SUPER-RESOLUTION AND NONLINEAR ABSORPTION
WITH METALLODIELECTRIC STACKS

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SUPER-RESOLUTION AND NONLINEAR ABSORPTION
WITH METALLODIELECTRIC STACKS

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ABSTRACT

SUPER-RESOLUTION AND NONLINEAR ABSORPTION WITH METALLODIELECTRIC STACKS

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We investigate sub-wavelength imaging, i.e. super-resolution, in metal-dielectric film systems, which are simply referred to as metallodielectrics. Our simulations incorporate experimentally derived material dielectric dispersion properties across the visible region. For demonstration purposes we designed metallodielectric stacks for super-resolution containing GaP and TiO\textsubscript{2}, dielectric films, and either Ag or Au as the metallic materials. Using the known optical properties of the constituent materials found designs that could be good candidates for super-resolution. We did not have the resources to fabricate these samples; however, based on our computer simulations we are confident that the designed samples would produce super-resolution approaching one-twentieth of a wavelength in air.
We examined for the first time the broad bandwidth of the super-resolution phenomenon in metallodielectrics. We validate the results using the finite element method (FEM) and the transfer matrix method (TMM). We also show that the measurement of super-resolution is highly dependent on the distance of the probe from the exit surface; high resolution at the exit plane can quickly decay with a few tens of nanometers when high resolution is sought.

Secondly we numerically studied the nonlinear optical transmission of an optical beam through heterogeneous metallodielectric stacks under the action of nonlinear absorption. One film layer is a metal and the other layer is a dielectric; the heterogeneous material is called a metallodielectric stack (MDS). In these studies we also used applied FEM with two-dimensional transverse effects and TMM simulation techniques. Our samples consisted of Ag/ZnS, Ag/SiO$_2$ and Cu/ZnS. We numerically simulate using two transverse dimensions in our FEM codes, Z-scan experiments for two different MDS designs and draw general observations from these cases. We experimentally examined the nonlinear absorption effect in samples of Ag/SiO$_2$ when irradiated by a femtosecond pulse width beam. There is a significant nonlinear enhancement effect observed in high transmission spectral regimes, which is attributed to field confinement in the metal layers. We showed how the nonlinear absorption varies with wavelength, which changes the field penetration within the stack layers. These results can be applied for optical limiters and switches.
DEDICATION

I dedicate this dissertation in loving memory of my Father, Joseph Yato Katte and my Mother, Mary Kemayou Katte who despite many odds gave their all, to make our home a citadel of joy, love, and a place where learning was valued.
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*Through God we shall do valiantly: for he it is that shall tread down our enemies* *(KJV)*

*Psalm 60:12*
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CHAPTER 1

METAMATERIAL OPTICS

1.1 Introduction

In the recent years, there has been a surge in the publications exploring metamaterials and their unusual optical properties. Metamaterials are artificially engineered structures with electromagnetic properties normally unattainable in nature [1], which can make them useful for designing new devices. An optical metamaterial is a composite in which sub-wavelength features and the constituent material control the macroscopic electromagnetic properties of the material [2].

Metamaterials with negative dielectric permittivity and magnetic permeability, i.e. \( \text{Re}\{\varepsilon\} < 0, \text{Re}\{\mu\} < 0 \), over a certain frequency region were studied by Veselago in 1968, since then they have attracted substantial interest because of their ability to refract light in a direction that is characterized by a negative index material (NIM). The physics of NIMs requires that the refractive index is given by the negative square root of the permittivity and permeability.
A simple simulation showing positive and negative refraction is demonstrated in Figures 1.1 and Figure 1.2 below. In both figures a plane wave is obliquely incident from the bottom on a surface. In Figure 1.1 the bottom rhombus has an index of 1.5 and the cover material has an index of 1. In Figure 1.2 the index of the cover is -1.5. Positive refraction is observed in Figure 1.1 and negative refraction in Figure 1.2. This result simply shows that NIM refractive behavior is quite different from that observed in conventional positive index materials (PIM).

Figure 1.1: Refraction at an interface between two PIM materials.
A seminal paper by Pendry appearing in 2000 ignited renewed interest in NIMs. Pendry showed that a lossless NIM is a form of flat lens that can help form super-resolved images of objects because high spatial frequency evanescent waves are effectively “amplified” in the NIM. Conventional PIM-based optical systems resolve only to about a half of the wavelength of the incident light, but super-resolution in the sense used here surpasses the diffraction limit.

The first NIMs were fabricated to operate in the microwave region by Smith and collaborators [3]. The most challenging task in designing and building the first NIMs, was achieving a negative magnetic permeability, which was initially done by designing a metallic split ring resonator (SRR) structure [3]. Obtaining a negative dielectric permittivity is simple since most naturally occurring metals have a negative dielectric permittivity at wavelengths longer than the plasma wavelength; this is a so-called
plasmonic regime. The most applicable metals for this task will be those with a low absorption such as silver, gold and aluminum.

In the same paper by Pendry he also predicted that a simple metallic slab can exhibit behavior that mimics negative refraction. Technically speaking a silver slab is not a NIM, although its dielectric permittivity is negative, $\text{Re}\{\varepsilon\} < 0$, its magnetic permeability is positive, $\text{Re}\{\mu\} > 0$. This prediction opened the way for metals which were not considered as candidate materials for transmission optics, in the past because of their opacity [1]. Pendry showed that super-resolution can be achieved with metals because the evanescent waves which carry information about sub wavelength details are “amplified” at the image plane. He calculated the super-resolution effect by considering a silver slab in his numerical calculations. With numerical simulations, Pendry showed that this silver slab can super-resolve in the near field regime at an optical wavelength of 357nm. His results also concluded that the imaginary contribution of the dielectric function limits achievable super-resolution. Thus, super-resolution was experimentally achieved using a silver slab in the near field regime by making use of a material with negative permittivity, thereby eliminating the burden of engineering a negative magnetic permeability [4]. Hence one need not employ NIMs for making a near field super-lens [1]. To mitigate excessive absorptive losses Korobkin et al. have explored using “semiconductor materials” in a plasmonic wavelength regime, i.e. $\text{Re}\{\varepsilon\} < 0$, since semiconductors have lower losses than metals [2]. Doped-semiconductors can be used at infrared wavelengths where metals are too opaque to be used for resonant tunneling. The losses are controlled by carefully monitoring the doping levels in InGaAs [2]. Metals
Despite their apparent opacity can open up new pathway for optics, if engineered thoughtfully with resonant tunneling to yield a high transmission. An alternate approach to the problem of low transmission through metals, is building a photonic band-gap material containing alternating bands of a metal and a dielectric called a metallodielectric stack (MDS). MDSs have shown very high transmittance, compared to a single metal film where it is known that visible light only propagates to a few skin depths within the metal. This peculiar characteristic is attributed to a resonance tunneling mechanism within the MDS [5]. Simulations in which the choice of the metal and dielectric, the thickness of the various bands, and the entire layout of the stack is varied have yielded very high Transmission and Resonant characteristics. These results have been confirmed by experiments [5, 6, 7].

The other unusual and interesting property of metals is their ultrafast and high optical nonlinearities, which has been the subject of recent investigations [8, 9]. If metallic layers are sandwiched within dielectric layers to yield interesting transmission and resonant characteristics of the entire MDS, these effects contribute enormously to enhance nonlinear processes.

My dissertation, addresses two applications of MDS, the linear application of super-resolution and the nonlinear application of optical limiting. We have designed, fabricated and characterized MDS consisting of just a few hundred nanometers thickness of material, yet they show high nonlinear absorption coefficients. These MDSs have been proposed for optical limiting or photonic switching applications.
1.2 Overview of Dissertation

The other sections of this chapter will be devoted to a review of related research. In Chapter 2, a theoretical study of super-resolution in MDSs is presented. MDS designs are considered that can super-resolve with relatively high through output power.

In Chapter 2 the Transfer matrix technique (TMM) and COMSOL multiphysics based finite element method (FEM) technique are compared for calculating the transmission and super-resolving properties of MDS. Chapter 3 is devoted to a theoretical study of Continuous wave (CW) propagation in MDS. In Chapter 3, the analysis is extended to include the optical nonlinearities prevalent in MDS. Here the Z-scan technique is introduced and discussed, which is the experimental technique used to determine the nonlinear properties of MDS. The results of numerical Z-scan simulations for typical, but realistic, MDS designs are shown. In Chapter 4, the fabrication and experimental characterization of MDS samples are discussed. The experimental steps leading to the determination of the nonlinearities of MDS is also addressed. Finally Chapter 5 is devoted to the conclusion of my dissertation and potential future works with MDS.

1.3 Review of Related Research

The objective of my dissertation is to investigate the super-resolving and nonlinear optical characteristics of MDS designs. With this objective in mind it will be worth investigating if our proposed device can achieve super-resolution with realistic material characteristics and the limitations of the current art of nanofabrication.
Considering the rapid growth of research in this area, because of its evident potential applications, a concise review of results and concepts are presented that have propelled research in this area. Beginning with the Maxwell-Garnett and the effective medium theories, which are used in deriving expressions for the effective permittivities and permeabilities of metamaterials. The Metal lens is discussed with consideration of surface plasmons (SP), which play an important role in the interaction of light metal-dielectric interfaces. Then some common NIM designs are discussed, heterogeneous structures and finally the chapter is concluded with a discussion on the super-resolving properties and nonlinear enhancements observed with MDS. Specific MDS designs which have been characterized for super-resolution and nonlinear absorption will be addressed in the next Chapters 2 and 3, respectively.

1.3.1 Maxwell–Garnett and Effective Medium Theories (MGT/EMT)

The MGT/EMT has been used extensively to calculate the dielectric function (permittivity/permeability) of composite materials and it can be extended by analogy to magnetic materials. The MGT which was introduced in 1904 provides an effective way in describing the effective dielectric properties of composite dielectric media [10]. Later in 1930 Bruggeman came up with a self consistent model known as the EMT.

The MGT is a homogenization technique for finding the average response of the medium due to constituents that are much smaller than a wavelength. Homogenization theories are also important for our understanding of negative refraction with applications to super-resolution [11]. It is therefore necessary that a simple foundation is laid for this
theory since it will be re-visited and applied in the several NIM schemes that we shall consider.

Most metamaterials are made of a conglomerate of constituent materials with different dielectric constants arranged randomly or in a periodic fashion. For us to study the optical characteristics of such a metamaterial it is important that we understand how to qualitatively describe its optical properties.

Let’s begin with the well-known Maxwell’s equations in matter, which are written as

\[
\begin{align*}
\nabla \cdot \mathbf{D} &= \rho , \\
\nabla \cdot \mathbf{B} &= 0 , \\
\n\nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t} , \\
\n\nabla \times \mathbf{H} &= \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t} ,
\end{align*}
\]

(1.1)

where \( \mathbf{E} \) and \( \mathbf{H} \) are the macroscopic electric and magnetic fields, \( \mathbf{D} \) is the electric displacement vector and \( \mathbf{B} \) is the macroscopic magnetic induction, \( \rho \) and \( \mathbf{J} \) are the macroscopic net charge and current densities [10].

The effective medium parameters of interest are the effective dielectric permittivity \( \varepsilon_{\text{eff}} \) and the effective magnetic permeability \( \mu_{\text{eff}} \) related to the fields as;

\[
\begin{align*}
\langle \mathbf{D} \rangle &= \varepsilon_0 \varepsilon_{\text{eff}} \langle \mathbf{E} \rangle = \varepsilon_0 \langle \mathbf{E} \rangle + \langle \mathbf{P} \rangle ; \\
\langle \mathbf{B} \rangle &= \mu_0 \mu_{\text{eff}} \langle \mathbf{H} \rangle = \mu_0 \langle \mathbf{H} \rangle + \langle \mathbf{M} \rangle .
\end{align*}
\]

(1.2)

The bulk average polarization and magnetization in the medium is

\[
\begin{align*}
\langle \mathbf{P} \rangle &= N \langle \mathbf{p} \rangle , \\
\langle \mathbf{M} \rangle &= N \langle \mathbf{m} \rangle .
\end{align*}
\]

(1.3)
Consider a solid metallic sphere of radius $R$, and dielectric permittivity $\varepsilon_m$, placed in a medium of permittivity $\varepsilon_h$ under the action of constant electric field ($E_0 = E_0 z$). Where $N$ is the relevant density of polarizable or magnetizable structural units, and $p$ and $m$ are the microscopic electric and magnetic dipole, moment that develop in a structural unit [10]. The angular brackets denote a volume average to recover the macroscopic moments.

The electrostatic potential solution for a sphere embedded in a dielectric medium can be solved using the solution of Laplace equation in spherical coordinates. The electric potential, $\phi$, within and outside the metallic sphere is given by

$$
\begin{align*}
\phi_{in}(r, \theta) &= A r \cos \theta & r \leq R \\
\phi_{out}(r, \theta) &= -E_0 r \cos \theta + \frac{B \cos \theta}{r^2} & r \geq R,
\end{align*}
$$

(1.4)

where $A$ and $B$ are constants to be determined, using the boundary conditions of electrostatics and $R$ is the radius of the sphere. The boundary conditions are the continuity of the tangential component of the electric field (This condition is same as the potential being continuous) and the normal component of the electric displacement vector.

Applying the boundary conditions at $r = R$, we have the equations:

$$
\phi_{out}(R, \theta) = \phi_{in}(R, \theta) : -E_0 R \cos \theta + \frac{B}{R^2} \cos \theta = AR \cos \theta \Rightarrow B = (A + E_0)R^3;
$$

(1.5)

and
\[
D_{\text{out}}^{\text{norm}} = D_{\text{in}}^{\text{norm}} : \varepsilon_0 \varepsilon_h (\ - \frac{\partial \phi_{\text{out}}}{\partial r} = \varepsilon_0 \varepsilon_m (\ - \frac{\partial \phi_{\text{in}}}{\partial r}) \Rightarrow \varepsilon_h (E_0 + \frac{2B}{R^3}) = -\varepsilon_m A. \quad (1.6)
\]

Eliminating the parameter \( B \), we solve for \( A \):
\[
\varepsilon_h (E_0 + 2A + 2E_0) = -\varepsilon_m A \Rightarrow A = \frac{-3\varepsilon_h E_0}{(\varepsilon_m + 2\varepsilon_h)}. \quad (1.7)
\]

The parameter \( B \) is
\[
B = \frac{(\varepsilon_m - \varepsilon_h)R^3}{(\varepsilon_m + 2\varepsilon_h)} E_0. \quad (1.8)
\]

The potential outside the sphere is
\[
\phi_{\text{out}} = -E_0 \cos \theta r + p \cos \theta r^3. \quad (1.9)
\]

The induced electric field inside the metal sphere is
\[
E_i = -\nabla \phi_i = \frac{3\varepsilon_h E_0}{\varepsilon_m + 2\varepsilon_h}. \quad (1.10)
\]

The polarization density induced within the sphere is \((\varepsilon_m - \varepsilon_h)\varepsilon_0 E_i\) and since the dipole moment of the scatterer which is the sphere is the volume integral of the polarization density, then the dipole moment of the sphere is [11]:
\[
p = \frac{(\varepsilon_m - \varepsilon_h)3\varepsilon_0 E_0 V}{(\varepsilon_m + 2\varepsilon_h)}, \quad (1.11)
\]

where \( V \) is the volume of the sphere \((V = \frac{4\pi R^3}{3})\). It follows that the polarizability \( \alpha \), of the sphere which is defined as the relation between the dipole moment and the incident field \((p = \alpha E_0)\):
\[ \alpha = 4\pi R^3 \varepsilon_0 \frac{\varepsilon_m - \varepsilon_h}{\varepsilon_m + 2\varepsilon_h}. \]  

(1.12)

At low volume fractions MGT/EMT lead to very similar results of the effective dielectric constants [10]. The polarization is given by:

\[ \mathbf{P} = N\alpha \mathbf{E}. \]  

(1.13)

To illustrate the MGT: consider a composite medium is assumed to be composed of small particles of radius \( r << \lambda \) and dielectric permittivity \( \varepsilon_i \) randomly embedded in another bulk medium with dielectric permittivity \( \varepsilon_h \). The volume filling fraction of the included particles is denoted as \( f \). If \( f \) is small, then the particles effectively do not feel the scattered fields of other particles, and one can write the dielectric permittivity for a dilute medium of small spherical particles. We define the effective dielectric constant as

\[ \varepsilon_{\text{eff}} = \varepsilon_h + 3f \varepsilon_h \left( \frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h} \right). \]  

(1.14)

For high volume fractions of spherical particles (\( f \sim 1 \)) there is an additional local field correction derived by Lorenz-Lorentz and uses a self-consistent form of the equations where the field on the applied particle is affected by an additional local field that is due to all the other dipoles in the medium.

\[ \mathbf{E} = \mathbf{E}_0 + \frac{\mathbf{P}}{3\varepsilon_0}, \]  

(1.15)
The factor of three comes from averaging over all directions. Also, the field in the definition of the dipole moment is replaced by the effective field, i.e. \( \mathbf{P} = N\alpha\mathbf{E} \). The Lorenz-Lorentz result for the polarization is

\[
\mathbf{P} = \frac{N\alpha E_0}{1 - N\alpha/3}.
\]  

(1.16)

When the concentration of the particles is dense and applying corrections to the fields according to the Lorentz-Lorentz model gives an effective dielectric permittivity which can be determined by the equation below:

\[
\frac{\varepsilon_{\text{eff}} - \varepsilon_h}{\varepsilon_{\text{eff}} + 2\varepsilon_h} = \frac{N\alpha}{3}.
\]  

(1.17)

A system composed of materials with different dielectric permittivity can be treated as an effective medium, when an electromagnetic wave propagates through it. Thus the dielectric function of the entire system can be written as a weighted average of the dielectric function its sub constituents.

The occurrence of super-resolution in MDSs was first predicted with a condition derived from the effective medium theory known as permittivity matching, that is when the permittivity of the metal is approximately equal to the negative of the permittivity of the dielectric. The calculation of the effective medium parameter based on MGT/EMT for a system with spherical particles generally includes Mie Scattering coefficients, as result of scattering of the electromagnetic fields by these particles. In the case of planar layers as considered in our work. The effective index of a metallodielectric is calculated as follows.
**Case 1** If the electric field is applied parallel to the interfaces and it’s continuous across the layer, then;

\[ E_1 = E_2 = \langle E_{\parallel} \rangle, \]

\[ \langle D_{\parallel} \rangle = \frac{(d_1D_1 + d_2D_2)}{(d_1 + d_2)} = \varepsilon_0 \frac{\varepsilon_1d_1 + \varepsilon_2d_2}{d_1 + d_2} \langle E_{\parallel} \rangle. \]

The parallel permittivity is

\[ \varepsilon_{\parallel} = \frac{\varepsilon_1d_1 + \varepsilon_2d_2}{d_1 + d_2}, \quad (1.18) \]

where \( d_1, d_2 \) is the thickness of the metal and dielectric respectively, and \( \varepsilon_1, \varepsilon_2 \) are the dielectric constants of the metal and the dielectric respectively.

**Case 2**: If the electric field is applied perpendicular to the interfaces, and its continuous across the layer, then;

\[ D_1 = D_2 = \langle D_{\perp} \rangle, \]

\[ \langle E_{\perp} \rangle = \frac{E_1d_1 + E_2d_2}{d_1 + d_2} = \frac{1}{\varepsilon_0} \frac{\varepsilon_1d_1 + \varepsilon_2d_2}{d_1 + d_2} \langle D_{\perp} \rangle, \]

\[ \varepsilon_{\perp} = \frac{\varepsilon_1\varepsilon_2(d_1 + d_2)}{d_1\varepsilon_2 + d_2\varepsilon_1}. \]

In a compact form the perpendicular permittivity is

\[ \frac{1}{\varepsilon_{\perp}} = \frac{d_1}{\varepsilon_1} \frac{1}{d_1 + d_2} + \frac{1}{\varepsilon_2} \frac{d_2}{d_1 + d_2}. \quad (1.19) \]
It is worth mentioning that this layering gives rise to anisotropic response parallel and perpendicular to the layers. This follows by setting $\varepsilon_\perp = \infty$ of Eq.(1.19) to zero while assuming that $d_1 \approx d_2$. Super-resolution has been reported in schemes designed to meet this criterion [1] and in other schemes [5, 6], such as the schemes proposed later in this treatise. In the derivation of the MGT and the EMT it is assumed that the composite material consists of grains that are much smaller than the wavelength of light but large enough, so that they can be characterized by macroscopic dielectric constants [12]. In the EMT approximation the two components making up the metal – dielectric system or stack are treated in an equivalent manner. Grains of the metal and grains of the dielectric are assumed to be embedded in an effective medium whose dielectric constant is $\varepsilon_{\text{eff}}$ the same as that of composite material. In the MGT approximation it is assumed that the grains of one component are embedded in the matrix of the other component. [12].

