter approach it is important to note that the projected densities  $\tilde{\rho}$  are used to interpolate between materials, as  $\tilde{\rho}$  for high  $\beta$  represent the physical densities.

Figure 5.1 shows the three densities for a 1D example in which the  $\rho$  is filtered using an analytical solution to (5.3) with an arbitrary filter radius of r = 50. The filtered densities are then projected

<sup>&</sup>lt;sup>ii</sup>For low  $\beta$  values.



*Figure 5.1:* 1D example of filtering and projection of the density field  $\rho$  across two adjacent elements with boundary at x = 0. Filter radius is r = 50, with threshold value of  $\eta = 0.5$  for two values of  $\beta = 5$  and  $\beta = 15$ .

using (5.4), here shown for two projection levels  $\beta = (5, 15)$  for a threshold value of  $\eta = 0.5$ . Let the dashed lined for  $\rho$  indicate the boundary between to adjacent elements, the element at  $x \leq 0$  has density  $\rho = 0$ , eg. void, and the element at  $x \geq 0$  has density  $\rho = 1$  for some solid material. The filtered densities represent a relaxation of the optimization problem where the material boundary is smoothed to avoid discrete changes i the design variables (checker board). Hence the smoothed density is a contentious functions. The optimization is started for a low beta value, typically  $\beta = 1$ , and as the optimization iterates the value of beta is increased so that the projected densities approach 0 or 1 and the material boundary is well defined. In this way the optimization should approach a local minima as the design is optimized for a increasing value of  $\beta$ . The threshold value is constant through out the optimization and is typically set to  $\eta = 0.5$  in compliance problems as a conservative definition of when a given value of  $\tilde{\rho}$  represents the shift between the two materials.

Figure 5.2 shows the two dimensional version of the three density fields.  $\rho$  is represented by a circle. Here the filter radius is set to r = 15 and the projected density is showed for  $\beta = 5$ . At iterations where  $\beta = 1$  the projected density field is similar to the filtered density field.



*Figure 5.2:* Examples of 2D density fields. Left)  $\rho$ . Middle) Filtered density field  $\tilde{\rho}$  for r = 15. Right) Projected density field  $\tilde{\rho}$  for  $\beta = 5$  and  $\eta = 0.5$ . The colourbar indicates the density value for the given fields.

Performance of a optimized design could potentially vanish due to production tolerances which might alter important design features rendering the design useless. As a means to circumvent such situations [26] introduced robustness towards (uniform) design variations by formulating eroded and dilated versions of the original filtered densities. The original design is now referred to, as the nominal design. The optimization problem is then formulated as a min/max problem so that the worst of the eroded, dilated or nominal designs are optimized at any given iteration. Implementation of robustness ensures a minimum length scale of the design. [18] introduced robustness into acoustic optimization problems via a second filtering step where the nominal design is filtered two times for threshold values above and below the nominal threshold value,  $\eta_n$ .



*Figure 5.3:* Conceptual sketch of the robust double filter approach. Step 1-3) Standard filtering and projection using the density approach. Step 4-6) Additional steps in the double filtering robust approach. The top plot in the right column,  $\tilde{\tilde{\rho}}$ , is filtered version of the last plot in the left column,  $\tilde{\tilde{\rho}}$ .

Hence also obtaining eroded and dilated versions. Figure 5.3 shows a sketch of this approach with the steps 1-3 depicting the traditional filter and projection step. Step 4 the extra added filter step to obtain  $\tilde{\tilde{\rho}}$ . The eroded dilated designs are obtained though additional two projections steps (steps 5 and 6) with eroded and dilated threshold values,  $\eta_e$  and  $\eta_d$  respectively.  $\eta_e$  and  $\eta_d$  must then represent the span in production tolerances, and therefore have to be fitted.

