OPTICAL STUDIES OF SUBWAVELENGTH STRUCTURES

By

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To my Mom,
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In this study we have studied optical properties of subwavelength structures. There is still a debate about possible mechanisms responsible for enhanced optical transmission of subwavelength structures. Two different works have been produced to better understand the physics of enhanced optical transmission: hole arrays in silver films and bullseye structures. In hole arrays, the period of the grating and hole size are systematically varied to give peak transmittances at different wavelengths. The spectra coincide when scaled using the array geometry and this shows independence of the dielectric function of the metal. We conclude that the spectra can be explained by interference of diffractive and resonant scattering. The resonant contribution is due to electromagnetic modes trapped inside the structures. In bullseye structures we have shown enhanced transmission without apertures due to trapped modes and cavity resonances.
CHAPTER 1
INTRODUCTION

The ability to concentrate, localize and control the distribution of electromagnetic energy in micro and nanostructures offers fundamental research opportunities and numerous applications. An important field in optical research, called plasmonics or subwavelength optics, has been stimulated by work on optical transmission through subwavelength periodic hole arrays by Ebbesen and coworkers. In that work it was shown that transmission of hole arrays is larger than the area occupied by the holes at some certain wavelengths. In their case, the area occupied by the holes was only 2.2 %, and the transmission at 1.5μm was about 4 %. This was a surprise considering the classical calculation given by Bethe which predicted that transmission would be proportional to \((d/\lambda)^4\) where \(d\) is the hole diameter. For Ebbesen et. al. Bethe's calculation would correspond to practically zero transmission. Initially this extraordinary transmission was attributed to resonant excitation of surface plasmon polaritons at the metal/dielectric interface. Since then, a lot of work has been produced to understand and explain this mechanism and exploit it for possible promising applications. Different explanations other than surface plasmons are suggested. These explanations involve Composite Diffracted Evanescent Waves (CDEWs), dynamical diffraction, cavity modes and/or trapped modes. However, the explanation of enhanced transmission still remains controversial. If we can understand the mechanisms behind all these effects, we can manipulate the distribution of electromagnetic energy in micro and nanostructures for possible applications in near field microscopy, photolithography, display systems and thermal emission sources. Subwavelength optical structures in the near field are not necessarily limited by diffraction and smaller size structures can be fabricated to replace bulky optoelectronic components. These applications can be used in all-optical information processing which is necessary for faster computation and communication.
To further understand extraordinary optical transmission, in this work, we have fabricated and characterized two different sets of structures where optically enhanced transmission can be observed. In the first set of structures we have fabricated hole arrays in silver metal films deposited onto fused silica and zinc selenide substrates and measured optical transmission and reflection. For 44% open area fraction samples, we get extraordinary transmission on the order of 60%. The results are then compared with numerical simulations. By changing the hole size and periodicity of the hole arrays, the optical transmission and reflection measurements together with numerical simulations have shown that enhanced transmission can be explained without surface plasmon polariton excitations and therefore is independent of metal properties. The enhanced transmission is attributed to the role of diffraction in addition to the role of the solutions of the Maxwell equations with periodic boundary conditions, which are called trapped electromagnetic modes. The resonant scattering due to trapped modes and their interference with diffracted modes leads to extraordinary optical transmission. Trapped modes are quasistationary, long lifetime, localized electromagnetic modes inside the structures.

In the second set of the structures we have fabricated and analyzed bullseye structures and we have shown apertureless transmission due to the presence of trapped modes and cavity resonances. In this work we study the effect of metal thickness, dielectric thickness, groove periodicity, number of grooves, groove width relative to the periodicity, polarization and the presence/absence of a central aperture.

This thesis is organized as follows, first the theoretical background (review of literature) is given in Chapter 2 and then in Chapter 3 the experimental procedures are presented, and this is followed with the introduction of computational methods used in Chapter 4. In Chapter 5 and 6,
our experimental and numerical results are given for hole arrays and bullseye structures respectively. And finally conclusions are given in Chapter 7.
CHAPTER 2
THEORY

2.1 Introduction

In this chapter, the theoretical background that will be used to explain the effects observed in the experiments will be discussed. There are different theories that claim to explain the physics of enhanced optical transmission of subwavelength hole arrays. Here, we will review some of those theories with their strengths and the drawbacks. Chronologically, the explanations are, excitations of surface waves (mostly surface plasmon polaritons, but different versions of it have been suggested), dynamical diffraction, Composite Diffracted Evanescent Waves (CDEW) and as many others also claimed diffraction enhanced by resonant character of lattice or surface waves. We will not go to a detailed discussion about the theories, but summarize some of the important claims. The reader is directed to review papers given in the references.