Since I will consider MDS designs for this dissertation, the EMT approximations are more appropriate to compare with experiments for this our case. The MGT theory is appropriate for proposed experiments that involve metal nanoparticles embedded in liquid crystal media.

1.3.2 Metal Lens

Near-field imaging as considered by Pendry with a 40nm thick Silver lens is possible because of the reduced losses for the evanescent waves [4]. The final result in his initial work on NIM is that with this new lens both propagating and evanescent waves contribute to the resolution of the image. Therefore there is no physical obstacle to
reconstruction of the image beyond practical limitations of the apertures and the flatness of the “lens” surface. Pendry concluded that by using a layer of silver sub-wavelength features could be resolved. He illustrated the result using two slits. In the absence of the silver lens, the image was blurred at a distance $z = 80 \text{nm}$ away from the object. So the two slits can no longer be resolved because the higher order (evanescent) Fourier components were reduced in amplitude. When he used the silver slab of thickness $d = 40 \text{nm}$ as a lens, with an approximate dielectric function for silver the two slits were well resolved at 80 nm from the object plane. Figure 1.3 below shows the cross section power plot across the exit surface of the silver lens described above. We notice that the two slits which are 50 nm are resolved with a good contrast. The incident wavelength of light was 357 nm. This calculation was done with COMSOL multiphysics software which solves Maxwell’s equations by through the Finite element method (FEM).
Figure 1.3: Shows the cross section power at the image plane 40 nm away from the exit surface of the silver lens.

1.3.3 Surface Plasmons

Surface plasmons (SPs) are electron oscillations at the interface of dielectric and metal. Metals because of their negative dielectric permittivity can support these surface electromagnetic modes which decay exponentially inside the bulk medium of the metal. SP modes exist for transverse magnetic (TM) polarization at the surface and the transverse electric (TE) polarization is not supported. In [4] Pendry linked the focusing action of his NIM lens to the existence of surface plasmons, when $\Re \{\varepsilon\} = -1$. Pendry calculated the Transmittance and reflectance of a NIM for both polarizations TE and TM. The calculation of the Transmittance proved that if $\varepsilon = -1$ and $\mu = -1$, then the NIM slab will “amplify” evanescent waves. The term amplify as used in literature is actually be misleading. Giving the impression that gain is introduced to the system. Actually in a
lossless medium there is a buildup of the evanescent wave as energy is stored; this is the same as a cavity mechanism. Hence for a Pendry lens both propagating and evanescent waves contribute to the resolution of the image. Even though there are no natural materials with $\mu < 0$, yet Pendry suggested that with recent developments in technology such a material could be fabricated, based on a structure with loops of conducting wire, where the permeability $\mu$ is given by $\mu(\omega) \approx 1 - \frac{\omega^2_{mp}}{\omega^2}$, where $\omega_{mp}$ is the magnetic plasma frequency. Also because $\varepsilon = -1$ is also the condition needed for the excitation of a Surface Plasmon (SP) (electromagnetically driven electron oscillations at the interface of dielectric-metals) the amplification of the evanescent waves was naturally attributed to the excitation of SP. It is believed that the plasmon resonance is responsible for building up electromagnetic energy, which enables the exponential growth of evanescent components [10]. Furthermore in Zhang’s demonstration of a silver lens in [11], he attributed the amplification of evanescent waves responsible for super-resolution to the excitation of SP. Therefore SP modes are crucial for the performance of a super-lens [10]. They usually do not propagate for very long distances, but can amplify electromagnetic interactions significantly.

The plasmon modes also turn out to be crucial for the performance of a super-lens that we will consider. [10]. Here to understand the results of subsequent experiments a short derivation of the SP mode at the interface ($z=0$) between a metal and dielectric is given. Light of frequency $\omega$ is incident from a medium with permittivity and permeability ($\varepsilon_1, \mu_1$) to a medium with permittivity and permeability ($\varepsilon_2, \mu_2$). The
transverse magnetic field is in the y direction and the propagation direction is in the z
direction. The expression for the magnetic field is:

\[
H_y = A e^{i k_x x} \begin{cases} 
  e^{-\gamma z} & z > 0 \\
  e^{\gamma z} & z < 0 \end{cases}.
\] (1.20)

The solution of the above equation requires that the permittivities of the two media
have opposite signs. We can therefore write the dispersion relation for TM mode as

\[
k_x = \left[ \frac{\varepsilon_1 \varepsilon_2 (\varepsilon_2 \mu_1 - \varepsilon_1 \mu_2)}{\varepsilon_2^2 - \varepsilon_1^2} \right]^{1/2} \frac{\omega}{c},
\] (1.21)

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

If medium 2 is a metal as \(\varepsilon_2 \approx -\varepsilon_1\) and the magnetic permeabilities are equal to the
vacuum value, then the expression above reduces to:

\[
k_x = \left( \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \right)^{1/2} \frac{\omega}{c} = k_{xr} + i k_{xc}.
\] (1.22)

The propagation length of the surface plasmon is \(l_{sp} = 1/2 k_{xc} \).
For a waveguide with metal boundaries the dispersion relation is more complicated and has anomalous dispersion frequency regions. The polarization choice for conventional plasmonic excitation is the TM mode. Lezec et al. used a prism shaped Au-
Si3N4-Ag waveguide with variable dielectric core thickness and edge angle to display negative refraction as seen in Figure 1.4. This negative refraction is attributed to plasmon modes which are excited on the leading edge of the prism. They later propagate through the prism and impinge on the slanted interface of the prism, and are refracted at a negative angle as seen in Figure 1.4. Non-resonant negative index behavior was achieved for visible incident wavelengths 476-514nm as the thickness of the dielectric layer was varied from 150nm to 50nm [13].
1.3.4 Photonic Crystals and Negative Refraction

This section briefly treats the achievement of negative refraction with 2D and 3D photonic crystal (PhC), which is still another legitimate route to achieve negative refraction. Negative refraction with PhC is fundamentally different from the other metamaterials mentioned in this work in that, the other metamaterials are composed of unit cells that are small compared to the operating wavelength and can be characterized by effective constitutive parameters, photonic crystals are based on the complex superposition of multiple Bragg scattering due to periodicity and a unit cell on the other of the operating wavelength [10]. A major advantage of these crystals over metallic systems is that these PhC can be made of dielectric materials whose absorption is very low. The 1D photonic crystal possess a photonic band gap for any given direction of propagation of light, yet these band gaps occur at different frequencies and there is no frequency region where the propagation of optical waves is completely stopped for all directions. In order to stop propagation at a given frequency for all directions, a 2D or 3D periodic variation of index may be needed [10].

To obtain negative refraction with 2D and 3D PhC, iso-frequency surfaces are needed for the photonic crystal, that are both convex and larger than those in the incident medium at the operating frequency. The crystal must be cut in order to allow phase matching from the incident medium. The iso-frequency contour of the PhC and that of the incident medium should phase match at the given frequency [10]. An illustration of negative refraction with 2D PhC is shown in Figure 1.5 below.
Prather et al have also reported negative refraction and super-resolution in the microwave with the usage of 3D photonic Crystal. The 3D photonic crystal was fabricated using a micro-milling machine. It had a body center cubic structure and was made out a material with a permittivity of 2.5. The photonic crystal was irradiated by an incident wave of wavelength 18.3mm generated by a horn antenna, placed very close to the surface of the crystal. The resolution of two pin holes by the 3D photonic crystal is shown in Figure 1.6. The pinholes have the same diameter and are separated by a distance 10 mm [14].

Figure 1.5: Correspondence between the spectral domain (Left) and the spatial domain (Right) for two crystal orientations and illustration of the associated phase matching [10].

Figure 1.6: (Left) The structure of the 3D photonic crystal. (Right): The surface intensity plot showing the two pinholes 10 mm apart. The intensity along the horizontal axis through the two pinholes is shown by the blue curve [14].
1.3.5 Metallodielectric Stacks for Super-resolution

An early attempt to confirm Pendry’s theories was accomplished by Zhang et al when they demonstrated super-resolution using a silver film, and compared the silver layer design with a purely polymer, Polymethylmethacrylate (PMMA) layer design. The entire MD Stack structure is illustrated in Figure 1.7 (left) below.

Figure 1.7 (Left) Schematic configurations for the silver super-lens experiment. The Silver layer is sandwiched between a photo resist (PR) layer and a PMMA layer, the super-lens image was revealed by means of photolithography at wavelength $\lambda = 365\text{nm}$. (Right): The imaging of a two-dimensional object ‘NANO’. Blue line shows the cross section profile when silver is used as the super lens. Red line shows the cross section profile when the silver is replaced with PMMA [15].

The design of the super lens is seen in Figure 1.7 (Right) above, consisted of sub wavelength objects milled in a Chromium (Cr) film by focused ion beam (FIB). The word ‘NANO’ was written on the Cr layer. This Cr layer was deposited on a quartz substrate by electron beam evaporation. A planarized PMMA layer was built above the Cr layer, by successive spin coating and an etching process. A silver layer was then deposited by electron beam evaporation on the planarized PMMA layer. The silver
surface was finally coated with a negative Photoresist (PR) as seen in Figure 1.7 (Right) [15]

At 365 nm which was the operating wavelength of the super lens the dielectric permittivity of Silver (Ag) \( \varepsilon_m = -2.4012 + i0.2488 \) and that of PMMA \( \varepsilon_d = 2.301 \) closely matched. In this work it was found that the enhancement of the fields was strongly dependent on the thickness of Ag and PMMA layers. The transmittivity of the TM-polarized waves was computed, while varying both silver and PMMA thicknesses. The silver thickness was varied from 15 to 55nm while the PMMA layer thickness was held fixed at 40 nm, the 35nm slab of silver gave the optimum transfer function with a resolution limit up to \( 4k_0 \). Thinner silver slabs 15nm and 25 nm showed higher but narrow enhancement bands. Thicker silver slabs 45 to 55 nm showed smaller enhancements in reference to zero-order transmission, which results in low image contrast of sub-diffraction–limited features [15]. This super-resolution optimization result has been very useful in my design studies of a MDS.

Scalora et al numerically demonstrated negative refraction in the visible spectrum by the use of metal dielectric photonic band gap structure [5, 6, and 7]. The physical interpretation of the results in these works is different from Zhang’s paper. Zhang attributed the super-resolution result as arising from the buildup of evanescent waves by excitation of surface plasmons on the surface of metal. Scalora et al. results revealed that the main physical mechanism for sub-wavelength focusing comes as result of resonance tunneling, field localization, and propagation effects within the MDS. This work also
concludes that negative refraction and superlensing is as a result of the propagating modes within the MDS instead of evanescent modes as in preceding works.

In the research presented in this dissertation, the validity of the role of resonance tunneling, field localization, and wave guiding within the MDS is numerically validated. Another distinguishing conclusion about the work by Scalora et al is the conclusion that surface plasmons do not play a significant role in the superlensing task. Thus super-resolution is attributed to a wave guiding phenomenon, which makes it possible for light to remain confined within a small fraction of a wavelength. In a previous work in reference [5], not devoted to super-resolution, it was found that a stack of $Ag/MgF_2$ containing a total of 150 nm of silver had a peak transmittance of 52% at 532nm. Such a result points to the fact that high transmittance can be achieved even with the usage of several metallic thin films.

Scalora et al coined the term “transparent metal” to describe such MDS with relatively high transmittance over a large broadband. These simulations led to the conclusion that a MDS could be transparent in the visible. They were also able to resolve two 50 nm slits sources with a transparent metal MDS 50 nm away from the MDS with a contrast of about 95% at a source wavelength of 532 nm. These results led to the conclusion that a MDS could be used to super-resolve fine features. When considering the MDS, we should see that the metal provides the negative refraction as predicted by theoretical calculations, while the dielectric is responsible for normal refraction. Several results have been previously published on this topic [6, 7].
1.3.6 Nonlinear Response of Metallodielectric Stacks

Metallodielectric (MDS) which would find potential applications in biomedical sensing and imaging because of their super-resolving properties, have also been shown to possess high third order nonlinearity, particularly because of the high $\chi^{(3)}$ value of the metal layers. These MDS could be important in the design of optical switches and limiters [8, 9].

The nonlinear response of metallic systems has been studied widely particularly metal nanoparticle systems dispersed in a solid or liquid matrix. In such systems the nonlinear optical response is enhanced by surface plasmons. Even though this beneficial, for increasing the total nonlinear optical response, yet the intrinsic nonlinear response of the metal remains hidden. This is why the thin film approach discussed in this dissertation a more qualitative and useful means of accessing the nonlinear characteristics of metals [8].

Boyd’s group showed with picosecond pulses how the nonlinear response of a Cu based MDS is enhanced compared to a single layer as shown in Figure 1.8 below. They attributed this nonlinear enhancement to a Fermi smearing process. The MDS considered in this work was made up five periods of Cu (16nm)|SiO$_2$ (98nm), and an open aperture Z scan experiment was performed at 650nm around the maximum of its transmission spectrum.
Another important result, which identifies nonlinear optical response of MDS, is reported by Lippitz et al in [9]. They studied two Ag based MDS designs. The two samples considered were:

i) 2 periods of Ag(30nm)|MgF$_2$(225nm) (single cavity);

ii) 3 periods of Ag (20nm)|MgF$_2$(215nm) (double cavity).

Even though they performed Z-scan measurements for these samples, they were concerned with the thermal issues associated with Z-scan measurements, since they were using a MHz repetition rate laser which produced picosecond pulses. The Z-scan results for nonlinear absorption measurements for double cavity stack is shown in Figure 1.9 below. In order to separate the electronic from thermal contributions; they performed pump-probe measurements. They also described the heating process taking place in the MDS with a two temperature model (TTM) and solved Maxwell’s equation’s including thermal modifications of the dielectric permittivity in the silver layer. The transmission spectra obtained showed significant agreement with the experimental measurements.
Figure 1.9: Open aperture Z-scan measurement of the double cavity sample at a pump wavelength of 825nm. The sample was measured with continuous wave and a pulsed laser beam with the same average power (20mW) but peak powers differing by about five orders of magnitude. The inset shows calculated differential transmission due to thermal effects [9].

Experimental investigations of some metallodielectrics with Cu, Au and Ag have reported significant nonlinear enhancements. The origin and contribution of all the nonlinearities is still not fully understood, yet it is agreed that an electron thermalization process which changes the Fermi Dirac distribution is a significant contributor to observed nonlinear changes. This process is sometimes explained as $\chi^{(1)}$ or $\chi^{(3)}$ process [8, 9]. In this dissertation we have studied both numerically and experimentally the nonlinearities prevalent in MDS with the Z-scan technique.
1.4 Chapter Summary

So far we have seen how metallodielectric stacks have been used to super-resolve sub-wavelength structures. We will proceed in our work to simulate and demonstrate the super-resolving properties of MDS with use of high index dielectric in the visible region (This region is usually not very convenient since the metals are very lossy). The optical properties of the metals are very important for our study; we have used optical constants of metals given in literature [16], for our simulations. With proper design we will show how optimized super-resolving MDSs can function over a wide range of wavelengths. We will report how the finite thickness of high index dielectric materials is critical for the overall super-resolving performance of the MDS.

We have also presented selected experimental results for metallic systems which show significant nonlinear enhancements under ultrafast excitation. A recent result, with Silver (Ag) showed how its optical properties vary in a nonlinear fashion with temperature [17]. The cause of this nonlinearity may be attributed to a combination of electronic and thermal mechanisms. These intriguing results for metals motivated our present research.
CHAPTER 2

DESIGNING SUPER-RESOLVING METALLODIELECTRIC STACKS (MDS)

2.1 Introduction

In this chapter, we apply numerical techniques to study the channeling and super-resolving properties of MDS. The metals considered in this chapter are Ag and Au, because of their low loss, while the dielectrics are GaP and TiO\(_2\) because of their high refractive indices. We have designed several MDS composed of these materials. We have calculated the transmission properties of the MDS with the transfer matrix technique and finite element method (FEM). We have also performed TMM and FEM simulations for sub wavelength imaging with COMSOL multiphysics. To optimize the MDS designs for super-resolution, the Bloch wave theorem was used to calculate thickness dielectric layers within the stack. In this chapter we have demonstrated the possibility of high transmittance super-resolution at the visible wavelengths. The MDS contains metallic and dielectric layers whose optical constants have been measured at our fabrication facility.
We also compared the two main designs based upon combinations of Ag/GaP and Au/GaP, and calculate the super-resolving band widths. The super-resolving bandwidth of the Ag/GaP design is (520nm-560nm), while that of the Au/GaP design is (620nm-660nm). We compare these two designs, when used in resolving two 20nm wide apertures separated by a center to center distance of 80nm. We have also compared two numerical techniques, used to study these systems, namely the transfer matrix method (TMM) and the finite element method (FEM).

2.1.1 Optical Response of Metals (Ag, Au)

The metals considered in this work are sputtered metals, which usually show slightly different optical properties from bulk metals. The optical properties are also dependent on the fabrication conditions. Silver (Ag) often stands out as an attractive candidate for super-resolution essentially because it has a lower absorption than most other metals. For super-resolution we ideally seek a metal that will transmit as much light as possible. In our work the dielectric constants for Ag and Au are taken from [16]. The values of the dielectric constant of Ag, significantly matches the values of films with thickness > 20 nm fabricated at the Nanofabrication lab of the University of Dayton. In Figures 2.1 below are plots of the refractive index and the extinction coefficient of Ag and Au which are the two metallic candidates for our super-resolution task.
We note that the refractive index of Ag remains small all through our spectral region of interest. The value is typically between 0.1-0.16, while its extinction coefficient which is responsible for absorption and negative permittivity grows steadily across the visible to near R region shown in Figure 2.1. The extinction coefficient of Au also grows steadily across the spectrum while its refractive index, drops for wavelength values less than 650nm and then rises slightly again for higher wavelengths.

2.1.2 Optical Response of Dielectric (GaP, TiO$_2$)

Gallium phosphide (GaP) is a high index (n=3.3) semiconductor material with an indirect band gap of 2.26 eV. The polycrystalline material has the appearance of pale orange pieces. Titanium dioxide occurring in three forms (Rutile, Anatase and Brookite)
is a very useful material because of its brightness and very high refractive index ($n = 2.7$), in which it is surpassed only by a few other materials [16, 18-19].

![Figure 2.2: (Left) Refractive index of GaP and TiO$_2$ as a function of wavelength. (Right) Extinction Coefficient of GaP and TiO$_2$ as function of wavelength.](image)

2.1.3 **Optical Super-resolution and Super-guiding with MDS**

Scalora et al demonstrated that super-resolution is possible with MDS because of successive momentum positive refraction at the dielectric boundary and negative momentum refraction at the metallic boundaries. When the vector sum of all the momentum vectors within the respective layers, doesn’t diverge, from the initial direction of the propagation vector, then diffraction is suppressed. This finally leads to super-guiding, channeling, sub wavelength resolution and ultimately super-resolution of sub wavelength features [5,6].

A Fourier decomposed electric field can be represented by Eq (2.1) below

$$E(x, y, z; t) = \frac{1}{4\pi^2} \iint dk_x dk_y E_t(k_x, k_y)\exp \left[i(k_x x + k_y y + k_z z - \omega t)\right]. \quad (2.1)$$
$k_x$ and $k_y$ represent the Fourier components of the spatial variation in the source in x and y respectively. The spatial variation of the fields on the object plane can be thought of as arising from the superposition of periodic functions with different periodicities. Each spatial frequency represents a periodicity $\Delta_{x,y} = 2\pi/k_{x,y}$ of variation of the electromagnetic fields on the source plane. If we consider the case of an isolated molecule on a surface emitting radiation in which case the $\Delta_{x,y}$ can literally be on atomic length scales [10].

The corresponding transverse wave vectors $k_x$ and $k_y$ are very large, and the waves are highly evanescent ($k_x > k_0, k_y > k_0$) these evanescent waves decay in amplitude away from the source [10]. So we see that sub wavelength features in the object plane will result to large k vectors propagated as evanescent waves which will be lost at the image plane in most conventional optical systems. The MDS design functions by engineering all the k-vectors to limit diffraction and capture sub wavelength details of an object.

2.2 Transfer Matrix Method (TMM)
2.2.1 TMM Plane Wave Technique

The TMM technique has been used extensively in this work to calculate the transmission properties of MDS. The TMM technique is a reliable technique used to study the propagation of electromagnetic waves in thin films. The technique used in this case is based on Maxwell equations. Given that the tangential component of the Electric field $\mathbf{E}$ and the tangential component of the Magnetic field $\mathbf{H}$ are continuous at all boundary layers.

Two by two matrices called transfer matrices can be written which describe the propagation of the electromagnetic waves within each layer. The wave vector is dependent on the index of refraction of the layer so it is going to change as we go from one layer to another. Another quantity which should be handled properly in the simulations is the phase change.