### 5.2 Optimization problem

Currently in the SunTune project all physical experiments are based upon a stratified setup in which gold (Au) metal nanoparticles are places directly upon a thin film, often  $\text{Er}^{+3}$  doped TiO<sub>2</sub>, grown on a substrate typically of fused quarts, SiO<sub>2</sub>. From an upconverting perspective this setup is not ideal as much of the enhanced light from the (plasmonic)metal nanoparticles will not reach the film and thereby contribute to upconversion. However the setup is used due to the simplicity which ease prototype manufacturing and the physical experimental efforts. The end case scenario is a setup with nanoparticles embedded into the host matrix/film it self. Figure 5.4 shows sketches of the computational 2D models currently used in the topology optimization. A stratified and a embedded model. For both cases the goal is to obtain the topology of the nanoparticles in the design domain  $\Omega_d$  such that the objective function  $\Phi$  is minimized in the evaluation domain  $\Omega_e$ . Both models are excited via the background excitation method<sup>iii</sup> by an incoming plane wave propagating in the negative y-direction with the E-field polarized in the x-direction. The plane wave is incident at a normal angle. Scattering boundary conditions are used to truncate the computational domain, thus a optimized single nanoparticle/unit cell is sought for.

minimize	${}^{-\!\Phi(E({\boldsymbol{\bar{\rho}}}),{\boldsymbol{\bar{\rho}}})$	$orall ar{oldsymbol{ ho}}\in\Omega_e$	objective function		
subject to:	S(x)u(x) - f(x) = 0		state problem, eq. (3.10)	(5.5)	
	$0 \leq \rho \leq 1$ ,	$\forall  \boldsymbol{ ho} \in \Omega_d,  \boldsymbol{ ho} = 0  \forall  \boldsymbol{ ho} \notin \Omega_d$	design variable bounds.		

The optimization problem is formulated<sup>iv</sup> in (5.5) with  $\Phi$  in (5.6) formulated using equation (4.2) relating the upconversion yield to the intensity of the incoming light raised to some power,  $n_p$ ,

<sup>&</sup>lt;sup>iii</sup>Described in chapter ??

 $<sup>^{</sup>m iv}$ As a minimization of the negative of  $\Phi$ , which essentially is a maximization: min. - $\Phi o \,$  max.  $\Phi$ .



**Figure 5.4:** 2D optimization models. Left) Stratified cases with the design domain placed on top of the thin film which resides on a substrate. Right) Embedded case with the design domain embedded into the film.  $\Omega_d$  denotes the design domain, and  $\Omega_e$  denotes the evaluation domain in which the objective function is evaluated.  $\Omega_e$  is indicated by the dashed lines. Scattering boundary conditions are placed on the boundaries,  $\Gamma_{scat}$ .

representing the number of photons required to excite the upconverting ions. The intensity is of a plane wave is proportional to the electric field squared,  $I \propto |E|^2$ . The exponent factor is set to  $n_p = 1.5$ , as this is the experimental average.  $\Phi$  is normalized with respect ti the intensity of the background field, turning the objective function is a measure of upconverting performance with respect to that of the background field.

$$\Phi = \frac{\int_{\Omega_e} I^{n_p} dA}{\int_{\Omega_e} I^{n_p}_b dA} = \frac{\int_{\Omega_e} |E|^{2n_p dA}}{\int_{\Omega_e} |E_b|^{2n_p} dA}, \quad \text{for } n_p = 1.5.$$
(5.6)

The filtered densities are obtained via the Helmholtz PDE in equation (5.3) and projected via the tangent hyperbolic formulation of the Heaviside step function in equation (5.4). The continuous  $\beta$ -updating scheme used in [18] for acoustic wave problems, are also used in this work. Also the design domain dimensions are in the following fixed to (height, width) = (1000,1000)nm.

### 5.3 Material interpolation

Interpolation between materials was initial performed directly in the permittivity using the power law scheme in (5.7). However this was found inefficient due to a high sensitivity to changes in the design variables,  $\rho$ . Figure 5.5 shows real part of the permittivity in the design domain,  $\epsilon'_d$ , as a function of the density when interpolating between the air and Au. Top left plot shows  $\epsilon'_d$ for direct interpolation in the permittivity and bottom left plot shows the interpolation in the refractive indices form which then the permittivity is calculated<sup>v</sup> for three power factors p = 1, 2, 3.