2.2 Electromagnetics of Metals

For a linear, isotropic, nonmagnetic medium Maxwell's equations in CGS units are given as,

\[ \nabla \cdot \vec{D} = 4\pi \rho \]  
\[ (2-1) \]

\[ \nabla \cdot \vec{B} = 0 \]  
\[ (2-2) \]

\[ \nabla \times \vec{E} = -\frac{1}{c} \frac{\partial \vec{B}}{\partial t} \]  
\[ (2-3) \]

\[ \nabla \times \vec{H} = \frac{1}{c} \frac{\partial \vec{D}}{\partial t} + \frac{4\pi}{c} \vec{J} \]  
\[ (2-4) \]

where \( \vec{D} \) is the electric displacement field, given as, \( \vec{D} = \varepsilon \vec{E} \) and the induction field \( \vec{B} \) is given as \( \vec{B} = \mu \vec{H} \). In SI units, \( 4\pi \) is replaced with \( 1/\varepsilon_0 \) and \( c \) is set to unity.
2.3 Surface Plasmons

Surface plasmon polaritons are electromagnetic excitations that occur between at the interface of a metal and a dielectric. Electrons in a metal couple with the electric field of the incident radiation, giving rise to surface waves that decay into the metal. Only p-polarized (transverse magnetic, TM) light, (electric field is in the plane of incidence) can couple to surface plasmons.

Wavevectors of surface plasmons are given as

\[ k_{wp} = k_0 \sqrt{\varepsilon_d \varepsilon_m} / (\varepsilon_d + \varepsilon_m) \]  \hspace{1cm} (2-5)

where \( k_0 = \omega / c \) is the wavenumber of the incoming light. For a metal, the dielectric function can be expressed with the Drude model result as,

\[ \varepsilon_m(\omega) = 1 - \omega_p^2 / \omega^2 \]  \hspace{1cm} (2-6)

Since surface plasmon polaritons are excited only for TM mode (electric field is in the plane of incidence), it is important to have phase matching; gratings (and corrugations make this possible) so that energy can be transferred to the excitations. The electric field distribution (blue curves) on the interface of a dielectric (air) and a metal (gray color) is shown in Fig. 2-1. To be consistent with an alternating surface charge distribution, the electric field has to be in \( x \)-direction here (has to be in the plane of incidence, TM polarization, here magnetic field is into the page). The field distribution falls off more rapidly inside the metal than it does within the dielectric layer above. The dispersion relations for the light line (\( k = \omega / c \)) and surface plasmons are shown in the figure. To generate surface plasmons, the reciprocal lattice wave vectors of the periodic structure must be included in the momentum conservation equations. Incident light can only couple to surface plasmon polaritons if there is a grating.
\( k_{||} + k_{\text{spp}} = G \) \quad (2-7)

where \( k_{||} \) is the wavevector of light parallel to the film surface, \( k_{\text{spp}} \) is the wavevector of surface plasmon and \( G \) is the reciprocal lattice vector given as,

\[
G = \frac{2\pi}{D_g} (\hat{x} + \hat{y}) \quad (2-8)
\]

At low \( k \) values, the dispersion relation of surface plasmon polaritons is linear, because at long wavelengths dielectric function of metal is much larger than the dielectric constant of the dielectric, so dispersion equation approaches the light line. Their momentum does not match to the phase match they need corrugation (momentum conservation). The surface plasmon is essentially an evanescent wave traveling parallel to the interface with intensity rapidly diminishing within the metal.

Enhanced optical transmission (EOT) is attributed to surface plasmon polaritons (SPPs). According to theory, they are excited and resonantly decay through the metal film, or through the hole arrays. There are two different types of surface plasmons: propagating surface plasmon polaritons, and localized surface plasmons. Propagating surface plasmons are usually on patterned films like hole arrays. Localized surface plasmons are associated with voids and/or metal particles and they do not propagate.\(^{11}\)

Resonant absorption of light by SPPs and their decay is claimed to be the reason for extraordinary optical transmission. Even today, this is the most commonly accepted hypothesis of EOT. Near-field considerations and the difference between localized and propagating surface plasmons is given by Smolyaninov et. al.\(^{11}\) A new field, involving applications that depend on surface plasmons is called plasmonics.\(^{8}\)
However the surface plasmon explanation has drawbacks. It only works for TM polarization case, and fails to explain TE polarization. There are experimental results for 1D gratings and 2D hole arrays for the TE polarization case, and extraordinary transmission is shown. Also numerically, for perfect electric conductors extraordinary transmission is shown, where perfect electric conductors cannot support surface waves and surface plasmons. Also for tungsten thin films at visible wavelength extraordinary transmission is shown, but tungsten is a dielectric in visible wavelengths.12

The dispersion relation for surface plasmons is obtained for flat films, but it is not known to modify it for corrugated surfaces. Still surface plasmon explanation is the most commonly accepted theory.