Our TMM technique was written as Matlab code and was used to study different configurations and layout of MD layers. We were also able to use our program to check the results obtained in [5, 6, and 7] and found that it the results did matched significantly. I have performed numerical computations with the TMM technique to validate that result.

The formulation of TMM technique is given below:

Consider a field incident from medium 1 to medium 2 as shown below. This derivation is based on [20, 21] describing the field in the $\alpha^{th}$ medium as a linear combination of plane waves:
\[ E = (A_a \exp(-i \beta_a z) + B_a \exp(i \beta_a z)) \exp(-i \kappa x) \] ,

(2.2)

where \( A_a \) is the amplitude of the forward travelling waves along (along +z direction), \( B_a \) is the amplitude of the backward travelling wave (along –z direction) \( \kappa \) is the x component of the wave vector, \( \beta_a \) is the z component of the wave vector.

Based on the definition of Fresnel reflection and transmission coefficient one can write the following equations below, where \( r \) represents the reflection coefficient, and \( t \) represents the transmission amplitudes from medium 1 to medium 2 (or from medium 2 to medium 1):

\[
\begin{align*}
\begin{cases}
\frac{n_1 \cos \theta_1 - n_2 \cos \theta_2}{n_1 \cos \theta_1 + n_2 \cos \theta_2} & \text{(TE wave)} \\
\frac{n_1 \cos \theta_2 - n_2 \cos \theta_1}{n_1 \cos \theta_2 + n_2 \cos \theta_1} & \text{(TM wave)}
\end{cases} & \\
\end{align*}
\]

\[
\begin{align*}
\begin{cases}
\frac{2n_1 \cos \theta_1}{n_1 \cos \theta_1 + n_2 \cos \theta_2} & \text{(TE wave)} \\
\frac{2n_1 \cos \theta_1}{n_1 \cos \theta_2 + n_2 \cos \theta_1} & \text{(TM wave)}
\end{cases} & \\
\end{align*}
\]

\[
t_{12} = t_{21} ; \quad t_{12}t_{21} - r_{12}r_{21} = 1 ; \quad r_{12} = -r_{12} ;
\]
and the coefficients of the field are

\[
B_1 = A_1 r_{12} + B_2 t_{21} , \\
A_2 = A_1 t_{12} + B_2 r_{21} , \\
A_1 = \frac{1}{t_{12}} A_2 + \frac{r_{12}}{t_{12}} B_2 , \\
B_1 = \frac{r_{12}}{t_{12}} A_2 + \frac{1}{t_{12}} B_2 .
\]

In matrix notation we can therefore write the above equation as

\[
\begin{bmatrix}
A_1 \\
B_1
\end{bmatrix} = \frac{1}{t_{12}} \begin{bmatrix}
1 & r_{12} \\
r_{12} & 1
\end{bmatrix} \begin{bmatrix}
A_2 \\
B_2
\end{bmatrix} = T \begin{bmatrix}
A_2 \\
B_2
\end{bmatrix} .
\]

\[ (2.3) \]

\( T \) denotes the transition matrix between layers 1 and 2. After the incident wave makes the transition to layer 2 it later picks up a phase as it propagates through layer 2. This can also be represented as matrix equation

\[
\begin{bmatrix}
A \\
B
\end{bmatrix} = \begin{bmatrix}
\exp(i\phi) & 0 \\
0 & \exp(-i\phi)
\end{bmatrix} \begin{bmatrix}
A' \\
B'
\end{bmatrix} = P \begin{bmatrix}
A' \\
B'
\end{bmatrix} ,
\]

\[ (2.4) \]

where \( A \) and \( B \) are the field amplitudes on the left medium; \( A' \) and \( B' \) are the field amplitude on the right end of the medium, \( P \) is the propagation matrix. \( \phi \) is the phase given by \( \phi = \frac{2\pi m_i d_i \cos(\theta)}{\lambda} \). The angle \( \theta \) is the angle between the normal vector at the boundary of medium, and the wave-vector. \( n_i \) is the refractive index of the layer, and \( d_i \) is its thickness. We see that the product of a transition matrix \( T \) and propagation matrix
$P$ results in a transfer matrix $F$. If we consider light incident from a region with refractive index $n_0$ unto an $m^{th}$ layered MDS, the light exits the MDS transiting it a region of refractive index given by $n_{m+1}$; then the resultant transfer matrix is given by

$$F = T_1 P_1 T_2 P_2 T_3 P_3 \cdots T_{m-1} P_m T_{m+1},$$

(2.5)

where $T_1$ is the transition matrix from incident medium unto layer 1, and $P_1$ is the propagation matrix within layer 1. The other $T$ and $P$ matrices are defined in a similar fashion.

$F$ can then be calculated for any number of layers, and will result in a matrix as shown below

$$F = \begin{bmatrix} f_{11} & f_{12} \\ f_{21} & f_{22} \end{bmatrix}.$$  

(2.6)

The Reflectance and Transmittance respectively are given by

$$R = \left| \frac{f_{21}}{f_{11}} \right|^2, T = \left| \frac{1}{f_{11}} \right|^2.$$  

(2.7)

In our study we have used a TMM codes to calculate the transmittance of various layouts of MDSs. The TMM technique is computationally economical compared to a full vector simulation such as the FEM [22]. The TMM is limited by the fact that the spatial variations at the interface cannot be included.
2.2.2 TMM Technique for Super-Gaussians (SG)

The TMM discussed in the previous section is implemented with an initial plane wave which is described by its electric field. This description is sufficient enough, to enable us to calculate the Transmission properties of a MDS. What about the super-resolving capabilities of MDSs, can these be easily studied using a plane wave only description of electromagnetic field through a Stack? Yes of course, but there are some obstacles, since we are interested in looking at the transverse profile of the output beam, which has been affected by the MDS. We would need a transform technique such as the Fourier transform in order to see spatial characteristics of the beam so it is logical to make the input beam a Gaussian, because it’s Fourier transform remains a Gaussian. To check for Super-resolution we would have to bring two Gaussians of specified waists close to each other and observe to see what happens after they propagate through the MDS with diffraction working to prevent super-resolution. Because the type of Gaussians used in this study have very small nanometer size spot size which is far from real world Gaussian beams, we coin the term “Super-Gaussians” to describe these beams.

Consider two Gaussian fields separated by a distance $s$ as shown below

$$E_{inc} = E_1(y - s/2) + E_2(y + s/2)$$, where $E_1$ and $E_2$ are Gaussians of form:

$$E_0 \exp \left[ -y^2 \left( \frac{1}{w^2} + \frac{ik_0}{2R} \right) \right] \exp(-k_z z) .$$  \hspace{1cm} (2.8)

The Fourier Transform of the incident field can be analytically performed and the output field is just the product of incident field and the transmission coefficient, which is
calculated by the TMM method. The TMM technique calculates transmission and reflection coefficients between materials interfaces in the MDS. A detailed description of the Fourier decomposed TMM technique for Gaussian beams can be seen [23].

2.2.3 Bloch Wave Analysis

The Bloch wave analysis is an important analysis for the characterization of the super-resolving properties of a MDS. Assume a two layer MDS, stacked along the z direction. The thickness of the dielectric layer is \( d_1 \) and the thickness of the metallic layer is \( d_2 \). The total thickness of the MDS is \( d = d_1 + d_2 \). The metal and the dielectric are all have a permeability of 1. The propagation mode within the MDS are Bloch waves whose phase accumulation is given by \( K_B(k_x) \) where \( K_x \) is the Bloch wave that is function of the transverse spatial components. A diffraction relation can be derived according to the set up of equations shown in Appendix C. The diffraction relation below is used to plot Isofrequency curves which will indicate the suppression or the enhancement of diffraction within the MDS. This diffraction relation is derived with a similar approach as those discussed in above for the TMM method.

\[
\begin{align*}
\cos(K_z d) &= \cos(\alpha_1 d_1) \cos(\alpha_2 d_2) + \frac{\epsilon_2^2 \alpha_1^2 + \epsilon_1^2 \alpha_2^2}{2 \alpha_1 \alpha_2 \varepsilon_1 \varepsilon_2} \sin(\alpha_1 d_1) \sin(\alpha_2 d_2) \cos(K_B d) = \\
\cosh(\alpha_1 d_1) \cosh(\alpha_2 d_2) + \frac{\varepsilon_2^2 \alpha_1^2 + \varepsilon_1^2 \alpha_2^2}{2 \alpha_1 \alpha_2 \varepsilon_1 \varepsilon_2} \sinh(\alpha_1 d_1) \sinh(\alpha_2 d_2),
\end{align*}
\]

where \( \alpha_i^2 = k_x^2 - (\omega/c)^2 \varepsilon_i \mu_i \) \((i=1, 2)\). The thickness of the metal and dielectric are \( d_1 \) and \( d_2 \) respectively, \( d = d_1 + d_2 \). For the MDS, the Bloch modes represent propagating waves when \( \alpha_i^2 < 0 \) and evanescent waves when \( \alpha_i^2 > 0 \). A flat diffraction curve, when
$K_z$ is plotted versus $k_x$ would indicate, that the phase accumulation upon propagation has the same rate for different spatial frequencies, hence diffraction is suppressed. While a rapidly changing slope of the curve would indicate strong diffraction [24].

Consider 2 MDS built from metals Ag or Au, and dielectric GaP. The layers distributed as follows: [(GaP (30nm)/Au (20nm)] and [GaP (30nm)/Ag (20nm)]. The incident wavelength of propagation is 640nm. If we plot the dispersion curve based on the Bloch wave calculations we obtain the result below for both MDSs. We see that the bold line representing the GaP/Au MDS is flatter than the dash curve, representing the GaP/Ag MDS. This is as a result of stronger diffraction in the GaP/Ag case.

![Dispersion at 640nm](image)

Figure 2.3: Bloch dispersion curve for [GaP (30nm)/Ag20nm)] and [GaP (30nm)/Au (20nm)]. The incident wavelength is 640nm. This result alludes to the fact that at 640nm, the [GaP (30nm)/Au (20nm)] design will resolve better than [GaP (30nm)/Ag (20nm)] design.
2.3 Finite Element Method (FEM)

The Finite Element Method (FEM) approximates a PDE problem with a problem that has a finite number of unknown parameters, that is, a discretization of the original problem. This concept introduces finite elements, or shape functions, that describe the possible forms of the approximate solution.

This discretization is done with the introduction of mesh elements, which are sub-domain or regions for which you seek solution for the particular partial differential equation (PDE). The PDE of interest in this dissertation is the Maxwell’s wave equation in TE or TM polarization. Once the meshing is done, then an approximate numerical solution of the equation is designated for each mesh element. This solution is dependent on the boundary conditions. The boundary conditions determine how the approximate solutions are established. The incident field is often known and it is represented with its parameter as an E-field, H-field or as an exact value scattering boundary condition (SBC). Other important boundary conditions are perfect electric conductor (PEC) this is good to represent reflecting surface where there is no electric field within the material. There is also the Perfect Magnetic Conductor (PMC) boundary condition, which excludes the penetration of magnetic fields within the region. Perfectly Matched layer (PML) guarantees the absorption of the field at the boundary without reflection. Boundaries far from the domain of computational interest can be represented as zero value SBC. COMSOL multiphysics which is the software used in this dissertation solves
PDEs by FEM methods. It has built in equations which implement the boundary conditions defined by the user. Using this package we study the transmission and super-resolving properties of MDS. Once the physics of the problem is well defined, approximate functions are selected for each sub-domain, and the whole system is assembled into a large linear system represented by matrices. The final solution is then obtained by LU decomposition technique. COMSOL Multiphysics has several solvers used to solve partial differential equations, describing the physics of the processes involved. The great power of COMSOL multiphysics is its ability to bring together many physical processes and solves all of them. For example it is able to solve for the electromagnetic fields within a MDS when light is incident on it, while also solving for the heat transfer processes generated as a result of this irradiation [25].

2.4 Super-resolving Properties of MDS

In this section the transmission and super-resolving properties of 4 different MDSs designs is examined. It is worth mentioning, that it is not always reliable in a practical sense to rely on published results of dispersion properties, of thin films, since it depends on many fabrication parameters. Nevertheless a careful study of these MDSs was performed, beginning with those with very ideal characteristics, which have not been attained by our state of art in fabrication to more realistic designs which we can fabricate. In order to demonstrate super-resolution and super-guiding there is need to build a Base structure (BS). The BS is the prior layers on which the MD stack multi-layers are built. The BS will be composed of glass, Cr patterned with PMMA filled apertures and a final planarized PMMA layer. In the simulations the buffer PMMA layer was omitted for
simplicity. In a MDS the metal focuses a diverging wave front to a tight spot inside the medium, thus exciting large spatial, spectral components (k-space), including evanescent k-resolved beyond the limits imposed by ordinary materials and by diffraction. [6].

Super-resolution is determined by propagating electromagnetic waves through the MDS and later examine if the objects inscribed at the object plane are resolved at the image plane by looking at the Poynting vector or the power at the exit layer of the MDS or some distance away from it. It may be interesting to also look at the square of the magnetic field, because this quantity is easily diffraction suppressed [6].

It is our desire in this study to push for super-resolution at longer visible wavelength. Nevertheless there are tough challenges to overcome, especially with the material characteristics of the MDS at these frequencies. In designing MDS for super-resolution, the following steps have been carefully considered. First identify the materials of choice a metal and a dielectric. Then choose realistic thicknesses for each of the layers. In some numerical studies, the authors have used very small thickness of a metal such as silver and dozens of layers [26]. It is difficult to use thicknesses of less than 10nm because in most practical systems, smaller film thicknesses than 10 nm is almost certainly dominated by island formation which also leads to a corresponding large surface roughness of sputtered or electron beam evaporated metal films.

Next we can use the TMM technique to calculate the transmission spectrum of the possible MDS layout. The goal here is to aim for super-resolution at significantly high transmission. Usually when the dielectric layers are made a lot (about 2 times) thicker than the metal layers. The super-resolving properties of the MDS will be weaker. A good
way to see this effect is to plot the dispersion curves for the MDS based on the Bloch wave dispersion properties as shown in the previous section.

Another quantity to examine is what I describe as the permittivity matching parameter $\delta_\varepsilon$ where $\delta_\varepsilon = |\text{real}(\varepsilon_m(\lambda) + \varepsilon_d(\lambda))|$. The sub-wavelength features are better resolved when $\delta_\varepsilon$ is small. We have the condition of perfect lensing when $\delta_\varepsilon = 0$.

### 2.5 Super-resolving Stack Results

#### 2.5.1 Super-resolving Stack 1 (SRS1)

For the first design, which we call SRS1, the optical transmission and field distribution characteristics are shown below. This design consists of 11 layers distributed as follows [GaP (20nm)/ 4.5 periods of Ag (20nm)/ GaP (30nm) /GaP (20nm)]. Figure 2.4 shows the transmission spectrum of SRS1. The transmission peaks around 45% even though there is 100 nm of silver in the MDS design.

![Figure 2.4: Transmission spectrum of Ag/GaP SRS1[27].](image-url)
In Figure 2.5 below a plot of the magnetic field squared within SRS1 is shown when the propagation wavelength is 532nm. We notice that the TMM and FEM technique used in calculating these fields agree strongly. This verifies that the FEM method is consistent in solving our MDS design problems for a case where we can compare with TMM results [27].

![Figure 2.5: Plot of the Magnetic field squared across the thickness of SRS1 consisting of Ag and GaP layers. There is an overlay of the bold line over the dash line. The red line is the FEM simulation, while the blue line is TMM solution. Both methods give a transmission of 44% at an incident wavelength of 532nm [27].](image)

When 30nm apertures are inscribed in the Cr layer, as seen in Figure 2.6 below where SRS1 exhibits super-guides the incident waves through out the structure. The apertures are separated by a center to center distance of 110nm. We see from that from the above figures that the magnetic field calculation by the TMM and the FEM gives the same result.
Figure 2.6: Plot showing the channeling of waves and resolution of two apertures inscribed on a Cr mask. The apertures are each 30nm wide and are separated by a center to center distance of 110nm. The whole structure is built on a glass substrate [27].

The Transmission at 532 nm is 0.4429. Both TMM code and COMSOL simulation gives the same result. The refractive index of GaP at 532 nm is taken as $n_{GaP}=3.2996-i*0.0384$, while that of Ag is $n_{Ag}=0.1301-i*3.1947$. If we plot the power (Poynting vector) at the exit surface of SRS1. When obtain Figure 2.7 below.
Figure 2.7: Shows the cross section of the power at the exit surface of SRS1 at 532nm. We notice the resolution of the two apertures inscribed on the mask. The apertures inscribed on the Cr mask are 30nm wide and separated by distance of 110nm [27].

If the apertures on the Cr mask are made smaller (20nm wide) and are now separated by a center to center distance of 80nm we can solve the wavelength were maximum super-guiding is observed. We can plot the power at the exit surface for 13 wavelengths within our bandwidth of interest (500nm-595nm) as shown in Figure 2.8 below. We notice that super-guiding is maximized at a wavelength of 532nm, where we noticed the highest contrast.
Figure 2.8: Shows the cross section of the power at the exit surface of SRS1 for a series of wavelength [500nm, 510nm, 515nm, 520nm, 525nm, 532nm, 540nm, 550nm, 560nm, 570nm, 580nm, 590nm, 595nm]. The apertures are each 20nm wide and are separated by a center to center distance of 80nm. The whole structure is built on a glass substrate.

If we calculate the power at several position after the exit layer of SRS1, we can estimate how far away from the lens, the super-resolving properties will be lost. In Figure 2.9 below, the Power is calculated at 4 positions. First at the exit surface of the lens and later at 3 other position. We notice that 30nm away from the lens, super-resolution is completely lost.
Figure 2.9 Plot of Power at several z-positions (exit surface, 20nm, 40nm, and 60nm) behind SRS1. For this simulation the apertures are each 20nm wide and are separated by a center to center distance of 80nm. The whole structure is built on a glass substrate.

SRS1 can resolve subwavelength features for several wavelengths in the range (500nm to 595nm). The resolution is best around 532nm. The resolution as drops significantly as distance behind the exit layers is increased from 0-60nm. This result is important for experimental validations of the concepts discussed in this chapter, as it will be necessary to know how close one bring a microscope tip.

2.5.2 Super-resolving Stack 2 (SRS2)

A second MDS design, denoted here as SRS2, is made up of Au and GaP distributed as follows: [GaP (20nm)/3.5periods (Au (20nm)/GaP (30nm))/GaP (20nm)]. Its transmission spectrum is shown below in Figure 2.10 and the magnetic field squared profile is shown in Figure 2.11.
Figure 2.10: Transmission spectrum of Au/GaP MDS SRS2 [27].

Figure 2.11: Plot of the Magnetic field square across the thickness of SRS2 consisting of Au and GaP layers. The thin solid line is the TMM result, while the dash line is the FEM result. Both methods give a transmission of 42% at an incident wavelength of 600nm [27].
Figure 2.12: $\log_{10}$ (Power) plot of the electromagnetic field propagating through two 50 nm slits separated by 100 nm at a wavelength of 600 nm. The apertures are 50 nm wide apertures separated by a center to center distance of 100 nm [27,28].

The Transmission at 532 nm is 42% Both TMM code and COMSOL simulation gives the same result. The refractive index of GaP at 600 nm is taken as $n_{GaP}=3.2405-i*0.0294$, while that of Au is $n_{Au}=0.2188-i*2.8625$. 

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Figure 2.13: Shows the cross section of the power at the exit surface of SRS2. We notice the resolution of the two 50nm wide apertures inscribed on the mask. The incident wavelength is 600 nm [27, 28].
Figure 2.14: Shows the cross section of the power at the exit surface of SRS2 for a series of wavelength [620nm, 625nm, 630nm, 635nm, 640nm, 645nm, 650nm, 655nm, 660nm]. The apertures are each 20nm wide and are separated by a center to center distance of 80nm. The whole structure is built on a glass substrate.
Figure 2.15: Plot of Power at several z-positions (exit surface, 20nm, 40nm, and 60nm) behind SRS2. For this simulation the apertures are each 20nm wide and are separated by a center to center distance of 80nm. The whole structure is built on a glass substrate. The wavelength is 640nm.

Similar to SRS1, SRS2 also shows a drop in resolution as the distance behind the stack increases. At about 30nm away from last layer, the resolution collapses.

2.5.3 Super-resolving Stack 3 (SRS3)

The third MDS design, which we call SRS3, consists of Au and TiO$_2$ layers. Its layout is described below. It has a total of 9 layers distributed as [TiO$_2$ (20nm)/3.5periods (Au (20nm)/TiO$_2$ (30nm))/TiO$_2$ (20nm)]. Figure 2.16 below shows the transmission spectrum of SRS3. The transmission is highest at about 620nm with a value of 0.3. In Figure 2.17 SRS3 is used to resolve two 50nm apertures separated by a distance of 110nm, when the propagation wavelength is 532nm.
Figure 2.16: Transmission spectrum of Au/TiO$_2$ SRS3.