$$\hat{\epsilon}_d(\rho) = \hat{\epsilon}_{air} + \rho^p(\hat{\epsilon}_{Au} - \hat{\epsilon}_{air}), \quad p = 1, 2, 3, \dots, \text{ permitivity interpolation}$$
 (5.7)

 $\hat{n}_d(\rho) = \hat{n}_{air} + \rho^p (\hat{n}_{Au} - \hat{n}_{air}) \quad p = 1, 2, 3, \dots, \text{ refractive index interpolation}$  (5.8)

<sup>&</sup>lt;sup>v</sup>Using equation (3.7)

The right column of figure 5.5 shows the corresponding reflectance<sup>vi</sup> and shows that even for increasing values of *p* high sensitivity towards changes in  $\rho$  exist using (5.7). This did cause issues in the for low  $\beta$  values. However, the reflectance curve for interpolation in  $\hat{n}$  shows smoother charterer, thus all material interpolation is conducted using (5.8).



*Figure 5.5:* Material interpolation using a power law between air and Au. Left column)  $Real(\epsilon_d)$  calculated by direct interpolation in the permittivity  $\hat{\epsilon}$  (top left) and calculated from material interpolation in  $\hat{n}$  (bottom left). Right column) Corresponding reflectance using direct interpolation in the permittivity  $\hat{\epsilon}$  (top right) or through  $\hat{n}$  (bottom right.)

### 5.4 Projection threshold

The projection threshold parameter,  $\eta$ , essentially controls the shift between materials, for e.g.  $\eta = 0.5$ , densities above this threshold,  $\rho > \eta$ , are per definitions one of the two materials and for  $\rho < \eta$  the other. At high  $\beta$ -values less intermediate densities exists as they are projected towards either 0 or 1, which essentially constrains the optimization. One strategy could then be to set  $\eta$  to favour one material over the other and another strategy could be to set  $\eta$  conservatively with projection around densities representing the average of the materials used.

Moreover for an objective function with multiple local minima the initial design used to start the optimization could potential bias the optimization towards a non-optimum minima. For objective functions with fewer local minima the initial design choice might might be less important as the optimization would converge towards the local minima minimizing  $\Phi$  the most. From experience the objective function in equation (5.6) consist many of such local minima in which the choice of initial design does influence the final outcome.

The following presents the work conducted in an effort to establish a strategy or "rule of thumb" for choosing both  $\eta$  and the initial design density, as these are prerequisites for obtaining consistent designs.

<sup>vi</sup>Calculated for normal incidences using,  $R(\rho) = \left| \frac{\hat{n}_1(\rho) - \hat{n}_2(\rho)}{\hat{n}_1(\rho) + \hat{n}_2(\rho)} \right|^2$ .

To test the influence of  $\eta$  a density scan has been conducted. Using the stratified case with air and Au as materials, the initial design<sup>vii</sup> was changed in the interval  $0.1 \le \rho_0 \le 0.9$  in steps of 0.1, with the projection threshold set to  $\eta = \rho_0$ . This scan was performed using the material interpolation in equation (5.8) for values of p = 1, 2 and 3. For all tested interpolation powers three qualitative tendencies were observed depending upon the start density,  $\rho_0$ .

- All Au is removed, leaving the design domain full of air as the optimal design.
- A "design" is obtained.
- The design domain is mainly filled with Au. Which is not optimal as such nanoparticles would block much of the upconverted light propagating back towards the solar cell.

Figure 5.6 shows the three design tendencies obtained for the scan with a interpolation power of p = 2. Table 5.1 shows the intervals of  $\rho_0$  for all three interpolations powers where a "design" were observed. In all cases for p the optimization returned a "design" for initial densities corresponding to a permittivity  $\epsilon'_d \approx 0$ .

р	$ ho_0$	$\epsilon_d'( ho_0)$		
Interpolation power	Initial density	Permittivity (real part)		
1	0.06 - 0.12	0.6 - (-0.7)		
2	0.25 - 0.36	0.4 - (-1.0)		
3	0.42 - 0.50	0.3 - (-0.8)		

**Table 5.1:** Density intervals from which a "design" were observed, with corresponding real part of the permittivity (rounded numbers) shown for interpolation powers of p = 1, 2, 3. Negative permittivity values corresponds to the high density values in the intervals. Stratified case.