### 2.4 Dynamical Diffraction

Another explanation for extraordinary optical transmission is an explanation by Treacy.3 In that work it is claimed that surface plasmons are a part of the diffracted light but they do not play an important role in enhancement. The enhancement comes from the Bloch waves that peak inside the holes are strongly excited, but the Bloch waves that peak on the metal are absorbed strongly and decay.

### 2.5 Composite Diffracted Evanescent Waves

Composite diffracted evanescent waves (CDEWs) are a description based on scalar diffraction theory. The theory is developed to overcome problems with the surface plasmon explanation.4 It does not rely on surface plasmons but on interference of diffracted evanescent waves, which are claimed to predict the right amount of transmission amplitude, and suppression as well. The authors also claimed that in the first work of Ebessen et. al.1 larger values of transmission ratios of enhancement were reported because Ebbesen et. al. underestimated the aperture sizes.
Incident light is scattered both into radiative modes and evanescent modes. The collection of all evanescent waves is a surface wave, which is named as composite diffracted evanescent wave (CDEW). The CDEW propagates with the wavevector of the incident light, and its amplitude decays with a reciprocal dependence (1/x) on distance as it propagates together with a phase that is shifted by $\pi/2$ compared to the excitation at the source.\(^4\) When the CDEW arrives at another scattering center (for hole arrays it is a neighboring hole) it interferes with the light that is directly hitting that hole, giving rise to electric field enhancement at the aperture entrance when the interference is constructive, or field suppression when the interference is destructive. The grooves, or corrugations are only a means of increasing the amplitude at the entrance of the hole. As the light emerges from the aperture on the backside of the film, some of it again is diffracted into radiative modes and some of it will diffract as evanescent modes, the collection of evanescent waves on the backside also forms another CDEW.\(^4\)

The total intensity transmitted through the hole is expressed as

$$T_C(\lambda) = A_1(\lambda) T_H(\lambda) A_2(\lambda) f_C$$  \hspace{1cm} (2-7)

where $A_1(\lambda)$ is the field enhancement at the entrance of the aperture, $T_H(\lambda)$ the intrinsic transmission coefficient of the aperture, and $A_2(\lambda)$ the effective collection enhancement due to scattering off the corrugation on the exit side of the film, and $f_C$ is the fraction of the total light emerging from the aperture exit.\(^4\)

According to the author's claim, the CDEW explanation can replace the SPP explanation because it predicts the correct peak positions of the transmittance observed in hole arrays and also includes enhancement and suppression effects. And it is more general than surface plasmons in the sense that the enhanced transmission effect seen on dielectrics and perfect conductors can be explained. Another paper where these effects are observed is by Gay et. al.\(^13\) Results of this
work have been controversial and it seems is that the CDEW explanation has some shortcomings in its interpretation. There are two papers analyzing the CDEW interpretation. The first of these\textsuperscript{14} showed that CDEWs are not adequate for handling s-polarization case, and mishandles p-polarization case, even though it is a scalar theory. Also the authors pointed out that, omitting radiative modes in calculations is a serious mistake because the radiative modes can cancel the contribution of evanescent modes. They also claimed that the CDEW description only works after several fitting parameters have been included. They conclude that the CDEW explanation is physically and mathematically flawed.\textsuperscript{14} Another work also concludes that there are flaws in CDEW picture.\textsuperscript{15}

2.6 Trapped Modes

Trapped modes are solutions of the Maxwell equations when they are expressed as in the same form as the Schrödinger equations. When these Schrödinger-like equations are solved, for our structures and similar subwavelength structures, the solutions are in the form of eigenstates. These eigenstates solutions are electromagnetic modes trapped inside the structures.\textsuperscript{5} The trapped modes explanation of extraordinary transmission of subwavelength structures solves all the problems with TE, TM polarization cases; it works with metals (and perfect conductors, numerically shown) and the dielectric structures. The trapped modes are shown to have long lifetimes.

To convert the Maxwell equations into Schrödinger equations we start with the relations\textsuperscript{16},

\[
\begin{align*}
\dot{D} &= c\nabla \times H \\
\dot{B} &= -c\nabla \times E
\end{align*}
\]
for no current or free charge case. In these equations $B$ and $D$ are inductions and $E$ and $H$ are fields. Their relations to each other are given as,

$$ D = \varepsilon E $$

$$ B = \mu H $$

The time-dependent Schrödinger equation is

$$ i \frac{\partial \psi}{\partial t} = H \psi $$

Here $\psi = \begin{pmatrix} E \\ H \end{pmatrix}$ and the Hamiltonian, $H$ is given as,

$$ H = \begin{pmatrix} 0 & ic\varepsilon \nabla \times \\ -ic \nabla \times & \frac{\mu}{\mu} \end{pmatrix} $$