Figure 2.17: Plot showing the channeling of waves and resolution of two apertures inscribed on a Cr mask. The apertures are each 50nm wide and are separated by a center to center distance of 110nm. The whole structure is built on a glass substrate. The propagation wavelength is 532nm.
The Transmission at 532 nm is 24% Both TMM code and COMSOL simulation gives the same result. The refractive index of TiO$_2$ at 532 nm is taken as $n_{\text{TiO}_2}=2.4379$, while that of Au is $n_{\text{Au}}=0.4522-i*2.4067$.

![Graph](image)

Figure 2.18: Shows the cross section of the power at the exit surface of SRS3. We notice the resolution of the two apertures inscribed on the mask. The incident wavelength is 532nm.

Consider a control sample where all the metallodielectric layers are become a dielectric of refractive index $n=3.5$. It is shown below in Figure 2.19 that this design will not super-resolve the 50nm structures separated by a Center to Center distance of 110nm. This example goes on to confirm the relevance of using metallic layers within the Stack, because some authors have argued that it is irrelevant to build metallodielectric that the diffraction limit can simply be pushed down with use of high index dielectric.
Figure 2.19: Surface plot of the $\log_{10}$ (Magnetic field) when all the metallodielectric layers are replaced with a high index dielectric of refractive index 3.5. We notice that diffraction dominates and there is no super-guiding through the stack the resolution of the two holes. The incident wavelength is maintained at 532nm.

One might feel that a high refractive index value and the absence of loss in the dielectric will guarantee an improvement in super-resolution as in the work by Y Jin [26]. But this is not generally the case as most often the extinction coefficients of the dielectric which contributes to the material loss is necessary to achieve near permittivity matching conditions and flat dispersion curves.

The results above agree with the Scalora et al. results which identifies two regimes a focusing regime at the edges of the pass band with no surface plasmon excitation, and a beam channeling, or super-guiding regime at the center of the pass band for a transparent metal Ag/GaP structure, that favors the onset of transverse surface plasmons and super-resolution [5]. The metallodielectric photonic crystal structures are
thicker than single metal layers; they contain more metal and have higher transmission.
The focusing regime is prevalent at shorter wavelengths, near the edge of the pass-band.
The output shows a focused spot at a standoff distance from the output plane. The
channeling regime is apparent at longer wavelengths; the beam width in the
metalldielectric structure can be maintained to a smaller fraction of a wavelength, but
the features are eventually lost as the distance from the surface is increased.

2.6 Comparison of TMM and FEM

In Section 2.2.2 we had discussed the extension of the TMM to handle “Super-
Gaussian” beams. With this capability is then possible to analyze the super-resolving
properties of MDS with the TMM method and compare the results to those obtain with
FEM.

First we propagated a super-Gaussian beam of beam waist 50 nm through a single
50nm aperture with beam with TMM and FEM in 200nm of air and found that both
results match perfectly as shown below in Figure 2.20. Next we introduce two apertures
separated by a center to center distance of 700nm and keep the beam characteristics the
same and propagate through 200nm of air.
Both the TMM and FEM agree reasonably well for a single beam in air. When we introduced two beams we begin to notice the differences with, the TMM exhibiting a higher contrast in resolution than the FEM.

Consider a 200 nm thick MDS with the following distribution [TiO2 (40nm) 1.5periods of (Ag (20nm)/TiO₂ (80nm)/TiO₂ (40nm)] denoted as SRS4. If we incident a “Super Gaussians” unto a SRS4 at 500nm. The FEM results reveal a broader waist of the Gaussian at the output compared to TMM result indicates that the FEM is picking up more of the diffraction terms. This is shown in Figure 2.21 below. The fields are all normalized with respect to maximum value.
Figure 2.21: Normalized electric and magnetic field squared at the exit layer of MDS. A SG beam is incident unto a 50 nm aperture inscribed on the MDS with both TMM (black) and FEM (blue). The SG has a spot size of 50nm, the incident wavelength is 500nm.

The result is different from the case in air in which both methods gave identical results; this seems to indicate the multiple reflection terms which is created by the introduction of multilayer brings about diffraction terms which are difficult to track with the TMM. When two 100 nm waist SG, separated by a center to center distance of 300nm are incident unto the MDS at 500nm wavelength, in this case TMM fail to show the side lobe pattern which is seen for the FEM case.
Figure 2.22: Normalized electric field squared at the exit layer of MDS. A SG beam incident unto two 100 nm apertures, separated by a center to center distance of 300nm inscribed on the MDS with both TMM (Black) and FEM (Blue). The SG has a spot size of 100nm, the incident wavelength is 500nm.

2.7 Hyperlens Design

The planar superlenses discussed in the previous section will only guarantee near field imaging with a magnification of 1. In order to magnify images at the output of the stack and realize far field imaging, a Hyperlens designed was first proposed and verified by Zhang et al [29]. With this design they were able to resolve 60 nm structures with a MDS made up of Ag and Al$_2$O$_3$ layers with an incident wavelength of 365nm. The Hyperlens is essentially composed of cylindrical layers of metallodielectric instead of planar layers. We have considered a Hyperlens design which is analogous to SRS1 design and show how channeling and super-guiding occurs in this MDS at a wavelength of 532nm. This design is shown in Figure 2.23 below.
Figure 2.23: A plot showing the surface power as channeling and magnification is experienced with a Hyperlens whose sequence of layers is the same as SRS1. The incident wavelength is 532nm and the two apertures of about 42 nm width are separated by an angular separation of 30 degrees.

In Figure 2.24 below, that the Magnetic field squared is resolved at the exit layer of Hyperlens. This simulation also demonstrates how the Hyperlens can magnify the images of Sub-wavelength details as this will be necessary to enable super-resolution in the far-field.
Figure 2.24: Plot of the Magnetic field squared along the exit layer of Hyperlens, when the wavelength is 532nm.

2.8 Chapter Summary

So far we have seen how permittivity matching, diffraction suppression affect the choice of metallodielectric designs for the visible region. We have discussed specific designs whose super-resolution ability is phenomenal given that we are considering realizable material characteristics.

In order to fabricate and validate these devices there are some practical barriers that have to be overcome, which includes mainly the surface roughness of the thin films after deposition. If the films are very rough then the properties predicted by theory will
not be achieved scattering will altogether smear the beam profile. In the FEM simulations for super-resolution it is more practical to include a buffer layer just before the MDS this buffer layer should be well planarized in order to avoid scattering. A good candidate for such a layer is PMMA [11]. In the past we have deposited and etched PMMA layers which served as buffer layers for the MDS in order to prevent surface roughness from destroying the resolving of the superlens.

We have verified that the usage of higher index dielectrics for the superlens design in the main way to achieve super-guiding in the visible region. We have calculated optimal super-resolution regime for an Ag/GaP MDS (SRS1) and Au/GaP MDS (SRS2), and showed how 20nm apertures can be resolved, when visible incident light is used. The transmission function which confirms our findings for SRS1 and SRS2 by calculating the transmission in both the propagating and the evanescent regime is shown in Appendix D. Previous research endeavor have not been devoted to this region because of the high losses of the metals at visible wavelengths and studies were done mainly for cases with idealized materials, which have not been realized with the current art of nanofabrication. We have compared the FEM and TMM technique; two popular techniques used in predicting super-resolution, and have shown how they differ as the complexity of the MDS increases. Blaikie et al did a similar calculation and concluded that the FEM is more robust in handling such problems; they came up with a modified TMM code which agreed better with their FEM simulations [25]. Our TMM doesn’t deviate a lot from our FEM results, yet we believe that experimental results will provide a final guide in adapting both codes for the Super-resolution task.
CHAPTER 3

NONLINEAR ENHANCEMENTS WITH METALLODIELECTRIC STACKS

3.1 Introduction

In this chapter, we examine the contribution of the nonlinearity of the separate layers to the entire nonlinearity of the Metallodielectric stack (MDS). Experimental investigations have reported significant nonlinear response of MDSs with constituent metal films of silver (Ag) and gold (Au) [30-32,33]. High nonlinear susceptibilities \( \chi^{(3)} \), have been reported for Cu based MDS [8,33,34].

Strong nonlinear coefficients are found in the layers MDS, which causes enhancements of the electromagnetic field within the MDS. In this chapter finite element simulation with COMSOL multiphysics software is performed, together with Transfer matrix method technique, to calculate the electromagnetic fields within the MDSs.

The dispersion data for the MDS materials are taken from [16] for the metals Ag, Cu and while that of ZnS and TiO\(_2\), are measured data. We have numerically simulated the propagation of a continuous wave (CW) through MDSs.
This was done by assuming the initial incident wave to be a typical Gaussian beam, as expressed in Eq (3.1) below, which is propagated through the MDS. Once the field at the exit layer of stack is known, we take its Fourier transform, which gives us the field characteristics at the far field. Once we can calculate the field at the far field, then we can repeat the process for several positions (i.e. z values) around the focus of the beam. This leads exactly to the popular Z-scan technique, which reduces to finding the transmission through a sample at the far-field as a function of sample placements. The main difference here is that, our technique handles the complexities of the metallic photonic band gap material (PBG), and doesn’t assume a homogenous material which is often the case for most Z-scan experiments.

Our results show the typical nonlinear absorption (NLA) for CW propagation in MDS, is similar to what is seen in experiments. Pulsed simulations will be discussed in Chapter 4, since we used pulsed lasers to conduct the experiment, which does not significantly change the physics of problem. The layers of metallodielectric are very thin, so for the short interaction time, the field could be assumed to be quasi CW. Principally in this chapter we show how a finite element method provides an efficient means to model a numerical Z scan experiment involving MDS which are 1D photonic band gap devices. The problem of modeling the Z-scan experiment for 1D PBG poses new constraints for the traditional Z-scan techniques, which rely essentially on the beam propagation method formulated for homogenous materials. The 1D PBG is a non homogenous material which shows strong back reflections which are often ignored in the traditional Z-scan models [35, 36, 37]. This approach to the problem also solves the
corresponding Maxwell’s equations in the most rigorous sense without using popular approximation techniques such as the slow varying envelop approximation.

3.2 Field Enhancements in MDS

In Chapter 2 we were able to calculate the electromagnetic fields within a MDS under a plane wave incidence at TM and TE polarizations. We used the TMM algorithms and completely neglected the typical third order nonlinearities which are significant for metals. In this section we proceed to show how the inclusion of the nonlinear properties of the materials can affect the fields within the MDS. Typically we expect that if the input beam intensity is strong enough, the corresponding nonlinear phase shift induces a nonlinear complex index change which is recognizable when the numerical Z-scan experiment is performed. For simplicity we would assume a typical input beam to be a continuous wave (CW) of TEM$_{00}$ mode. The input beam is described by Eq. (3.1) below for CW propagation $E_0(t) \rightarrow E_0$.

$$E(z,r,t) = E_0(t) \frac{w_0}{w(z)} \exp \left( -\frac{r^2}{w^2(z)} + i \frac{\pi r^2}{\lambda R(z)} + i \phi \right). \quad (3.1)$$

In the experiment the MDS is moved about the focus of a beam through positions, which are multiples of the diffraction length $z_0 = \frac{\pi w_0^2}{\lambda}$. The beam waist and radius of Curvature of the beam is described by $w$ and $R$ below.
\[ w(z) = w_0 \left( 1 + \frac{z^2}{z_0^2} \right)^{1/2}, \]
\[ R(z) = z \left( 1 + \frac{z_0^2}{z^2} \right). \]  

\((3.2)\)

3.2.1 The Theory of Beam Propagation

The propagation of light within metals has not gathered enough interest since metals are essentially known to be reflective. Yet there are great opportunities to study the interaction of electro-magnetic fields and metals. As an electric field tunnels through a typical MDS stack at high intensities, it dynamically changes the local index of refraction, which alters the local field profile. Since analytical solutions of Maxwell’s wave equation for the propagation of field inside MDS structures is only possible under very restrictive conditions [7], we are compelled to resort to numerical techniques.

In Chapter 2, we saw the how the FEM describes quantitatively the diffraction of a beam as it propagates through the stack. The traditional method which has been used to illustrate in simplicity the diffraction effects of a beam as it propagates has been the fast Fourier transform beam propagation method (FT-BPM). This method has used extensively especially in fiber optics research [38]. The weakness of this method with regards to the problem we are considering in this work is that, this method does not include back reflections, secondly it is usually implemented with the assumption that the second order derivatives of the amplitude of the field with respect to time and the displacement along \(z\), the propagation axis is negligible. Scalora et al developed a
modified time domain version of FT-BPM which handles back reflections and retains the simplicity of the standard FT-BPM [39, 40]. This modified version of the FT-BPM does not assume slowly varying envelope approximations for the amplitude of the fields since, this approximation will not be suitable because of the significant refractive index changes across the layers that make up the one-dimensional photonic band gap structure. The limitations of the mentioned techniques guided us to consider a finite element approach which we know will handle the diffraction problem and the complexities of back reflections accordingly. We also envisage applying it to correctly handle the nonlinear propagation problem.

Therefore a full finite element method (FEM) approach to the problem will also avoid such over simplifications and provide a good validation check for similar calculations performed by other methods such as the TMM. The significant refractive index change across an interface requires that the propagation equation be supplemented by matching boundary conditions. Although this can be handled by using the above description treating the field as slowing varying everywhere except at boundary, the problem becomes cumbersome and somewhat unmanageable if the number of such boundaries is large. The standard method then fails if there are many such boundaries within a wavelength as might be the case for MDS [39]. The equation system showing the interaction of laser beam propagation with a MDS with third order nonlinearities is as shown below. For brevity and simplicity, we will work with the complex fields. The physical (real) field is given by half the sum of the field and it’s conjugate. Later we will solve for specific cases of MDS with COMSOL.
\[
\n\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad (3.3)
\]
\[
\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t}. \quad (3.3)
\]

If the medium is nonlinear then, the electric displacement vector, can be written as

\[
\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}_L + \mathbf{P}_{NL}, \quad (3.4)
\]

where \( \mathbf{P}_L = \varepsilon_0 \chi^{(1)} \mathbf{E} \) and \( \mathbf{P}_{NL} = \varepsilon_0 \chi^{(2)} \mathbf{E} \mathbf{E} + \varepsilon_0 \chi^{(3)} \mathbf{E} \mathbf{E} \mathbf{E} + \ldots \) represent the linear and nonlinear polarization terms, respectively. The tensor product of the electric susceptibilities \( \chi^{(i)} \) and electric field(s) always gives us a vector.

\[
\nabla \times \nabla \times \mathbf{E} = -\mu_0 \left( \varepsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} + \frac{\partial^2 \mathbf{P}_L}{\partial t^2} + \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2} \right), \quad (3.5)
\]
\[
\nabla (\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} = -\mu_0 \left( \varepsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} + \frac{\partial^2 \mathbf{P}_L}{\partial t^2} + \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2} \right). \quad (3.5)
\]

3.2.2 Z-Scan Technique Development for Photonic Band Gap Materials

The standard Z-scan technique which has been used extensively to characterize several kinds of materials including a 1D photonic band gap device, such as our MDS, usually ignores the losses due to internal multi-interference and back reflections, which contributes to the absorption within each layer [41, 42]. Even when these phenomena are considered as in the case of a bidirectional beam propagation method, transverse effects important in describing the beam profile are often approximately handled [33].
scan experiment a sample is scanned through a range of $\pm n z_0$, \( n \geq 1 \) and the transmitted power at the detector is recorded as function of position $z$ of the sample. The incident beam is focus with a positive lens at the center position of the scan range [42]. With this technique both real and imaginary parts of the third-order susceptibility can be determined as opposed to the complicated degenerate four-wave mixing technique (DFWM) which required very precise alignment of beams yet could only account for the magnitude of the third order nonlinear susceptibility [43]. The Z-scan technique is a method which can rapidly measure the nonlinear absorption (NLA) and nonlinear refraction (NLR) of solids and liquids [42]. We will consider only a Transmittance Z-scan with thin samples ($L \ll z_0$) and exclude higher order nonlinearities. Where $z_0$, is the diffraction length of the incident beam and $L$ is the length of the sample. The technique is often modified to measure Reflectance instead of transmittance and the theory can be enhanced to account for thick samples ($L > z_0$) and fifth order nonlinearities. The incident beam in the analysis considered in this section will be Gaussian TEM\(_{00}\) mode. A simple schematic of a Z scan set-up is shown in Figure 3.1 below.
The open aperture Z-scan is used to characterize the NLA of materials. In the open aperture Z scan mode, all the light transmitted through the sample is captured by the detector; this is implemented by keeping the iris close to the detector wide open. The detector is always placed at a distance considered to be at the far field. Then the aperture transmittance $T = 1 - e^{-2r_a^2/w_a^2}$, for the open aperture is equal to 1. Where $r_a$ and $w_a$ denote the radius of the aperture and the beam waist at the aperture. The two-photon absorption coefficient, $\beta_2$, is determined from the normalized transmitted energy $T(z)$ through the sample in the open aperture mode given by Eq. (3.6) below. Where $q = \beta_2 I_0 L_{\text{eff}}$ and $I_0$ is the $z$-axis irradiance; $L_{\text{eff}} = (1 - \exp(-\alpha L))/\alpha$ is the effective length. And $\alpha$ is the linear absorption of the sample.

$$T(z) = \sum_{m=0}^{\infty} \frac{(-\beta_2 I_0 L_{\text{eff}})^m}{(1 + z^2/z_0^2)^m(m + 1)^{3/2}} .$$

(3.6)
For a closed aperture experiment the change in transmittance between the peak and valley in the Z-scan is denoted as $\Delta T_{pv}$. This quantity is proportional to the nonlinear refractive index as seen in Eq. (3.7) below: The quantity $\Delta \phi_0$ is the induced phase shift [42].

$$\Delta T_{pv} \approx 0.406 (1 - S)^{0.27} \left| \frac{2\pi}{\lambda} n_2 I_0 L_{eff} \right|, \quad \Delta \phi_0 = \frac{2\pi}{\lambda} n_2 I_0 L_{eff}. \quad (3.7)$$

### 3.3 Linear Properties of MDS with Transfer Matrix Method

We applied the transfer matrix method (TMM) to study the linear properties of the field for different angles of incidence and wavelengths to determine the dispersion characteristics like the group index. In the linear regime we used the TMM to verify that the FEM results accurately represent the physical problem. Using the TMM method the field is decomposed into plane waves and the forward- and backward-propagating amplitudes are found by using a series of 2x2 matrices that contain the optical path lengths in the adjacent media and the polarization-dependent boundary conditions [20, 33]. For linear media the TMM is a faithful representation of the field across the sample. The properties that are most often examined are the transmittance and reflectivity of the sample. The transmittance and reflectance amplitudes are expressed in complex form as

$$t = |t| e^{i\phi}, \quad r = |r| e^{i\phi}. \quad (3.8)$$
The quantities $\phi_t$ and $\phi_r$ are the phase changes in the transmitted and reflected wave amplitude due the complex interference between the different wave paths. The plane-wave transmittance and reflectance coefficients are defined for any angle of incidence as

\[ T = \frac{n_t \cos \theta_t}{n_0 \cos \theta_i} |t|^2, \quad R = |r|^2, \]

(3.9)

$\theta_i$ and $\theta_t$ are the incident and transmitted angles of incidence, and $n_0$ and $n_t$ are the incident and transmitted medium refractive indices, respectively. The absorbance $A$ is calculated using the relation

\[ A = 1 - T - R. \]

(3.10)

The phases in Eq. (3.8) contain information about spectral dispersion of the transmitted and reflected waves [20, 33]. The phase of the transmitted wave is related to an effective propagation constant used for optical interactions:

\[ \varphi_t = k_e L, \]

(3.11)

where $L$ is the thickness of the sample and $k_e$ is the effective propagation constant. A quantity of interest in the present studies is the group index, which is defined as:

\[ N_g = \frac{c}{\omega} \frac{\partial k_e}{\partial \omega} = \frac{c}{L} \frac{\partial \varphi_t}{\partial \omega}. \]

(3.12)
3.3.1 Energy Velocity and Group Velocity Contribution

In a 2001 paper Scalora et al analyzed the notions of group velocity $V_g$ and energy velocity $V_E$ for light pulses propagating inside one-dimensional photonic band gap structures of finite length. They found that that the two velocities are related through the transmission coefficient $t$ as $V_E = |t|^2 V_g$ [44]. We seek to examine the role of energy velocity and the group velocity of electromagnetic pulses in determining significant nonlinear activities within a MDS. They considered a periodic photonic band gap material with indices of 1 and 1.42857 respectively. The incident wavelength was 1 micron and the thickness of the layers was 250nm and 350nm respectively. The plotted energy velocity versus the reduced frequency $f_n = \omega / \omega_0$, for different number of periods $N$, of the respective materials. We have used our TMM program to compare this result for $N=20,100$ and found them to match exactly. These results are shown in Figure 3.2 below. With this check we can safely examine consider the energy velocity of our MDSs of interest [44].
Figure 3.2: Energy velocity versus reduced frequency for N periods structure of a 1D photonic band gap material made up layers with refractive index 1 and 1.42857 respectively. The incident wavelength is 1 micron and thicknesses of the respective layers are 250nm and 350nm respectively. (Left): Result from [44] when N=20, N=100, and N=infinity. (Right): Our results when N=20 and N=100.