*Figure 5.6:* Results from density scan of the initial design in the interval  $0.1 \le \rho_0 \le 0.9$  with projection threshold set to  $\eta = \rho_0$ . The plots shows the electric field enhancement  $|\mathbf{E}|/|\mathbf{E}_b|$ , w. dark blue areas containing Au. Top left) All Au is removed. Top right) A "design" is obtained. Bottom) Design domain mainly filled with Au.

This might indicate that setting  $\eta$  to, a value where the intermediate densities shifts between dielectric and metallic properties could be advantageous. That is where  $\epsilon'_d(\eta) = 0$ . On the other

<sup>&</sup>lt;sup>vii</sup>Referring to the initial density value of every element in the design domain  $\Omega_d$ .

hand, according to the interface conditions in equation (4.11), surface plasmon polaritons only occur if  $\epsilon_m = -\epsilon_d$ . Hence another strategy could be to set  $\eta$  such that  $\epsilon'_d(\eta) = -\epsilon_{sur}$ , where "*sur*" indicates the surrounding medium. To test these two projection threshold strategies another density scan were performed scanning for different values of the start density,  $\rho_0$  but with a fixed  $\eta$  according to the two projection thresholds strategies. This was performed for three types of dielectrics surrounding the design domain, air, Er doped TiO<sub>2</sub> and SiO<sub>2</sub> for both the stratified and the embedded case. Such that the material in the design domain can be either; air/Au, Er doped TiO<sub>2</sub>/Au or SiO<sub>2</sub>/Au. The interpolation power were fixed, at p = 3.

Table 5.2 shows the best objective function obtained for both the stratified and the embedded case when optimizing between Er doped TiO<sub>2</sub> or Au. No significant difference was found when using the two projection threshold strategies, when comparing the value of the objective function. In both the stratified and the embedded case  $\Phi$  is within the same order of magnitude when comparing between the two thresholds. The optimum is also obtained for the same initial density,  $\rho_0$ . Thus, one might chose to set the projection threshold  $\eta$ , such that,  $\epsilon'_d(\eta) = -\epsilon'_{sur}$ , to satisfy the interface condition in equation (4.11).

$ ho_0$	$\Phi$	$\Phi$	$\epsilon_d'( ho_0)$	R	$\rho_0$	Φ	Φ	$\epsilon_d'(\rho_0)$	R
	$\epsilon'_d(\eta) = 0$	$\epsilon'_d(\eta) = -\epsilon'_{sur}$			-	$\epsilon'_d(\eta) = 0$	$\epsilon'_d(\eta) = -\epsilon'_{sur}$	u ·····	
0.5	-3	-3	2	9%	0.4	-7	-6	4	2%
0.6	-5	-3	-1	23%	0.5	-4	-3	2	9%
0.7	-8	-7	-10	<b>46</b> %	0.6	-4	-4	2	23%

**Table 5.2:** Results using the two different projections for optimization between Er doped TiO<sub>2</sub> / Au with a material interpolation power of p = 3. Left) Stratified case. Right) Embedded case. In both cases  $\eta = 0.57$  for  $\epsilon'_d(\eta) = 0$  and  $\eta = 0.66$  for  $\epsilon'_d(\eta) = -\epsilon'_{sur}$ . R is reflectance. The tables shows the ranges of  $\rho_0$  where "designs" were obtained. All values are rounded.

In the stratified case best values of  $\Phi$  are obtained for values  $\rho_0$  resulting in a slightly negative  $\epsilon'_d(\rho_0)$ , i.e. when the initial design possess light metallic character. Thus, for  $\epsilon'_d(\rho_0) \approx -10$  in the left of Table 5.2. For the embedded case best values of  $\Phi$  was obtained for initial densities with light dielectric character, the opposite of the stratified case. Here  $\epsilon'_d(\rho_0) \approx 4$ . This might be due to the nature of the embedded case where the optimization potentially benefit (i.e. improving the objective function) from even small amounts of reflection/scattering, as the evaluation domain  $\Omega_e$  is placed around the design domain  $\Omega_d$ . The stratified case does not benefit from reflection/scattering unless it is directed towards the film. By looking at the reflectance, R, columns this is also evident as the reflectance is significantly lower for the  $\rho_0$  resulting in good designs in the embedded case compared to the stratified case.