The derivation of the energy velocity is based upon a treatment of the subject in [45]. The energy velocity is determined as follows; first we calculate the total energy density per unit volume. This is usually given by a sum of the electric and magnetic components as shown in Eq (3.13) below

\[
U = \frac{1}{4} \left( \frac{\partial (\omega \varepsilon)}{\partial \omega} |E|^2 + \frac{\partial (\omega \mu)}{\partial \omega} |H|^2 \right) = U_E + U_M. \tag{3.13}
\]

\(U\) is the energy per unit volume.

We can simplify Eq (3.13) above as follows
Let \( g \) be known as a material permittivity factor, so for each material such as Cu and TiO\(_2\) considered we will calculate its permittivity factor at various wavelength. We take just the real values of the function \( g \).

The Poynting vector is defined as

\[
S_{av} = \frac{1}{2} \text{Re}(E \times H^*) .
\]  

(3.15)

For a MDS of length \( L \) the energy velocity

\[
V_E = \frac{\int_0^L E \cdot dz}{\int_0^L U dz} .
\]  

(3.16)

Therefore analogous to the group velocity index the energy velocity index is defined as

\[
N_E = c / V_E .
\]  

(3.17)

The Matlab program for energy velocity is given in Appendix B. We report two different MDS designs. The first, denoted as MDS1, which has 9 layers consisting of a first layer of TiO\(_2\), then 3.5 periods of Cu and TiO\(_2\) and a final layer of TiO\(_2\). This distribution is simply denoted as: (TiO\(_2\)(40nm)|[Cu(20nm)|TiO\(_2\)(80nm)]\(_{3.5}\)|TiO\(_2\)(40nm)). The thickness
of the layer is shown in brackets. called MDS2 has 7 layers, distributed as

:ZnS(40nm)[ Ag(30nm)|ZnS(80nm)][2.5]ZnS(40nm) . The energy velocity of MDS1 is
plotted as function of wavelength in Figure 3.2. The transmittance, absorbance and the
group index of MDS1 is plotted in Figure 3.4 as a function of wavelength. The energy
velocity of MDS2 is plotted as function of wavelength in Figure 3.5. The transmittance,
absorbance, group velocity index, and energy velocity index of MDS2 is plotted in Figure
3.6 as a function of wavelength the group index serves as an initial guide where we can
identify slow light wavelength regions that could be examined further for enhanced
nonlinear effects. The group index has a strong peak at the edges of the transmission
spectral bands, which approaches or exceeds 20 for these portions of the transmission
band.
Figure 3.3: Optical properties of the TiO$_2$ and Cu multilayer system called MDS1. The sequence is a 40 nm thick TiO$_2$ layer followed by three and one-half periods of 20 nm thick Cu film and 80 nm thick TiO$_2$ film and finally a 40 nm thick TiO$_2$ film at the other end. This design is a transparent metal and we denote it by the following notation describing the thickness of each layer: TiO$_2$(40nm)/[Cu(20nm)/TiO$_2$(80nm)]$_{3.5}$/TiO$_2$(40nm). The Energy velocity versus the wavelength is shown above for MDS1.
Figure 3.4: Optical properties of the TiO$_2$ and Cu multilayer system called MDS1. The sequence is a 40 nm thick TiO$_2$ layer followed by three and one-half periods of 20 nm thick Cu film and 80 nm thick TiO$_2$ film and finally a 40 nm thick TiO$_2$ film at the other end. This design is a transparent metal and we denote it by the following notation describing the thickness of each layer: [TiO$_2$(40nm)/[Cu(20nm)/TiO$_2$(80nm)]$_{3.5}$/TiO$_2$(40nm)]. (Left): Transmittance and absorbance versus wavelength. (Right): Group index and Energy velocity index versus wavelength [33].

For the ZnS/Ag MDS denoted as MDS2. We have plotted the energy velocity as function wavelength in Figure 3.5 below. We see that the energy velocity is at its maximum at about 600nm. Its transmittance, absorbance, energy velocity index and group velocity index is show in Figure 3.6.
Figure 3.5: Optical properties of the ZnS and Ag multilayer system called MDS2 [ZnS (40nm)/[Ag(20nm)/ZnS(80nm)]_{2.5}ZnS(40nm)]. The Energy velocity versus wavelength is shown above for MDS.

Figure 3.6: Optical properties of the ZnS and Ag multilayer system called MDS2 [ZnS (40nm)/[Ag(20nm)/ZnS(80nm)]_{2.5}ZnS(40nm)]. (Left): Transmittance and absorbance versus wavelength. (Right): Group index, Energy velocity index versus wavelength [33].

The spectral width of the transmission bands is determined by the properties of the materials, the thickness of the films and the number of layers. The first and last layers are dielectric layers and the outer films are half as thick as the bulk dielectric layers. This
has the effect of raising the transmittance and reducing the local Fabry-Perot-like peaks in the spectrum [7, 33]. The designs with this feature are called transparent metal structures.

For both structures the group index has a peak near the pass band edges in the transmission spectrum. At the long wavelength pass band edge there is a peak in the absorbance that corresponds with the group index peak; the absorbance indicates that the field is localized in the metal film. The short wavelength pass band either does not have an absorbance peak or the absorbance peak does not coincide with the local maximum of the group index. We find that the field near these wavelengths is largely localized in the dielectric [33].

3.4 Nonlinear Characterization with Finite Element Method

The open aperture Z-scan experiment leads to the determination of the two-photon absorption coefficient. The FEM used in this calculation is implemented using COMSOL 3.5a. The FEM method can be used to describe closed aperture Z-scan experiments. The corresponding nonlinear wave equation which is solved by FEM in cylindrical coordinates, when the input beam of wavelength \( \lambda \) is TE polarized and the free space wave number is \( k_0 = 2\pi / \lambda \). The wave propagates in the z direction is given below.

\[
\nabla \times \left( \frac{1}{\mu_r} \nabla \times E_\varphi \right) - \varepsilon_r k_0^2 E_\varphi = 0.
\]  

(3.18)
The relative magnetic permeability $\mu_r$ is unity for all the materials used in our calculations. The dielectric function depends on the irradiance with the following dependence:

$$\varepsilon_r = (n_L + j S_{av,z})^2,$$

(3.19)

where $n_L$ is complex the refractive index of the metal or the dielectric in the linear regime, and the nonlinear coefficient, $\gamma = n_2 - \frac{i\lambda \beta_2}{4\pi}$, has a complex value. The Kerr coefficient $n_2$ affects the refractive index and $\beta_2$ creates a nonlinear absorption effect. The effect on the phase and log-amplitude is proportional to the irradiance profile. We take advantage of the cylindrical symmetry of the beam in our calculations. The vector field is given by $\mathbf{E}_\phi = \hat{e}_\phi \mathbf{E}$ where the unit vector is transverse to the direction of propagation.

The beam width at the focal plane is $w_0$ and The Rayleigh range $z_0 = \frac{\pi w_0^2}{\lambda}$. The beam width at position $z$ is $w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_0}\right)^2}$. The radius of curvature of the phase is

$$R(z) = z \left[1 + \left(\frac{z_0}{z}\right)^2\right].$$

The scalar field is given by Eq. (3.19) above [33].

MDS1 is made of Cu and TiO$_2$. The Cu metal was chosen because it is reported to have a high third-order nonlinear coefficient among the metals of interest [8, 34]. For MDS2 designed from materials Ag and ZnS. In comparison to Cu, relatively small third-order nonlinearity has been reported for Ag [9, 30-31]. The linear optical transmission and absorption characteristics of MDS1 using TMM are plotted in Figure 3.4. The FEM
transmission spectrum was calculated for specific wavelengths and was in close agreement with the TMM results.

Using the TMM and FEM simulations the field inside the sample was calculated. On the left in Figure 3.6 is the TMM calculated field amplitude throughout the sample at a wavelength of 650 nm for three angles of incidence. The wavelength was chosen because it is near the transmittance maximum. TMM results on the left shows the TE-polarized field dependence inside the sample for several angles of incidence. We note that the field localization inside the metal has the largest amplitude for normal incidence. Normal incidence should have a large nonlinear effect in the sample. On the right is a comparison of the fields at normal incidence in the sample found between the FEM and the TMM simulations. The two results are nearly identical and provide a direct verification of the FEM approach for this case.

![Graphs showing field squared at different angles and at normal incidence](image)

**Figure 3.7:** Shows field squared at a wavelength of 650nm throughout the sample. The vertical lines are positions of the metal/dielectric interfaces. (Left): TMM plot of field squared for TE polarization at different incident angles (0 degrees (solid), 30 degrees (long dash) and 45 degrees (dotted). (Right): FEM and TMM plot of the field at plane wave normal incidence [33].
To verify the nonlinear transmission characteristics a Z-scan experiment of a homogeneous material was simulated. An illustration of the transmission results for a Z-scan experiment is shown in Figure 3.8. The sample is moved a distance $z$ from the focus of a Gaussian beam, as illustrated in Figure 3.8. The Rayleigh range, $z_0 = k w_0^2 / 2$, is used to scale the position of the sample relation to the beam’s focal plane. The beam waist at the focal plane is $w_0$. In the far field an aperture is placed in front of the detector and the irradiance is recorded as a function of $z$. In a linear medium the transmittance is constant. We considered a 400 nm thick sample with Kerr nonlinearity $n_2 = 9 \times 10^{-16}$ m$^2$/W. The refractive index of the sample was chosen as 1.2 and the laser wavelength is 532 nm. The Gaussian beam waist is $w_0 = 20$ microns and the peak irradiance at the focal plane is 11.9 GW/cm$^2$. A CW beam is used in the simulations. The transmittance as function of position $z$ for the Z-scan experiment is given by Eq (3.20) below. Where $E_{NL}$ and $E_L$ are the nonlinear electric field and linear electric fields respectively at aperture which is determined by a Fourier transform of the electric field at the exit layer of the MDS, and $r_d$ is the radius of the aperture in front of the detector.

$$T(z) = \left[ \frac{\int_0^{r_d} r dr |E_{NL}(z,r)|^2}{\int_0^{r_d} r dr |E_L(z,r)|^2} \right].$$

(3.20)
Figure 3.8: Z-scan for a 400nm thick material whose refractive index at 532nm is 1.2 and has a nonlinear refractive index $n_2 = 9 \times 10^{-16} \text{m}^2/\text{W}$ [33].

The numerical Z-scan results are compared with the analytical results derived for a thin nonlinear medium [41]. The equation for the analytical result is:

$$T = 1 + \frac{4Z\Phi}{(Z^2 + 9)(Z^2 + 1)},$$

(3.21)

where $Z = z/z_0$ and $\Phi = k_0 n_2 L I$, where $I$ is the irradiance, $L$ is the sample thickness, and $k_0$ is the free space wavenumber. The transmittance is normalized to unity in the linear regime. The position and size of the peaks in our FEM simulations are in close agreement with the analytical results. The steeper drop of the FEM curves may be due to additional nonlinear effects in the simulation.

We also examined the group index and look for wavelength regions for slow light within the material; as a first guess we expect the slow light regions will lead to
enhancement of the nonlinear optical response. We consider the propagation of a typical CW Gaussian beam through the MDSs. We save the complex field at the exit layer of the stack and use it to calculate the normalized transmittance according to Eq.(3.20). We assume a CW source; with COMSOL we extract the phase and the amplitude of the Electric field at the exit layer of MDS, which is numerically integrated by taking a Bessel transform to determine the electromagnetic field at the far field.

This gives us the Z-scan trace, from which the overall nonlinear optical coefficients can be determined. We obtain the nonlinear optical constant of the materials used in the simulations. As a check to our nonlinear FEM calculations we compared both FEM and TMM results for the linear case and check agreement before solving the nonlinear problem. The results are presented for two MDS samples. First we consider the sample composed of Cu and TiO$_2$ layers (MDS1) and then we study the sample made from Ag and ZnS layers (MDS2).

The origin of the nonlinearity in our samples is attributed to electronic transitions which produce third-order susceptibility. There are intraband and interband transitions which affect the susceptibility when electrons fill empty states. A Fermi smearing effect, commonly known as the hot electron contribution, creates a sea of thermally excited electrons with energy levels just around the Fermi energy. These effects were recently examined for Ag films by Owens et al. [30, 31]. The electron temperature changes under strong fluence irradiation are large enough to elicit strong transmittance changes. Classical electromagnetic local field enhancements caused by electron oscillations known
as surface plasmons will amplify the nonlinear effect. For our simulations we describe the nonlinearity by purely instantaneous third-order nonlinearity.

3.5 Numerical Z-scan Results

The sample labeled MDS1 in this paper has a total of 9 layers with material and layer thickness as follows: TiO$_2$ (40nm)/[Cu(20nm)/TiO$_2$(80nm)]$_{3.5}$/TiO$_2$(40nm). The sample has 80 nm of Cu metal and is 400 nm thick. From the transmittance, absorption and group index results in Figure 3.4 we observe a wide transmission spectral band extending from below 500 nm to greater than 800 nm. Near the ends of the spectral bands the group index has a distinct maximum. The group index alone though does not reveal whether or not the field is localized in the metal films. The nonlinear parameters we use for Cu are: $n_2 = 5 \times 10^{-11} \text{cm}^2/\text{W}$ and $\beta_2 = 5 \times 10^{-6} \text{cm/W}$ similar to the results published in [46,47]. The nonlinear parameters for TiO$_2$ are $n_2 = 1.33 \times 10^{-16} \text{cm}^2/\text{W}$ and $\beta_2 = 13 \times 10^{-9} \text{cm/W}$ [48]. The normal incident electric field amplitudes for five wavelengths across the transmission band are shown in Figure 3.9. The four regions delineated by vertical lines are the Cu metal films. The field profile at 820 nm has the strongest amplitude in the first two metal films and the one at 500 nm penetrates the least. These are both wavelengths where the group index has a peak, but the absorbance peak only coincides with the long wavelength group index maximum. The metal nonlinearity produces its strongest affect at the long wavelength peak.
Figure 3.9: The electric field amplitude squared versus position in the sample MDS1. The vertical lines mark the interfaces between the TiO$_2$ and Cu films [33].

The numerical results for the open aperture Z-scan simulation of MDS1 is shown in Figure 3.10. In all cases the irradiance was constant and at the focal plane it is 1.9 GW/cm$^2$. The linear transmittance in each case has been normalized to unity. The numerical results for the open aperture Z-scan MDS1 simulation are shown in Figure 3.9. In this case the irradiance was constant and at the focal plane it is 1.9 GW/cm$^2$. The linear transmittance in each case has been normalized to unity. The strongest relative change around 15% in the transmittance is at 650 nm, where the penetration of the electric field in the metal films is strong. A strong dip of about 8% is observed at 820 nm and its linear transmittance is about five times lower than at 650 nm. The field localization in each metal layer at 650 nm is high and coincidentally it has the largest transmittance of the cases considered in Figure 3.9. We do not find a correlation between slow light regimes and strong nonlinear responses [33]. So the Stack experiences a lower nonlinear
absorption as compared to 650nm. At 600nm the absorbance and transmittance of MDS1 is also high, but the field is slightly less confined in the metallic layers as compared to the field at 650nm. The change in transmission at 600nm is about 13%. At 500 nm the field penetration is very weak; therefore the nonlinear transmission change is small.

![Open Aperture Z-scan Curves for MDS1](image)

**Figure 3.10**: Open Aperture Z-scan Curves for MDS1. The position of the sample is normalized by the Rayleigh range, $z_0$ [33].

The MDS2 sample has seven alternating layers of ZnS and Ag in the following sequence: ZnS (40nm)/ [Ag (20nm)/ ZnS (80nm)]$_{2.5}$ ZnS (40nm). The Electric field amplitude plotted in Figure 3.10 shows the beam’s confinement in both the longitudinal and transverse directions. The input radial beam shape is a Gaussian function, Eq. (3.1) with the sample placed at the focal plane. The nonlinear parameters we use for Ag are: $n_2 = 2 \times 10^{-11} cm^2/W$ and $\beta_2 = 3.3 \times 10^{-8} cm/W$. The nonlinear refraction coefficient of ZnS is zero, while its absorption coefficient $\beta_2 = 3.4 \times 10^{-9} cm/W$. These values are taken
from [48]. The propagation of a Gaussian beam through MDS2 is shown in Figure 3.11 below.

Figure 3.11: The electric field amplitude squared for the radial coordinate (vertical) and the longitudinal coordinate at 600 nm in the focal plane. The field is largely localized in the ZnS regions delineated by vertical lines separating the Ag and ZnS films.

The field amplitudes on the symmetry axis are plotted in Figure 3.12 for five wavelengths. At the short wavelength peak of the group index the field is localized outside the metal films. The largest reported field localization occurs at the long wavelength peak in the group index. The field profile through the metals has a relative minimum in the metals to avoid excessive absorption losses in the structure. The field penetration for wavelengths 650 nm and 680 nm is comparable with an overall edge at the 650 nm wavelength.
Figure 3.12: The electric field amplitude squared versus position in the sample MDS2. The vertical lines mark the interfaces between the ZnS and Ag films [33].

The Z-scan simulation of MDS2 is shown in Figure 3.13 for five different wavelengths. The irradiance at the focal plane was the same as applied in our simulations of MDS1. The weakest nonlinear effect was observed at the short wavelength edge of the transmission band. The 650 nm wavelength shows the strongest nonlinear effects and its value does not correspond to a peak in the group index or an absorption maximum. The smaller value of $\beta_2$ for Ag leads to a smaller minimum in the normalized transmittance curves. The results correlate nicely with the field localization in the metal films. Namely, higher localization means larger nonlinear changes.
Figure 3.13: Open Aperture Z-scan Curves for MDS2. The position of the sample is normalized by the Rayleigh range, $z_0$ [33].

### 3.6 Chapter Summary

Using FEM numerical simulations we studied the open aperture Z-scan results of nonlinear absorption in metallodielectrics. The localization of the field in the metal region always corresponded to a greater nonlinear effect. The group index or absorption maximum used as an indicator of the most effective nonlinear wavelengths was not reliable for both cases we studied. The simulations used a FEM technique to characterize the nonlinear transmission properties of 1D metallic PBG structures with realistic material parameters. Our method incorporates the transverse beam profile, interference and diffraction effects.

The accuracy of the simulations was verified using TMM techniques and analytical results. Our purely refractive nonlinear results agree very well with the Z-scan
theory and we conclude that the FEM technique is a faithful model for such systems where back reflections and resonances play an important role in the linear and nonlinear processes. This method applies equally well for multi-layer stacks made from purely dielectric materials. It can be adapted to study nonlinear optical properties of higher-dimensional photonic crystals.

With our FEM tool we can consider this very interesting physics of photonic crystals that includes open and closed aperture Z-scan geometries. Besides potential broad-band optical limiter applications [48] metallodielectric stacks have potential for super-resolving imaging properties [7, 27, 33]. The FEM technique can be applied to these problems, as well, resolves the very memory intensive problem, by assuming axial symmetry scaling down the dimension of the problem from three to two.

We included the nonlinear refractive indices of the dielectric materials in our simulations; however, for the examples in this paper the two photon absorption coefficients of the metals dominated the nonlinear optical properties in these simulations. These results are a prelude to future experimental investigations of the nonlinear optical properties of MDSs. In the future we will apply realistic input beam characteristics, including pulse operation, and we will use experimentally derived material properties to provide a direct comparison with experimental results [33].
CHAPTER 4
EXPERIMENTAL Z-SCAN CHARACTERIZATION OF
THIRD-ORDER NONLINEARITIES IN MDS

4.1 Introduction

This chapter is dedicated to experimental determination of the effective nonlinear absorption coefficients \( \beta_{\text{eff}} \) of Metallodielectric stacks (MDS). The technique used to determine the nonlinear absorption of MDS is the Z-scan technique which was discussed in Chapter 3. In order to observe and characterize the nonlinear optical properties of MDS with the Z-scan technique some careful steps have to be taken. It will be worthwhile to point out here that femtosecond pulses from an amplifier laser were used. This is different from the CW beams considered in the numerical calculations of Chapter 3. The overall governing physics however doesn’t change very much since the nonlinear materials we are considering are very thin, so the interaction time is very short compared to the pulse width, and we can treat it as a series of time slices where the field amplitude could reasonably be assumed to be a constant within a slice.
4.2 Fabrication of Metallodielectric Stacks

The MDS used in our experiment were fabricated by RF sputter deposition method in a class 1000 clean room by Dr. Andrew Sarangan and Mr. Jian Gao. RF sputtering is a popular physical deposition method to fabricate thin films. Sputtering involves bombarding a target of bulk material with high energy ions, thereby releasing atoms of ejected material which settles on a substrate forming a thin film. The sputter deposition technique is a reliable one for the thin film deposition, under favorable conditions it yields good quality thin films in terms of uniformity and surface roughness compared to other thin film deposition technique such as electron beam evaporation.

There are commonly 2 types of sputter deposition techniques. They are direct current (DC) sputtering and alternating current modulated at radiofrequency (RF) sputtering. The DC sputtering is used only for conducting material, while RF sputtering is good for both insulators and conductors such the metals in our work.

In RF sputtering, the charge build-up on insulating targets can be avoided when the sign of the anode-cathode bias is varied at a high rate. RF sputtering yields highly insulating films but only with the added expense of RF power supplies and impedance matching networks. Stray magnetic fields leaking from ferromagnetic targets also disturb the sputtering process. Specially designed sputter guns with unusually strong permanent magnets must often be used for compensating magnetic field aberrations. The physical mechanism of sputtering involves the use of energetic ions in plasma to knock out atoms of a bulk material known as the target thereby releasing these atoms to form thin films on a substrate. This process takes place in a well regulated gas filled chamber, which is fed
by electrodes: the cathode and the anode. The target material is always attached to the cathode. When the voltage between the cathode and anode is high enough it causes a breakdown of gas in the chamber thereby creating plasma of ions. The gas of choice in most chambers is Argon; so Argon ions are created which are heavy and energetic enough to knock out target atoms when they are attracted to the cathode. RF magnetron sputtering uses strong electric and magnetic fields to trap electrons close to the surface of the magnetron, which is known is a target. The trajectories of electrons are helical paths around magnetic field lines thereby undergoing more ionizing collisions with gaseous neutrals near the target surface. Excess argon ions are created by this ionizing process. The plasma can be sustained at a lower pressure.

The sputtered atoms, knocked out from the target are neutrally charged and are unaffected by the magnetic trap. Sputtered films are often porous and are not very resistant to stress. Such films could be annealed (heated) at high temperatures such that the grains could reflow and settle to form a much harder, uniform and less porous film. The annealing process should be a carefully controlled one. In the case of multilayer structure of a metallodielectric, annealing can cause the metal to diffuse into the dielectric forming a composite. In RF Sputtering for the short periods of time when the electrode is positive, the electrode will attract electrons, however because this process is short-lived, it cannot cause a depletion of electrons in the plasma [49, 50].

In our work three different MDSs were fabricated by RF sputtering and used in our nonlinear optics experiments. We have Stack 1, made up Ag and ZnS. This is the MDS which we have investigated best, because of its well understood properties. I
proposed the design, and its fabrication and characterization steps were carefully followed through, for good agreement with numerical simulations.

For the fabrication of **Stack 1** the deposition rate of ZnS was 0.09nm/s and that for silver was 0.03nm/s. The tooling factor $TF$ which is an important parameter in fabrication defined as: $TF = TF_p (th_m / th_r)$, where $TF_p$ is the initial estimated tooling factor, usually it is the tooling factor for the previous deposition of that material, $th_m$ and $th_r$ are the measured thickness and thickness read from the instrument respectively. The tooling factor for the deposition of ZnS layers, and the Ag layers was 253 and 168 respectively. The RF power at the electrodes for ZnS and Ag depositions was 100W and 20 W respectively. The work pressure was 4mTorr.

**Stack 1** consist of 7 layers, distributed as ZnS (35nm)|[Ag (35nm)/ZnS (85nm)]$_{2.5}$|ZnS(35nm) and **Stack 2**, made up Ag and SiO$_2$, with a total of 8 layers that is 4 periods of Ag (20nm)|SiO$_2$(150nm). This sample was an older sample which was fabricated several years ago for other applications. This sample has been damage on so many spots, since I used so often as a practice sample and had several accidents with as I struggled to acquire mastery in my work. Nevertheless it has a pretty high transmittance and we could still locate some good spots on which we could focus a beam for our experiment.

**Stack 3** is a Cu and ZnS MDS, having 9 layers in total distributed as follows (ZnS (20nm)|[Cu (20nm )/ZnS (80nm)]$_{3.5}$|ZnS(20nm)). For the fabrication process of stack3, the tooling factor for the deposition of ZnS layers, and the Cu layers was 253 and 200
respectively. The RF power at the electrodes for ZnS and Cu depositions was 80W and 100 W respectively. The work pressure was 4mTorr. The deposition rate of ZnS was 0.12nm/s, and that of Cu was 0.08nm/s. There were several setbacks with this sample especially because it was our first time working with Cu. The fabricated sample showed significantly lower transmittance than the predicted numerical model. It also had a lot of pin holes which could affect the experimental data.
4.3 Experimental Set-up

The set-up of the main experiment, discussed in this chapter is shown in Figure 4.1 below.

Figure 4.1: Z-scan setup, for third order nonlinear measurements on metallodielectric stacks. F1 = filter 1, F2=filter2, F3= filter 3, M1=Mirror 1, M2=Mirror 2, M3=Mirror 3, L1=Lens 1, L2=Lens 2, BBO=Nonlinear Crystal, BS = Beam Splitter, FD1= filter/detector 1, FD2=filter/detector 2 and MS= motorized stage with the sample.

Shown in Figure 4.1, is a schematic of our experimental, ultrafast-laser setup. The pump laser is a Ti-Sapphire laser, Spectra Physics Tsunami, producing pulses 35fs to 150fs, with a repetition rate of 80MHz. The beam average power is about 750mW. The output wavelength is 790-810nm. The Spitfire Pro is an optical regenerative amplifier with a
lower repetition rate. Its standard repetition rate is 1 kHz. This amplifier accepts pulses with pulse widths less than 115fs and can produce pulses with pulse energies greater than 1mJ. The TOPAS (Traveling-wave optical parametric amplifier of white light continuum) is a wavelength converter. At 1 kHz, it sends out pulses of about 20-300fs, with wavelengths in the 1100nm to 1400nm. Its pump energy can be as high as 5mJ.

There are several filters (Corning glass color filters) in the setup. The first filter removes 800nm pump light which is co-propagating with the IR light produced by the TOPAS parametric amplifier. The second filter absorbs the IR beam after a second-harmonic signal is generated by the Beta barium borate (BBO) crystal. Mirrors M1, M2 and M3 are used to reflect the beam and direct it to the nonlinear crystal, the sample or the reference power detector. The beam Splitter sends path of the beam to M3, which becomes the reference arm. The rest of the beam on the signal arm is focused by L1 a 75mm lens onto a MDS sample on a motorized stage. The Lenses L2 (focal length 75mm) and L3 focus the signal and reference beams onto a filter/detector 1 and filter/detector 2 respectively. It is important to introduce these filters before the detector, to prevent oversaturation. The electrical signals generated at the detectors are carried are modified by resistor circuits before they are sent to the computer via cables. The data is analyzed at the computer with a Labview program. The program also keeps track of the position of the sample on the motorized stage. The motorized stage is controlled by a micro-controller which is also linked to the computer.
4.4 Beam Characteristics

Femtosecond pulses are sent from the Tsunami laser, to the Spitfire which amplifies the pulses. The amplified pulses are received by the Topas which down-converts them to infrared wavelengths. The BBO crystal converts the IR pulses by second harmonic generation to visible pulses. The wavelength of operation of our system is selected by tuning the Topas laser and adjusting the BBO crystal for phase matching. The wavelength is measured using a spectrometer. The beam spot size is determined by taking a knife edge transverse and vertical scans and by taking the images of beam at various positions after the focusing lens. Both methods yield very similar results.

4.4.1 Beam Spot Size Determination

An experimental set-up in which transverse cuts are made through the beam, using an opaque object while measuring the power of the unblocked portion of the beam on the detector is known as the knife edge experiment. A plot of the power measured versus the position of the “knife edge” contains information about the beam spot size at that particular location. For example in Figure 4.2, normalized power is plotted against a transverse horizontal distance in the x direction. The data is fit to an error function and the beam waist is determined. The wavelength of light is measured with a spectrometer to be 592nm. The transverse knife edge is done in both the vertical (y) and horizontal directions so that any asymmetry in the beam could be determined. The result can be expressed in terms of a beam quality factor, called $M^2$. It is important that the beam should be approximately Gaussian, i.e. $M^2 \sim 1$, for us to reliable measurements in our Z-scan experiment.
The results of the knife edge experiment on our beams showed that the beam is approximately Gaussian since the beam qualities valued obtained are less than 1.5. The beam spot size was also estimated using Camera images of the beam spot size at focus. Both measurements give a similar value of 40 microns for wavelength of 592nm.

Figure 4.2: Transverse Knife edge x direction result showing power profile at a distance of 12 cm from focus of the beam.
Figure 4.3: Shows both the x and y radius of the beam at several z positions. These data are fitted according to equation 4.1 and the quality factor of the beam is calculated as a fit value of $M_x$ or $M_y$.

$$w_{x,y} = w_{0x,y} \sqrt{1 + M_{x,y} \frac{(z - z_f)^2}{z_0^2}}.$$  \hspace{1cm} (4.1)

In the equation above $z_f =$ focal length of lens $L_1=75$mm. The quality factor $M^2 = M_x^2$ or $M_y^2 = 1.1^2 = 1.21$. To estimate the beam spot size with the Camera (COHU, High performance CCD Camera), first we determined the pixel pitch of the Camera. An attenuated He-Ne laser 632nm lase beam illuminates the Camera, which is placed on a translation stage. The Camera was moved in the x and y directions, while capturing images of the beam. The position of the centroid of the images is divided by the distance moved by the Camera, to give the pixel pitch of Camera in each direction. This pixel pitch was found to be $9.83\mu m(x) \times 13.29\mu m(y)$. To measure the beam the beam
spot size for the Z-scan experiment, first we aligned the system according to the
experimental set-up shown in Figure 4.1, with few modifications. We take out the sample
and introduced the Camera just after the focus of the Lens L1.

The wavelength of the light is read with the Ocean Optics Spectrometer, by
placing its detector on the beam. For this particular case. The system was tuned and
wavelength was 592nm. With the Camera was placed around the focus of the beam with
the beam significantly attenuated with 3D filter. Then images of the beam were taken.
The image of the beam was saved as 240 by 300 matrix. From this matrix the row and
column of highest intensity was located. In performing the matrix operations noisy signal
levels were substracted in order to facilitate direct comparison with numerical fits. Then
the intensity along the row (x) and column(y) was plotted and was compared to a fitted.
From this fit we can get a good estimate of the spot size. According to these
measurements the beam spot size in the vertical axis (y direction) was 40.38microns, and
beam spot size in the horizontal (x direction) was 36 microns. The plots of intensity and
the fits are shown in Figure 4.4 below.
Figure 4.4: (Left) Intensity versus pixel position in x direction. (Right) Intensity versus pixel position in y direction.

From these measurements we see that both techniques gave us very similar values of the beam spot size in both directions. Even though the beam is not a perfect Gaussian, the eccentricity of the ellipse describing the beam is close to zero. So we can rightly approximate the beam as Gaussian, which will validate our later Z-scan measurements.

4.4.2 Beam Pulse Width

The pulse width of the beam was estimated using a real time short pulse measuring device known as VideoFROG. For this measurement the output of the TOPAS was about 1400nm, which was doubled by the crystal to a wavelength of 700nm. The minimum wavelength the VideoFROG instrument could detect was 700nm, so we can used it to measure the Pulse width of a 700 nm beam.
Even though this wavelength was not of interest in our Z-scan experiments, yet we can reasonably assume that the pulse width doesn't change very much when the wavelength is tuned down to the 570nm-680nm range which is the range where most of our Z-scan measurements were performed. From these measurements we estimated the pulse width to be about 100fs.

4.5 Z-scan Experiments

We performed open aperture Z-scan experiments for several MDSs at several wavelengths. Our experimental set up is shown in Figure 4.1 above. We used the fundamental output from the TOPAS laser which is IR light. The filter F1 in our setup (Figure 4.1) removed the light at 800nm, which also passes through the TOPAS. We introduced the filter F2 to eliminate extra IR light and to ensure that only the Fundamental IR light is seen by the crystal. For our experiments the fundamental IR light is usually tuned from 1130nm to 1400nm. In the crystal the fundamental IR light is doubled by the nonlinear BBO crystal by a second harmonic generation process. The filter F3 filters out the fundamental IR light in order for us to ensure that only the second harmonic visible beam is used for nonlinear characterization. With a 75 mm lens L1, the beam is focused onto the sample which sits on a motorized translation stage. The sample is translated through the focus of the beam about a distance of 7-9cm. In order to improve signal to noise ratio it was expedient to split the beam, such that one beam serves as a reference beam.

FD1 is filter/detector combination that collects light from the signal arm, where the beam interacts with the sample. The other filter/ detector combination FD2 collects
light from the reference arm. The photocurrents generated by these detectors are converted to voltages which can be seen on a computer screen with aid of a Labview program.

Before acquiring data, there are some essential calibration steps to check. A good alignment of the system results in getting optimum amount of light to both the signal and reference detectors. A balance detection technique was been crucial in achieving meaningful results with very lower signal to noise ratio. The voltage signals at each detector are displayed on the computer screen and one obviously checks for linear relationship between these two signals.

Another good check is to run the data acquisition program after the alignment, without the sample and its movement. The program spits out the background signal/reference ratio. We normally look for a smooth background with a low signal/reference ratio. The system is aligned and the linearity of the filters is checked by looking at the voltage traces, on the computer screen. The background signal/reference ratio after a 100 time step is also read. Typical a good range of values that give a good background is often between 0.35 -0.60 percent. This step usually helps in the alignment process too. Because if the alignment is poor and the detectors are not located at the focal points it is impossible to have a low signal/reference values.

Before drawing conclusions about our results we had to investigate the sources and possibility of erroneous data. The first one we observed was sample damage. All the samples we used had a damage threshold. Therefore if high intensity beam impinges on it, it might produce a hole, which we could mistakenly read as a strong
nonlinear signal. This situation was observed when a 100 nm thick Cu layer deposited on glass substrate was exposed to the focused beam with an average power of 3.0 mW. Also putting a screen behind the sample proved to be a good check on the experiment, since on the screen we could look for a diffraction pattern, which would occur when the beam is drilling a hole in the sample. For this reason all of the reported results were performed at average laser power below 3.0 mW. The other concern was thermal blooming which is usually critical for measurements of the nonlinear refractive index, when high repetition rate and ultrafast lasers are used, as in our experiment. It is generally considered unavoidable, when Ti-sapphire lasers with repetition rate of 100 MHz to 1 kHz are used. The standard repetition rate of our laser is 1 kHz, just at the limit of the above mentioned interval [48].

In order to check if thermal blooming was could significantly falsify our data. A Z-scan experiment was performed at 671 nm when the repetition rate was 1 kHz; the average power at focus was about 0.80 mW. A second measurement was done when the repetition rate was reduced to 100Hz. The results of these measurements are shown in the Figure 4.5 below.
When the repetition rate was reduced to 100Hz and a new set of Z-scan data, was obtained there was no significantly noticeable difference in the transmission change as seen in Figure 4.5 above. This should indicate that, thermal blooming was not playing any strong role in modifying the signal data. There are other intrinsic thermal effects for the metals which are expected and unavoidable such as Fermi smearing, and thermo-optic index changes [8, 9, 52]. We will discuss those in the later sections of this chapter.

4.6 Experimental Results

4.6.1 Ag/ZnS MDS

The open aperture Z-scan experimental results yields very interesting results at the visible wavelengths considered. Interestingly we notice that the nonlinear absorption coefficient is high at high transmittance and absorbance values. Figure 4.6 below shows the transmittance and absorbance of a Ag/ZnS MDS with structure: Ag/ZnS [ZnS (35nm)| [Ag (35nm)/ZnS (85nm)]_{3.5}| ZnS (35nm) ].
Figure 4.6 (Left): Transmittance (Right) Absorbance spectrum of Stack 1 (Ag/ZnS).

Figure 4.7: The electric field amplitude squared versus position in the sample Stack 1. The vertical lines mark the interfaces between the ZnS and Ag films.
Experimental Z-scan results for Stack 1 is shown in Figures 4.8-Figure 4.12 for five different wavelengths. The on axis intensity of the laser beam for the Z scan experiment with Stack 1 was $I_0 = 159.61 \text{GW/cm}^2$.

We saw in Chapter 3 that two main factors critically affect the nonlinear absorption [33].

1) Field Confinement in the metallic layers.

2) The overall transmittance of the stack at a particular wavelength.

Open aperture Z-scan results are shown below for 5 different wavelengths. The data obtained is fitted according to Eq. (4.1), which is similar equation to Eq. (3.6); only in this case we are describing the total nonlinear absorption of the MDS by an effective value, since the MDS is not homogenous. The linear absorption of the MDS is known, so the effective length $L_{\text{eff}}$ is easily computed. By selecting an appropriate value of $\beta_{\text{eff}}$ we can fit the experimental data to a curve. On the curves below $\beta_{\text{eff}}$ is simply represented as TPA (Two-photon absorption). Even though we use the “two photon absorption” to describe these systems, we it loosely, bearing in mind that there are other physical processes which are occurring.

Two-photon absorption affects the open-aperture transmission and the following result is found:

$$T(z) = \sum_{m=0}^{\infty} \frac{(-\beta_{\text{eff}} I_0 L_{\text{eff}})^m}{(1 + z^2 / z_0^2)^m (m+1)^{3/2}}.$$  \hspace{1cm} (4.1)
Eq (4.1) was derived using the Gaussian beam decomposition is usually used to determine the nonlinear absorption coefficients of material. It is equally applied for both CW and Pulsed beams [41].

Figure 4.8: Open Aperture Z-scan Curve for Stack 1. The incident wavelength is 581nm and the experimental data is fitted to a curve in order to obtain the nonlinear absorption coefficient.

TPA=130cm/GW @581nm
Figure 4.9: Open Aperture Z-scan Curve for Stack 1. The incident wavelength is 600 nm and the experimental data is fitted to a curve in order to obtain the nonlinear absorption coefficient.

Figure 4.10: Open Aperture Z-scan Curve for Stack 1. The incident wavelength is 629 nm and the experimental data is fitted to a curve in order to obtain the nonlinear absorption coefficient.
Figure 4.11: Open Aperture Z-scan Curve for Stack 1. The incident wavelength is 661nm and the experimental data is fitted to a curve in order to obtain the nonlinear absorption coefficient.

Figure 4.12: Open Aperture Z-scan Curve for Stack 1. The incident wavelength is 671nm and the experimental data is fitted to a curve in order to obtain the nonlinear absorption coefficient.
The nonlinear absorption is strongest at 661nm when the absorbance is very high with \( \beta_{\text{eff}} = 146 \text{cm} / \text{GW} \) and the electric field is strongly confined in the Ag layers, especially the second Ag layer. At 671 nm the nonlinear absorption is \( \beta_{\text{eff}} = 144 \text{cm} / \text{GW} \), we notice very similar characteristics to the case at 661nm. For these first two cases we see that the strong field confinement and the high absorbance values are responsible for the overall strong nonlinear absorption. The transmittance for these cases is small. We also notice that at 629 nm the nonlinear absorption is high with \( \beta_{\text{eff}} = 140 \text{cm} / \text{GW} \). There is also strong field confine in the Ag layers in this case.

Even though the transmittance of the stack is highest at 581nm, the stack shows less nonlinear absorption compared to the cases, when the wavelength is 629nm, 661nm and 671nm, with an effective nonlinear absorption of \( \beta_{\text{eff}} = 130 \text{cm} / \text{GW} \). There reason for this is evident, because the stack at 581nm shows a small confinement of the electric field within the Ag layers. The value of the nonlinear absorption at 600 nm, is \( \beta_{\text{eff}} = 120 \text{cm} / \text{GW} \) the lowest value for all five wavelengths, this value is smaller than the value of the nonlinear absorption at 581 nm. The field confinement within the Ag layers at 600nm is also small and its absorbance and transmittance is less, compared to 581nm. For all the 5 wavelengths considered in this study the absorbance is least at 600nm. This result confirms some facts pointed out by Daniel Owens in [17] that the transmittance and absorbance will determine to a great degree the nonlinear enhancements observed in MDS. The weakest nonlinear effect was observed at 600 nm. Even though the field concentration within the metallic layers is looks higher at 600nm than at 581nm, yet because the overall transmittance is higher at 581nm, this seems to increase its
nonlinearity by a small amount. Nevertheless the measured nonlinear absorption is
generally high at wavelengths where the fields are highly confined in the metallic layers.
We conclude that these 3 competing quantities (electric field confinement in metallic
layers, transmittance and absorbance will determine the overall nonlinear absorption
process).