Figure 5.7<sup>viii</sup> shows some of the designs (i.e. with the best cost function while not containing any Au nor only/mostly Au) from the density scan studies using the two strategies for  $\eta$ . Both strategies for  $\eta$  results in overall similar designs with comparable objective function value.

<sup>&</sup>lt;sup>viii</sup>Only the field electric field enhancement plots are show to conserve space. As these plots shows both the electrical performance while also showing the contours of the topology





For the embedded cases "resonator" like designs are obtained for all three choices of dielectric (the embedded case with  $SiO_2/Au$  is not show) which seen from an upconverting perspective is beneficial as then more  $Er^{+3}$  ions are exposed to the enhanced field. The stratified case creates designs with Au along the design domain edges letting light propagate down the film while enhancing light near the film boundary.

Based on the tendencies described above and shown in Table 5.2 a general strategy/ "rule of thumb" for choosing  $\eta$  and the initial design density could be in the form:

#### Stratified case:

- Project densities around a value of  $\rho$  resulting in  $\epsilon'_d(\eta) = -\epsilon'_{sur}$ .
- Chose a density of the initial design resulting in light metallic properties. Around  $-1 \le \epsilon'_d(\rho_0) \le -10$  seems to be sufficient when using Au as the metallic material.
- The reflectance of the initial design,  $R(\rho_0)$ , should be within 20 50% in order not to block nor reflect to much of the incoming light.

#### **Embedded case:**

- Project densities around the value of ρ resulting in ε'<sub>d</sub>(η) = -ε'<sub>sur</sub>.
- Chose a density of the initial design resulting  $\epsilon'_d(\rho_0) \approx 0$ .
- The reflectance of the initial design,  $R(\rho_0)$ , should be within 1 10%.

In af first attempt to check the generality of these guidelines the objective function is changed setting the exponent  $n_p$  in (5.6) to  $n_p = 2$ , and performing a density scan. Figure 5.8 shows the best three "designs" obtained when using  $\text{ErTiO}_2/\text{Au}$  for both cases.

Comparing these plots with the corresponding plots figure 5.7 shows very similar designs obtained for approximately the same initial design,  $\rho_0$ . There is a slight difference in which  $\rho_0$  resulting in the best  $\Phi$ , which could be due to the change of objective function as well as difference in the resolution/step in the sweeps. However, the tendencies are very similar and thus validate the guidelines. At least to a first approximation.

A general addition to the guidelines could be to always perform a density scan and hereby get a general impression of the sensitivity towards changes in initial design. This would be beneficial in cases were a widely different objective function is initially implemented. Such a scan could either be rough scan in steps of 0.1 in whole density range  $0.1 \le \rho_0 \le 0.9$  or using smaller steps in the vicinity where;  $\epsilon'_d(\rho)$  is light metallic (stratified case) and  $\epsilon'_d(\rho) \approx 0$  (embedded case). Or both.







### 5.5 Robustness

Another important factor to consider is robustness of the design towards production tolerances as previously mentioned. A mapping from a production tolerance towards the choice of filter radius and eroded and dilated threshold values would be beneficial. In an effort to obtain such a mapping the following expressions have been derived.

The double filter approach is illustrated for a 1D case in figure 5.9 for a extremely high projection level<sup>ix</sup> of  $\beta = 999$ , a filter radius of r = 50 and for three values of  $\eta = [\eta_d, \eta_n, \eta_e] = [0.3, 0.5, 0.7]$ . In this case the eroded and dilated projection levels are set symmetrical around the nominal projection,  $\pm 0.2\eta_n$ , resulting in dilated and erode designs that are r/2 bigger and -r/2 smaller than the nominal design, respectively.



*Figure 5.9:* Robust double filter approach for a filter radius of r = 50, and  $\eta = [\eta_d, \eta_n, \eta_e] = [0.3, 0.5, 0.7]$  for dilated, nominal and eroded threshold values respectively. Extreme high projection level  $\beta = 999$ . The numbers at the top and bottom of the lines indicate the x position of the eroded, nominal and dilated boundaries.

Solving the Helmholtz equation (5.3) for the above 1D case an expression for the tolerance one would obtain at high  $\beta$  values, using symmetric projection equates,

$$\Delta x = \ln(2\eta_n - 2\Delta\eta)r. \tag{5.9}$$

Where  $\Delta x$  is the obtained one dimensional tolerance, and  $\Delta \eta$  is the symmetric projection value relative to the nominal projection threshold,  $\eta_n$ . That is,  $\eta_d = \eta_n - \Delta \eta$  and  $\eta_e = \eta_n + \Delta \eta_n$ . In the above example  $\Delta \eta = 0.2$  and r = 50 which would result in  $\Delta x = \pm 1/2r$  as observed in figure 5.9. Substituting this value of  $\Delta x$  into (5.9) and solving for  $\Delta \eta$  results in

$$\Delta \eta = \eta_n - \frac{1}{2} e^{-1/2}.$$
 (5.10)

Which is an estimate of what to set  $\eta_e$  and  $\eta_d$  to in order to obtain symmetrical eroded and dilated designs within  $\pm r/2$  of the nominal design. Here then the filter radius *r* would correspond to a given machine tolerance.

Adopting the min/max formulation and thus optimizing towards improving the worst of the eroded or dilated design could ensure a minimum performance of the nominal design towards production tolerances of  $\pm r/2$  in this particular example. Another approach could be to write the objective function as the average performance of the eroded and dilated design. However (5.10) remains still to be tested.

<sup>&</sup>lt;sup>ix</sup>In order emphasize the result. Such  $\beta$ -values would not be used in practice.

# Conclusion and outlook

A 2D topology framework has been implemented using COMSOL Multiphysics, controlled and manipulated though custom Matlab scripts. The framework is currently able to optimize the topology of nanostrucures and hereby enhance the local near field in a desired domain. Two cases have been studied, a stratified and a embedded case. The former represents an initial prototype setup easing production and experimental efforts. The latter represents the end case scenario in which nanostructures are embedded directly into the host matrix containing upconverting ions. A set of generic guidelines have been formulated from qualitative studies. These will function as tool to determine the nominal projection threshold value and initial design density, in cases where either the objective function is changed and/or when changing material interpolation function.

By relating the filter radius to a symmetric production tolerance using the robust double filtering method, a expression estimating values for the eroded and dilated projection threshold values have also been obtained. It has not been tested for this specific use case, however the robust approach have previously proven to successfully introduce a minimum length scale into optimized designs. Hereby ensuring a minimum performance towards production tolerances.

The guidelines obtained will serve as foundation in the future work. Here focus will be directed towards implementing constraints imposed by Electro Beam Lithography (EBL) and nanoimprint fabrication processes. Both techniques are so called top-down methods which constrains the topology in the third dimensions, due to necessary etching steps, as well as general production inaccuracies such as proximity effects when using EBL. Such constraint implementations will initially be conducted in the presented 2D models to gain basic experience with the robust double filtering formulation and/or addition of penalty terms. However, progressing to 3D is a necessity. Mapping 3D performance of 2D optimized designs could serve as an initial step prior to conducting true and more computational demanding 3D optimization. [27] showed that a 2D optimized metal nanoparticle could be tuned towards a desired resonance wavelengths by extruding it in the third dimension.

A one month external stay, at the Danish Technical University (DTU), at the TopOpt group (dept. of mech. eng.) is scheduled in the beginning of 2017. Here focus will be to work on a 3D topology optimization framework capable of solving electromagnetic problems using the PETSc interface. Furthermore experimentation with different objective functions are also planned, as the current objective function does not necessarily guarantee neither the best numerical performance nor optimum plasmonic topology. The following lists some of the future tasks:

- Implementation of robustness using the double filter approach
- Implementation of production constraints and EBL proximity effect correction, (inverse optimization problem).
- Experimentation with different objective functions.
- Transition from 2D to 3D topology optimization.

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