4.6.2 Ag/SiO$_2$ MDS

A second MDS considered in our study is an Ag/ SiO$_2$, which we denote as Stack
2. It is composed of 4 periods Ag (20nm)/ SiO$_2$ (150nm) deposited on a glass slide. This
stack showed very significantly high transmittance and high nonlinear absorption at
wavelengths in the range (620nm-660nm). Its experimental and numerical transmission
spectrum is showed in Figure 4.13. We notice that there are resonances at certain
wavelengths; such resonances are typical of a symmetric design of MDS as opposed to
non symmetric designs discussed in this work typically the transparent metal design
which has apodized layers of dielectric at the beginning and end of the stack which has a
play a role of broadening the transmission spectrum. A detailed discussion of the various
designs of MDS and their transmission and reflection properties is discussed in [23]. The
average beam power for the Z-scan experiment is appears in a box within the Figure as
“Pav” with the effective two-photon absorption coefficient using the fit from Eq. (4.1).
Figure 4.13: Experimental/ Numerical Transmittance for [Ag (20nm)/SiO₂(150nm)]₄ MDS.

Figure 4.14: The electric field amplitude squared versus position in the sample Stack 2. The vertical lines mark the interfaces between the SiO₂ and Ag films.
Figure 4.15: Open Aperture Z-scan Curve for Stack 2. The incident wavelength is 638 nm and the experimental data is fitted to a curve in order to obtain the nonlinear absorption coefficient.

Figure 4.16: Open Aperture Z-scan Curve for Stack 2. The incident wavelength is 658 nm and the experimental data is fitted to a curve in order to obtain the nonlinear absorption coefficient.
The experimental transmission closely follows the numerical predictions. The transmittance is higher at 638nm than at 658nm, but the field penetration in the metallic layer is higher for 658nm. From the experiments with Stack 2, we observed that the nonlinear absorption increased with an increase in intensity.

4.6.3 Cu/ZnS MDS

The third MDS investigated in our study is a Cu/ZnS MDS. The design is made of 9 layers of thin films deposited on a glass substrate, \((\text{ZnS (20nm)} | \text{Cu (20nm)/ZnS (80nm)} | \text{ZnS(20nm)})\). This sample was difficult to study because, the sputtered material did not settle uniformly on the substrate, leaving pinholes throughout some portions of the sample. So it was very necessary during the transmission and Z-scan measurements to avoid irradiating a pin hole. The experimental transmission spectrum of Stack 3 is shown below in Figure 4.17 along with our numerical result; we notice a large discrepancy in the correspondence between the two results. Clearly more research is needed to fabricate samples with these materials. Nevertheless, the experimental result from the Z-scan measurements confirms once more, that the nonlinear absorption of Cu based MDS could be very high. At 668nm the transmittance of the stack is about 23.55%.
Figure 4.17: Transmission spectrum for Stack 3 (Cu/ZnS).

For Stack 3, the ZnS/Cu MDS, an open aperture Z-scan was performed at 668nm. The average power of the beam read by the detector was 0.35mW. Therefore the on axis intensity was 69.96GW/cm$^2$. The plot of the experimental Z-scan and the fit is shown in Figure 4.18 below. The effective two photon absorption for this sample is

$$\beta_{\text{eff}} = 240\text{cm} / \text{GW}.$$
The Z-scan results for stack 3 do not show very high nonlinear absorption values as seen in literature [8, 34]. Yet the values are larger than those of the Ag based MDSs even for a lot lower on-axis intensity.

4.7 Simulation of Pulses

In Chapter 3, we discussed our novel FEM based, Z-scan technique which we used to study two different MDS designs. Our analysis however was based on a CW propagation which is different from the pulsed laser used in the experiments discussed in this chapter.

Figure 4.19: Schematic of time dependent amplitude of an electric field.
In order to theoretically and numerically track our current experimental measurements, a similar FEM based Z-scan simulation was carried out with the FEM tools of COMSOL multiphysics linked to MATLAB. Figure 4.19 shows our approach to simulating pulses in COMSOL; we perform a temporal discretization of the pulse into time steps and treat each step as a CW wave. This approach is valid when the pulse width is much larger than the sample length, as is the case in our experiments. Experimentally we have seen that is possible to determine the effective nonlinear absorption coefficient of the stacks, but to determine the contributions for each of its constituent materials is almost impossible. With our FEM Z-scan technique we can estimate the individual contribution of the metallic layers, since we know by theory that they are the main contributor to $\beta_{\text{eff}}$.

Some simplifications were introduced in the COMSOL simulations in order to save computer memory and facilitate calculations.

i) The MDS stack is grown on a 1mm thick glass substrate; we usually omit this layer even in Transmittance measurements. In our simulations we assume all the nonlinear activity is concentrated in the MDS. It is rewarding to neglect the substrate layer since it is exorbitant in size compared to the MDS wherein lies our nonlinearity. Including it will just make the problem extremely cumbersome and difficult to visualize in the graphical user interface.

ii) In the experiment there is a lens which focuses the beam after beam to the detector. This lens is also omitted in the simulation. Including this step could also be costly on our memory. Nevertheless it is possible to include it. This
will require a propagating field at the exit layer of the MDS to the lens, performing a Fourier transform at the lens and propagating to the detector to measure the irradiance. Our simulations are based on the set-up shown in Figure 3.1 with an open aperture to capture all the transmitted irradiance. The output field at the last layer is propagated directed to the detector using a Fourier (Bessel) transform, where the irradiance is calculated.

The computational time to calculate the transmittance for a single \( z \) value is about 5 hours. Without these simplifications, it would have taken a longer time to simulate the pulses.

![Z-Scan @ 581nm](image)

**Figure 4.20:** Open aperture Z-scan at 581nm for Stack 1. The red circles are the experimental data. The blue line is the numerical fit of the experimental data. The black line is the FEM simulation result of the Z-scan experiment, when 

\[
\beta_{2Ag}^2 = 40\text{cm/GW}.
\]

We see from the above result at 581 nm that, FEM simulation for this pulsed case can be adapted to match significantly the experimental Z-scan, if an appropriate fit for nonlinear coefficient of Ag is chosen. We can also compare the effective values of the
nonlinear absorption coefficients obtained in Chapter 3 for MDS2, which is an almost identical MDS design to Stack 1 studied in Chapter 4. We should recall that FEM based numerical simulation for this stack was performed when the on axis intensity \( I_0 = 47.74 \text{GW/cm}^2 \), with a CW laser signal. The experimental measurements was performed at a much higher intensity of \( I_0 = 159.6 \text{GW/cm}^2 \), with a pulsed laser. If we fit the data of Figure 3.12 according to Eq. (4.1) and calculate effective nonlinear absorption coefficients we obtain the following, values: [10cm/GW, 105cm/GW, 50cm/GW, 230cm/GW, 36cm/GW] for the wavelengths [510nm, 550nm, 600nm, 650nm, 680nm]. We have plotted this result together with the experimental result obtained in Chapter 4 in Figure 4.21 below.

![Figure 4.21: The nonlinear absorption coefficient versus wavelength is plotted for Stack 1.](image)

There are several reasons that could explain the discrepancies between the experimental and numerical results. The fact that we are comparing CW propagation to
pulsed propagation is a first concern, since for a pulse excitation the highest irradiance is sustained for only a short period and lower irradiance values are averaged in the signal.

Also from some theoretical calculations and comparison with other results, it should be obvious that intensity used in the experiments was really huge; this could probably turn on higher order effects such as fifth order susceptibility $\chi^{(5)}$ and higher orders. Finally, the thermal dynamics at high intensities could really change the optical properties in way that are unaccounted for in our models. The two temperature model (electron and lattice (phonon) temperatures) would introduce another complication that could affect the pulse dynamics. This effect is discussed in the next section.

4.8 Thermal Effects

Given that the electron temperature of such systems quickly rises from a few ten of degrees to thousands of degrees, thermal processes turn to play an important role is such Z-scan experiments. It is our goal to separate field induced transmission changes from thermally induced transmission changes. It is necessary for us to identify the sources of thermally induced transmission changes and see how much these have affected our results.

Owen et al enumerated at least five thermally driven processes that affect the permittivity of silver as mentioned in Chapter 3 [30]. The most significant of these processes is the Fermi smearing effect. However experimental work on the Fermi smearing has not been very convincing in providing a full physical picture of the phenomenon. The thermal energy in the Fermi smearing process is proportional to the square of the temperature of the metallic electron cloud. This process can change the
dielectric permittivity of metals and also generate nonlinear effects [43]. The other thermal processes 1D MDS include; thermal expansion due to the thermal expansion coefficient, change in the index of refraction due to the thermo-optic coefficient, change in the index of refraction due to the nonlinear refractive index $n_2$ and change in the plasma frequency of the metal $\omega_p$ in an expanded layer. The role of these processes in our experiments is negligible [9]. In order to limit the effect of thermally driven process especially the Fermi smearing process which is the most significant in our experiment, several measures can be taken.

i) Use a lower power laser.

This has the drawback of lowering the intensity of the laser will be the inability of accessing any nonlinearity. The fact that the pulses are so short does work in our favor because most of the pulse has passed through the sample before the thermal effects can affect the transmission.

ii) Use a lower repetition rates

Even though our experiments were carried out at high intensities, this was necessary in order to probe the nonlinearity of the materials. Several steps were taken to limit thermally driven process. We carried out our measurements at 1 kHz as opposed to experiments performed at MHz. Lippitz et al [9], performed a similar Z-scan experiment on a Ag/MgF$_2$ MDS at 76MHz and observed transmission changes of 30%-40%. We noticed a transmission changes from 15%-20% for Z-scan with the Ag/ZnS sample. We also accounted that the high transmission changes was directly related to the
iii) Use shorter pulses

Femtosecond and picosecond pulses are often preferred to probe such thermally sensitive systems compared to nanosecond pulses which will generally have higher fluence [48]. Real time measurements have shown that it takes longer it takes about one picosecond for the electron gas and the lattice to settle at thermal equilibrium. From our calculations we are attributing the nonlinearities observed to be intensity dependent $\chi^{(3)}$ rather than a change in complex refractive index which is a $\chi^{(1)}$ effect as proposed by Rotenberg et al in [53].

The Fermi smearing process, responsible for the increase in the absorption, should be explained in this case as a $\chi^{(3)}$ process. Several publications especially dedicated to the study the nonlinear response of metallic nanoparticles distributed in dielectric matrix have treated the hot electron effect as a $\chi^{(3)}$ process. On the other hand Rotenberg et al, who showed a pulse-width dependence of the effective nonlinear absorption of Au film claims the hot electron effect is purely a temperature-dependent change in the complex refractive index of the metal. When the change in complex permittivity was measured in [30] for pulse widths ranging from 0.5ps -5 ps, over wavelengths ranging from 500nm to 700nm only a very small values were obtained. I have used those numbers to simulate the pulse propagation in an Ag/ZnS MDS and found very small amplitude and phase changes, which will ultimately lead to small transmission changes. These observations
enable us to conclude that in the picoseconds regime, Fermi smearing contribution to \( \chi^{(1)} \) is small. If higher transmission changes are measured, it may be a thermal induced contribution to the third order and higher nonlinearities.

The two temperature model (TTM) is often used to model these systems, calculating the electron temperature \( T_e \), the lattice temperature \( T_l \) and the relaxation time \( \tau_r \). The electron temperature may rise to about 8000K, under conditions as in our experiment and then relaxes within a few picoseconds to the lattice temperature of 500K [9, 53]. The last step in the relaxation is the diffusion of heat out of the metallic layers into the other neighboring layers and the glass substrate. The equations to model the two temperatures is given below. From our estimation this diffusion may be of the order of milliseconds, close to the time in between the pulses.

\[
C_e \frac{\partial T_e}{\partial t} = k \nabla^2 T_e - g (T_e - T_l) + P(z, t) \quad ,
\]
\[
C_l \frac{\partial T_l}{\partial t} = g (T_e - T_l), P(z, t) = (1 - R) e^{-\alpha z} I(t) \quad ,
\]

where \( C_e \) and \( C_l \) are the electron and lattice heat capacities respectively, \( k \) is the conductivity of electrons, \( g \) is the electron-phonon coupling constant of the metal, \( P(z, t) \) is the heat power source term, \( R \) is the reflectance of the sample, and \( \alpha \) is the linear absorption coefficient.

The polarization \( \mathbf{P} \) can be function of electron temperature as in equation 4.4 below.

\[
\mathbf{P}(T_e) = \varepsilon_0 (\chi_r^{(1)}(T_e) - i\chi_r^{(1)}(T_e)) \mathbf{E} + \varepsilon_0 (\chi_r^{(3)}(T_e) - i\chi_r^{(3)}(T_e)) |\mathbf{E}|^2 \mathbf{E} .
\]
As the electrons of the metals get heated, they will cause increases in $\chi^{(1)}_c$ and $\chi^{(3)}_c$ depending on the pulse width of the irradiated light. When shorter pulses are used, such as femtosecond pulses, the instantaneous $\chi^{(3)}_c$ dominates over $\chi^{(1)}_c$ as the pulse width increases. By solving an analogous equation to Eq. 4.3 in cylindrical coordinates. We estimate that the temperature rise for 20nm thick layer of is about 200K if its initial temperature was 300K. We assume the input intensity was 160GW/cm$^2$, the same value as in Section 4.5.1.

We can write the change in temperature as:

$$\Delta T(r,t) = \frac{w^2}{4D_t + w^2} T(0) \exp\left(-\frac{r^2}{4D_t + w^2}\right),$$

(4.6)

where $w$ is the beam spot size; and $D_t = k_T / \rho C_p = 2 \times 10^{-4} m^2 / s$ is the thermal diffusion coefficient of silver. $k_T$, $\rho$, and $C_p$ are the conductivity, the density and the specific heat capacity of silver respectively and $T(0) = \int_{-\infty}^{\infty} \frac{\alpha I}{\rho C_p} dt$. The temperature drops exponentially, so in 1ms which is the time in between pulses for the laser system described above in section, the temperature will drop by 90%. This will results in heating, and cooling cycles as the film is bombarded by a series of pulses. This will certainly lead to memory effect which is unaccounted for in our models. Even though our intensities in the experiments were high, it becomes clear to us that the temperature rise in the lattice of the MDS does not change the dielectric properties by much. If the temperature changes
were exorbitant, material damage would have inevitable, and the results of the Z-scan would not be repeatable.

4.9 Chapter Summary

We have measured the nonlinear absorption coefficient of 3 MDSs. For the Ag/ZnS MDS we have shown how the nonlinear absorption depends on transmittance absorbance and the field penetration within the metallic layers of MDS. Our pulse energy for most measurements we performed with the Ag/ZnS samples was about $0.8 \mu J$. We believe that it is also possible to access the nonlinear refraction of the sample by increasing the on-axis intensity as in [54], where $7 \mu J$ pulses were used and nonlinear absorption and nonlinear refraction with a 50fs pulse at 800nm and obtained values of $-4.41 \times 10^{-8} \text{ cm/W }$ for $\beta_{\text{eff}}$ and $-2.95 \times 10^{-13} \text{ cm}^2/\text{W}$ for $n_{\text{eff}}$.

We have also numerically characterized the Z-scan experiment at a wavelength of 581nm with a FEM technique, and show how it agrees with experiments. We believe that the nonlinear enhancements observed can be significantly attributed to the intrinsic third order susceptibility of the metals. Our results prove that the nonlinearities observed cannot be totally attributed to thermal processes, which could be dominant in such systems.
CHAPTER 5

CONCLUSIONS AND FUTURE WORK

5.1 Outcomes

In this dissertation, we studied unusual optical properties of MDS designs. First, we numerically examined optical super-resolution of simple images and transmission properties of MDS samples. We found MDS design that should exhibit adequate super-resolving properties in the visible region. Thick metallic films are highly absorptive in the visible region, which poses a tough challenge to overcome. We demonstrated that we can design a MDS with significant transmission (more than 50%), which will overcome the diffraction limit of light and enable channeling of electromagnetic waves through sub-wavelength apertures thereby suppressing diffraction which in other materials would have been dominant. We also compared numerical techniques used in characterizing MDSs for transmission and super-resolution. Devices that could be fabricated from these MDSs could have applications in lithography and biomedical imaging.
We also fabricated MDSs and checked how the numerical transmission matches the experimental one. In order to push the resolution to desired smaller dimensions we realized that a high index dielectric with low loss was necessary. Our main designs have GaP dielectric and the high dielectric constant reduces diffraction and decreases the required film thickness. Nevertheless the deposition of low loss GaP films remains an ongoing process development and we used realistic values of dispersion properties of annealed GaP films that were measured by Jian Gao at the Nanofab Lab facility to calculate. With these realistic dielectric constant data we were able to demonstrate the super-resolving properties of several MDS designs.

To experimentally verify the super-resolving ability of this device, one needs to design a mask structure on which periodic sub-wavelength objects are inscribed. This part of the work has not been achieved, so we could not experimentally verify the super-resolving abilities of MDS. Nevertheless we have carefully analyzed the linear optics problem of imaging and propagation within MDS.

We analyzed the problem of beam propagation in MDSs and examined the properties that affect nonlinear enhancements in MDS. We developed a FEM based solution to beam propagation involving a 1D photonic band gap material such as a MDS. This problem which includes quantities such as back reflections, Fabry-Perot resonances, transverse beam profile that are often ignored other solutions. We saw from the results in Chapter 3 that the numerical Z-scan experiment on MDS at several wavelengths is analogous to the Z-scan experiments done at the same wavelength, when the distribution of the metallic layers is spaced periodically [8, 32-33]. We then developed a FEM based
Z-scan technique which works well for MDSs. Experimental results were also obtained for fabricated MDS which definitely proves high nonlinear absorption for MDS. In the process leading to our Z-scan simulations we compared the FEM and TMM technique for agreement in the linear regime. The Z-scan simulation was also compared with the analytical results for agreement. In our CW Z-scan simulation we showed that the nonlinear absorption of a MDS depends on the field penetration within the metallic layers, the transmittance of the MDS and its absorbance. We saw that at particular wavelengths where the field penetration is very small, the change in nonlinear transmission is very small. This case is similar to the case where a bulk metal is irradiated and because the transmission is very small and the field falls exponentially, the nonlinear change in transmission is small. But if the bulk metal is split into smaller layers and dielectric layers and placed in between the metallic layers. The field is enhanced and the transmission of the stack increases, the stack will exhibit a higher nonlinear absorption in this case. In our analysis, we were expecting to notice a particularly higher nonlinear absorption in the slow light regime that is at wavelengths where the group velocity index and the energy velocity index are high. But this was not the case except at higher wavelengths of the transmission band pass which could also show a resonance in group velocity index. The effective nonlinear absorption coefficients calculated from this Z-scan simulation had a large variance, with the highest value obtained at 650nm for the two MDSs (MDS1, MDS2) when the transmittance was high and the field penetration in metallic layers was strong.

In the experiments we considered the samples named Stack1 which had almost the same design as our case labeled MDS2; the effective nonlinear absorption coefficients
were measured at a much high intensity, for this case the variance in the effective nonlinear absorption coefficients was significantly smaller. The values however followed a similar pattern, where they tend to be higher for strong field confinement in the metallic layers, and high transmittance. This experiment was also performed with a pulsed laser at a repetition rate of 1 kHz. A pulsed simulation was also performed at 581 nm with the FEM, which agrees reasonably with the experimental result, for this simulation a fit was sought for using only nonlinear absorption values of the metal. The discrepancy between the theory developed for CW beams and the experimental results of the nonlinear absorption coefficients could be accounted for by several reasons, which could include the following; the different nature and quality for the beam, one is CW, the other is pulsed. In theory we assume a perfect Gaussian. There are other effects unaccounted for in our model such as memory effects, optical bistability, and the generation of higher harmonics since the intensity for the experiments was very high. There is a worry about the inability of standard Z-scan theory used for the analysis of systems with Fabry-Perot resonance such the ones addressed in this work as it was shown in [55]. There is therefore a need for a better model to analyze the effective nonlinear parameters for MDS.

5.2 Future Work

There are several interesting challenges that will make use of the results showed forth in this dissertation. The experimental super-resolution task could be pursued making use of the design and numerical characterization demonstrated in this work.
We can also pursue an extensive simulation of pulsed propagation in nonlinear MDS, which could help account for nonlinear refraction as well as nonlinear absorption and give a more faithful comparison with experimental results. We can also incorporate additional physical phenomena into our investigation investigate that will especially the influence of high intensity regime for our Z-scan experiments. For instance, a detailed examination of the underlying physics of the thermal processes that are generated when light is incident MDS, and the optical effects is still a field which has not been explored thoroughly. We made estimates of the thermal diffusion effects based on lattice (phonon) temperatures. A true multi-physics simulation of these effects would help to unravel the importance of various contributions. A simple temperature calculation of a high intensity CW beam propagating through the MDS (stack1) is shown in Figure 5.1 below. We see that the temperature rises to about 533K from an initial temperature of 300K.

Figure 5.1: Temperature distribution across MDS (Stack 1)

We note that the temperature is highest at the first Ag layer, as expected.
5.3 New Horizons

Engineering MDSs have already opened up new ways of applying metals in optics, specifically for applications in lithography and sensing. These devices have the potential to be used as antennas for both RF and optical signals. The research community is seeking robust and well adapted materials for optical limiting, in order to protect sensors [56, 57]. We believe a MDS can be designed and fabricated which may meet this demand. If the nonlinear refractive index of MDS is properly characterized, then they could also be explored as potential materials for optical switching [58]. The access of electromagnetic waves to the metal nonlinearity and the presence of multiple surfaces provide a mechanism for optical bistability second and third harmonic generation in the MDS samples [59, 60]. A future project devoted to examining the dynamics of harmonic generation could yield yet more surprising results. These UV signals were not detected in our experiments, but there is evidence from the literature that the harmonic and even higher harmonic signals will be generated with high efficiency.
REFERENCES


APPENDICES

A. MATLAB Code for Energy Velocity

```matlab
function zn=mtimesTEval(lambda)
tic
% program calculates transmission, absorption, Energy velocity for plane
% propagating in N layers films
% sketches the electric and magnetic field within all N layers
% Written by Nkorni Katte (January 2011)
n0=1;
nf=1;
fi0=(0/180)*pi;
i=sqrt(-1);
d=layerput %[17 22 35 22 35 22 35 22 35 22 35 22 17]
N=indexput %[ndi nm ndi nm ndi nm ndi nm ndi nm ndi nm ndi nm ndi]
numl=length(d)%eps=N.*N
```
rr=length(N)

for c=1:rr
    km(c)=2*pi*cos(0).*N(c)/lambda;
end

% Compute last refraction angle
pm=km;
alphaf= 0;
k0=n0*2*pi*cos(fi0)/lambda;
knf= nf*2*pi*cos(alphaf)/lambda;
zsum=0
for c=1:length(d)
    zr(c)=d(c)+zsum
    zsum=zr(c)
end
lp=zr(numl)

zr(numl+1)=lp;

z=zr

zf=z(length(z)-1)
zff=z(length(z))

M0=[exp(-i*k0*0) exp(i*k0*0);-i*k0*exp(-i*k0*0)/n0/n0 i*k0*exp(i*k0*0)/n0/n0];
\[
\begin{align*}
M_1 &= [\exp(-i \cdot km(1) \cdot 0) \ \exp(i \cdot km(1) \cdot 0); -i \cdot km(1) \cdot \exp(-i \cdot km(1) \cdot 0) \\
& \ \quad i \cdot km(1) \cdot \exp(i \cdot km(1) \cdot 0)];
\end{align*}
\]

\[
\begin{align*}
M_{\text{last}} &= [\exp(-i \cdot knf \cdot zf) \ \exp(i \cdot knf \cdot zf); -i \cdot knf \cdot \exp(-i \cdot knf \cdot zf) / nf / nf \\
& \ \quad i \cdot knf \cdot \exp(i \cdot knf \cdot zf) / nf / nf];
\end{align*}
\]

\[
\begin{align*}
M_{\text{next}} &= [\exp(-i \cdot km(rr) \cdot zf) \ \exp(i \cdot km(rr) \cdot zf); -i \cdot km(rr) \cdot \exp(-i \cdot km(rr) \cdot zf) \\
& \ \quad i \cdot km(rr) \cdot \exp(i \cdot km(rr) \cdot zf)];
\end{align*}
\]

\[
\begin{align*}
\text{check} &= \text{numl} \\
c &= \text{numl} - 1 \\
C &= [1 \ 0; 0 \ 1];
\end{align*}
\]

\[
\begin{align*}
Cl &= (M_{\text{last}} \setminus M_{\text{next}}) \\
Cf &= (M_1 \setminus M_0)
\end{align*}
\]

\[
\begin{align*}
\textbf{while} \ (c <= \text{check}) && (c >= 1) \\
f_{11} &= \exp(-i \cdot km(c) \cdot z(c)) \\
f_{12} &= \exp(i \cdot km(c) \cdot z(c)) \\
f_{21} &= -i \cdot km(c) \cdot \exp(-i \cdot km(c) \cdot z(c)) \\
f_{22} &= i \cdot km(c) \cdot \exp(i \cdot km(c) \cdot z(c)) \\
b_{11} &= \exp(-i \cdot km(c+1) \cdot z(c)) \\
b_{12} &= \exp(i \cdot km(c+1) \cdot z(c)) \\
b_{21} &= -i \cdot km(c+1) \cdot \exp(-i \cdot km(c+1) \cdot z(c)) \\
b_{22} &= i \cdot km(c+1) \cdot \exp(i \cdot km(c+1) \cdot z(c))
\end{align*}
\]

\[
\begin{align*}
M_f &= [f_{11} \ f_{12}; f_{21} \ f_{22}] \\
M_b &= [b_{11} \ b_{12}; b_{21} \ b_{22}] \\
M &= (M_b \setminus M_f);
\end{align*}
\]
\begin{verbatim}
C=C*M;
c=c-1,
end
P=C
Mt=(Mlast\Mnext)*P*(M1\M0)
R=abs(-Mt(2,1)/Mt(2,2))^2;
TT=abs(Mt(1,1)-Mt(1,2)*Mt(2,1)/Mt(2,2))^2
r=(-Mt(2,1)/Mt(2,2));
E0=[1;r];
E1=(M1\M0)*E0;
Ep=E1;
A(1)=E1(1,1);
B(1)=E1(2,1);
tt=1;
while (tt<=numl-1)
f11=exp(-i*km(tt)*z(tt));
f12=exp(i*km(tt)*z(tt));
f21=-i*km(tt)*exp(-i*km(tt)*z(tt));
f22=i*km(tt)*exp(i*km(tt)*z(tt));
b11=exp(-i*km(tt+1)*z(tt));
b12=exp(i*km(tt+1)*z(tt));
b21=-i*km(tt+1)*exp(-i*km(tt+1)*z(tt));
b22=i*km(tt+1)*exp(i*km(tt+1)*z(tt));
Mf=[f11 f12;f21 f22];
\end{verbatim}
Mb=[b11 b12; b21 b22];
M=(Mb\Mf);
E=M*Ep;
A(tt+1)=E(1,1);
B(tt+1)=E(2,1);
 tt=tt+1;
 Ep=E;
end
Elast=(Mlast\Mnext)*Ep;
cc=1
zm=0:1:zff
zstart=1;
zend=z(cc)-1;
na=1; na2=na*na;
 nb=1.42857; nb2=nb*nb;
 while (cc<=numl)
   if mod(cc,2)==0
      deva=nb2;
   else
      deva=na2;
   end
   for kk=zstart:zend
      Ef(kk)=A(cc)*exp(-i*km(cc).*zm(kk))+B(cc)*exp(i*km(cc).*zm(kk));
      Ue(kk)=deva.*abs(Ef(kk)).^2;
end
cc=cc+1;
zstart=z(cc-1);
zend=z(cc)-1;
end
zf=z(length(z)-1);
zff=z(length(z));

dd=length(zm);
Ef(dd-1)=A(numl)*exp(-i*km(numl).*zm(dd-1))+B(numl)*exp(i*km(numl).*zm(dd-1));
Ef(dd)=A(numl)*exp(-i*km(numl).*zm(dd))+B(numl)*exp(i*km(numl).*zm(dd));
Ue(dd-1)=nb2.*abs(Ef(dd-1)).^2;
Ue(dd)=nb2.*abs(Ef(dd)).^2;
Etl=Ef;
Hx=mtimesTM(lambda);

H2=abs(Hx).^2;

Sz=0.5*real(Ef.* conj(Hx));

% U is the energy per unit volume
% epsilon_0 =1 and mu_0=1

U=0.25*(H2+abs(Ue));

%12000=20*(250+350)

LL=100*(350+250);

th=0:1:LL;
Pl(1)=0;
deltah=0.5;
Ea(1)=0;

for rr=1:length(th)-1
    Pl(rr+1)=Pl(rr)+0.5*(Sz(rr)+Sz(rr+1)).*deltah;
    Ea(rr+1)=Ea(rr)+0.5*(U(rr)+U(rr+1)).*deltah;
end

PLL=Pl
Eaa=Ea

% Energy velocity V_E
V_E=(Pl(length(th))./Ea(length(th)));

zn=V_E;
B. MATLAB/COMSOL Code for Beam Propagation in MDS

function zz = ebeneamp

% COMSOL Multiphysics Model M-file

% Generated by COMSOL 3.5a (COMSOL 3.5.0.603, $Date: 2008/12/03 17:02:19 $)

% Some geometry objects are stored in a separate file.
% The name of this file is given by the variable 'flbinaryfile'.

flclear fem

% COMSOL version

clear vrsn

vrsn.name = 'COMSOL 3.5';

vrsn.ext = 'a';

vrsn.major = 0;

vrsn.build = 603;

vrsn.rcs = '$Name: $';

vrsn.date = '$Date: 2008/12/03 17:02:19 $';

fem.version = vrsn;

flbinaryfile='Ebenezermat.mphm';

% Constants

fem.const = {'sigma','50[um]', ...

'E0','1.09e9[V/m]', ...

'w0','40[um]', ...

'lambda0','661[nm]', ...
'z0','pi*w0^2/lambda0', ...
'k0','2*pi/lambda0', ...
't0','100[fs]', ...
'dt','100[fs]', ...
'xx','3[F*m/V^2]', ...
'n0m','0.1403-i*4.2195', ...
'n0di','2.2858-i*8.6099e-4'};  

% Geometry

clear draw

g32=flbinary('g32','draw',flbinaryfile);
g33=flbinary('g33','draw',flbinaryfile);
g34=flbinary('g34','draw',flbinaryfile);
g35=flbinary('g35','draw',flbinaryfile);
g36=flbinary('g36','draw',flbinaryfile);
g37=flbinary('g37','draw',flbinaryfile);
g38=flbinary('g38','draw',flbinaryfile);
g39=flbinary('g39','draw',flbinaryfile);

draw.s.objs = {g32,g33,g34,g35,g36,g37,g38,g39};

draw.s.name = {'R1','R2','R3','R4','R5','R6','R7','R8'};

draw.s.tags = {'g32','g33','g34','g35','g36','g37','g38','g39'};

fem.draw = draw;

fem.geom = geomcsg(fem);

fem.mesh = flbinary('m1','mesh',flbinaryfile);

% (Default values are not included)
% Application mode 1

clear appl

appl.mode.class = 'InPlaneWaves';

appl.dim = {'Ez2','Hz2','Ax2','Ay2','Aphi','scEz2','scHz2','psi2'};

appl.sdim = {'z','r','phi'};

appl.name = 'rfwe2';

appl.module = 'RF';

appl.assignsuffix = '_rfwe2';

clear prop

prop.inputvar = 'lambda';

clear weakconstr

weakconstr.value = 'off';

weakconstr.dim = {'lm4','lm5','tlmx4','tlmy4','lm6'};

prop.weakconstr = weakconstr;

appl.prop = prop;

clear bnd

bnd.type = {'H0','cont','SC','SC'};

bnd.E0 = {{0;0;0},{0;0;0},{0;0;Ein},{0;0;0}};

bnd.ind = [3,1,1,2,1,1,2,1,1,2,1,1,2,1,1,2,1,1,1,2,1,1,4];

appl.bnd = bnd;

clear equ

equ.epsilonr = {'erd','erm',1};

equ.ind = [1,2,1,2,1,2,1,3];

appl.equ = equ;
appl.var = {'lambda0',650e-9', ...
'E0iphi','exp(-j*k0_rfwe2*zm')};
fem.appl{1} = appl;
fem.sdim = {'z','r'};
fem.frame = {'ref'};
fem.border = 1;
clear units;
units.basesystem = 'SI';
fem.units = units;

% Boundary settings
clear bnd
bnd.ind = [1,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2,2];
bnd.dim = {'Ez2'};

% Boundary expressions
bnd.expr = {'E_bnd','w0*exp(-r^2/w^2)/w',"'};
fem.bnd = bnd;

v=5:5:100;

% Scalar expressions
fem.expr = {'w','w0*sqrt(1+(zm/z0)^2)', ...
'eta','atan(zm/z0)', ...
'R','(zm)*(1+(z0/zm)^2)', ...
'c0','1/sqrt(epsilon0_rfwe2*mu0_rfwe2)', ...
'omega0','2*pi*c0/lambda0', ...
'erdi','(n0di+(n2rdi-i*n2cdi)*Idi)^2', ...

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'erm', (n0m+(n2rm-i*n2cm)*Im)^2, ...
'Ein', E0*(w0/w)*exp(-r^2/w^2)*exp(i*0.5*k0*r^2/R)*exp(-i*eta)};

% Coupling variable elements

clear elemcpl

% Integration coupling variables

clear elem
elem.elem = 'elcplscalar';
elem.g = {'1'};
src = cell(1,1);
clear pnt
pnt.expr = {{},{{}},{{}},{{}}};
pnt.ipoints = {{},{{}},{{}},{{}}};
pnt.frame = {{'ref'},{'ref'},{'ref'},{'ref'}};
pnt.ind = {{'1','2','3','4','5','6','7','8','9','10','11','12','13',... 
    '14','15','16','17','18'}};
src{1} = {pnt,{{},{{}}};
elm.src = src;
geomdim = cell(1,1);
geomdim{1} = {};
elem.geomdim = geomdim;
elem.var = {'w_bnd','eta_bnd','R_bnd','Eout'};
elem.global = {'1','2','3','4'};
elem.maxvars = {};
elemcpl{1} = elem;
fem.elemcpl = elemcpl;

% Global expressions

fem.globalexpr = {'Im', 'real(n0m)*abs(Ez2)^2/(2*mu0_rfwe2*c0)', ...
'n2rm', '2e-11[cm^2/W]', ...
'n2cm', 'beta*lambda0/4/pi', ...
'zm', '1e-10*z0', ...
'beta', '3.5e-8[cm/W]', ...
'amp', 'abs(Ez2)', ...
'phazz', 'atan(imag(Ez2)/real(Ez2))', ...
'th', '400e-9[m]', ...
'n2cdi', 'betadi*lambda0/4/pi', ...
'rf', '1.0468e-17[m^2/W]', ...
'betadi', '3.4e-9[cm/W]', ...
'n2rdi', '0*1.33e-20[m^2/W]', ...
'Idi', 'real(n0di)*abs(Ez2)^2/(2*mu0_rfwe2*c0)', ...
'E_pulse', 'exp(-(t-t0)^2/dt^2)', ...
'hm', 'exp(-i*(omega0*t-k0*z))', ...
'te', '1[fs]'};

% Functions

clear fcns

fcns{1}.type='inline';
fcns{1}.name='getphase(Ez2)';
fcns{1}.expr='atan(imag(Ez2)/real(Ez2))';
fcns{1}.dexpr={'d(atan(imag(Ez2)/real(Ez2)),Ez2)'};
fcns{2}.type='inline';
fcns{2}.name='pulz(v)';
fcns{2}.expr='exp(-(v-100)^2/100^2)';
fcns{2}.dexpr={'d(exp(-(v-100)^2/100^2),v)'};
fem.functions = fcns;

% ODE Settings
clear ode

clear units;
units.basesystem = 'SI';
ode.units = units;
fem.ode=ode;

% Multiphysics
fem=multiphysics(fem);

% Extend mesh
fem.xmesh=meshextend(fem);

% Solve problem
fem.sol=femstatic(fem, ...
     'complexfun','on', ... 
     'solcomp', {'Ez2'}, ... 
     'outcomp', {'Ez2'}, ... 
     'blocksize','auto', ... 
     'callback','postcallback', ...
'callbparam', {'tridata', {'Ez2', 'cont', 'internal', 'unit', 'V/m'}, 'trimap', 'Rainbow', 'title', 'Surface:

% Save current fem structure for restart purposes

fem0 = fem;
%
% for cc=5:length(v)
%  Em(cc)=exp(-(v(cc)-100).^2/100^2);
%
% end

dr = 1e-6;
for rr = 1:121
    Amp(rr) = sc * postinterp(fem, 'amp', [-5.5e-8; 0 + dr * (rr - 1)]);
    Phaz(rr) = postinterp(fem, 'phazz', [-5.5e-8; 0 + dr * (rr - 1)]);
end
r = 0:dr:1.2e-4;
zz = Amp;
C. Derivation of Block Waves Dispersion Relation

Consider a simple periodic layered medium consist of two different materials with a
dielectric permittivity profile given by

\[
\varepsilon(z) = \begin{cases} 
\varepsilon_2 & 0 < z < d_2 \\
\varepsilon_1, & d_2 < z < d_1 + d_2 
\end{cases} \ .
\] (C.1)

If we incident a TM polarized field unto a this stack of material, we can write the write
the incident magnetic field as

\[
H(z) = \begin{cases} 
a_n \exp(-ik_1(z - nd)) + b_n \exp(+ik_1(z - nd)), & nd - d_1 < z < nd \\
c_n \exp(-ik_2(z - nd)) + d_n \exp(+ik_2(z - nd)), & (n - 1)d < z < nd - d_1 
\end{cases} \ .
\] (C.2)

By writing the continuity condition for Maxwell’s equation at the interfaces  \( z = (n - 1)d \)
and  \( z = nd - d_1 \), we get the following matrix equations.

In the matrix equations below \( n \) stands for the \( n^{th} \) unit cell and \( a_n, b_n, c_n \) and \( d_n \) are
constants which depend on one another.

\[
\begin{bmatrix} 1 & 1 \\ ik_1 / \varepsilon_1 & -ik_1 / \varepsilon_1 \end{bmatrix} \begin{bmatrix} a_{n-1} \\ b_{n-1} \end{bmatrix} = \begin{bmatrix} \exp(ik_2d_2) & \exp(-ik_2d_2) \\ ik_2 \exp(ik_2d_2) / \varepsilon_2 & -ik_2 \exp(ik_2d_2) / \varepsilon_2 \end{bmatrix} \begin{bmatrix} c_n \\ d_n \end{bmatrix},
\] (C.3)

\[
\begin{bmatrix} 1 & 1 \\ ik_2 / \varepsilon_2 & -ik_2 / \varepsilon_2 \end{bmatrix} \begin{bmatrix} c_n \\ d_n \end{bmatrix} = \begin{bmatrix} \exp(ik_2d_2) & \exp(-ik_2d_2) \\ ik_2 \exp(ik_2d_2) / \varepsilon_2 & -ik_2 \exp(ik_2d_2) / \varepsilon_2 \end{bmatrix} \begin{bmatrix} a_n \\ b_n \end{bmatrix} .
\] (C.4)

By eliminating the column vector \( \begin{bmatrix} c_n \\ d_n \end{bmatrix} \) we can derive the matrix equation below which
relates the field amplitudes in a recursive fashion.
\[
\begin{bmatrix}
a_{n-1} \\
b_{n-1}
\end{bmatrix} =
\begin{bmatrix}
A & B \\
C & D
\end{bmatrix}
\begin{bmatrix}
a_n \\
b_n
\end{bmatrix}
\]
where the matrix elements \((A, B, C, D)\) depend terms which depend on the dielectric permittivity, the thickness and the wavenumber in the respective layers.

In a one dimensional photonic crystal of period \(d\), the magnetic field (perpendicular to the xz plane) according to the Bloch theorem can be written as

\[
\mathbf{H} = \mathbf{H}_K(z) \exp(-iKz) \exp(i(\omega t - k_z z)) \quad \text{where} \quad H_K(z) = H_K(z + d) \quad \text{is a periodic function of period} \quad d \quad \text{and} \quad K \quad \text{is known as the Bloch wavenumber.}
\]

From the periodic condition above we can derive the eigenvalue problem as shown below

\[
\begin{bmatrix}
A & B \\
C & D
\end{bmatrix}
\begin{bmatrix}
a_n \\
b_n
\end{bmatrix} = \exp(iKd)
\begin{bmatrix}
a_n \\
b_n
\end{bmatrix} \quad , \quad \quad (C.5)
\]

and

\[
\cos(Kd) = \frac{1}{2}(A + D) \quad . \quad \quad (C.6)
\]

Substituting the expressions of \(A\) and \(D\) in Eq (C.6) we get Eq (2.9) of Chapter 2.
D. Transfer Functions for SRS1 and SRS2

The transmission of the two MDSs is plotted in Figure D.1 as a function of $k_x/k_0$ wavelength of 532nm for SRS1 and 640nm for SRS2. The transmission function for SRS2 at 640 nm just beyond $k_x/k_0 = 1$, but it occurs over a small range of values. The transmission function of SRS1 grows higher and broader in the evanescent regime ($k_x/k_0 < 1$), than the transmission function of SRS2.

Figure D1: Transmission of the Fourier components at a wavelength 532 for SRS1, and 640nm for SRS2.