The initial value problem is solved and propagated with the time operator,

$$ \psi(t + \Delta t) = \psi(t) e^{-iH\Delta t} $$

Trapped modes are independent of the choice of the materials, and the only thing important is geometry. Space dependency is included in the dielectric constant $\varepsilon$. The solution of Maxwell's equations in periodic boundary conditions will impose these solutions. This dependence on geometry is shown in the scaling results of Selcuk et. al. 17

### 2.7 Cavity Resonances

Cavity resonances (Fabry Perot) type resonances 18 are shown to exist in 1D and 2D structures for perfectly conducting structures, s-polarization case. 19 When a wave is inside the resonator, it will reflect back and forth and will form a standing wave inside the cavity structure. For our bulleye structures, to be discussed in Chapter 6, with thick dielectric structures beneath
the metal layer, we will show that cavity resonances, which are localized and hence similar to trapped modes\textsuperscript{5}, play a role in optical transmission enhancement.
Figure 2-1. (left) Electric field distribution (blue curves) on the interface of a dielectric (air) and a metal (gray color). (right) Dispersion curves for the light line (dashed) and surface plasmon (solid).
3.1 Fabrication

The following summary gives an overview of fabrication procedures. The procedures for fabrication are determined by the minimum feature size in the structure. If the structures are on the order of 1.5 micron or above, photolithography rather than Electron Beam Lithography (EBL) or Focused Ion Beam (FIB) lithography is preferred for patterning. Photolithography techniques have limitations since it is inherently diffraction limited. EBL and FIB lithography techniques require conductive layers in the sample for charged particles to flow from source to ground. Otherwise excess charged particles at a local point will deflect or repel the beam and will distort the pattern. Also low throughput of EBL and FIB lithography is one of the main reasons it is not a widely employed lithographic process, because each element is scanned serially, whereas optical lithography has a significantly higher throughput. Another consideration in this study is the wavelength ranges over which optical data will be collected. As will be discussed later, the wavelengths where enhanced optical transmission occurs can be controlled with the design of the structures. Our measurements in near infrared (NIR) and far infrared (FIR) require bigger pattern sizes to get enough signal. Since it will take long time to pattern bigger areas with EBL or FIB, photolithography is the preferred method for most of the samples. Solution to this might come from imprint lithography, which could be an important tool to fabricate submicron structures with large areas. But its drawback is the fabrication of the mold, which requires large areas to be written again with EBL; so it is not commonly used yet.

3.1.1 Sample Cleaning

The very first thing is to remove the contamination on the sample. Contamination can be in the form of organic and inorganic particles or the thin films. For polymeric particles deionized
(DI) water sonication is the most efficient method for cleansing whereas acetone:ethanol (1:1) mixture is very efficient for removing inorganic particles.\textsuperscript{22}

Fused silica and zinc selenide (ZnSe) substrates are cleaned sequentially with sonication in deionized water, acetone, isopropanol and methanol at least five minutes for each. Samples are then dried with nitrogen blow. They are inspected visibly and then with optical microscopy for cleanliness. Acetone leaves a residue if it evaporates from the sample; to prevent that methanol must be used right after acetone without letting acetone evaporate. To remove residues of polymers dry cleaning procedures might be needed: these are oxygen reactive ion etch (RIE) (O\textsubscript{2} plasma), barrel asher and UV/ozone cleaning.

3.1.2 Photolithography

Photolithography also known as optical lithography is a common microfabrication technique. It involves a light source emitting at UV wavelengths, which is used to change the chemistry of a radiation sensitive polymer, photoresist, which is coated on top of a sample and exposed through a chrome mask. The change in the chemistry of photoresist with light makes it soluble or insoluble in a chemical called the developer depending on the type of the resist. Positive (negative) photoresist becomes soluble (insoluble) when exposed to UV light.

A Karl Suss MA-6 mask aligner is used in our experiments. Its mercury bulb's light is filtered at 365 and 436 nanometers. Even though different photoresists are used for different purposes, mostly Shipley S1813 thinned with P-Thinner with a ratio of 3:1 (Thinner : Photoresist)\textsuperscript{23} is used in our experiments. S1813 is produced by Shipley (now owned by Rohm-Haas) and distributed by Microchem Corporation. The photoresist is spin coated onto the samples without any adhesive layer. Spin speed is 4000 rpm for 40 seconds after an initial spin of 100 rpm to dispense the photoresist. The photoresist is prebaked at 115 °C for 90 seconds on a hot plate or in an oven for 30 minutes for 90 °C to dehydrate the surface. Baking in
an oven gives a more uniform photoresist film. The thickness of photoresist is about 550nm after baking as determined by atomic force microscopy measurements. If it is not diluted with thinner, S1813 photoresist has a thickness of approximately 1.3 \( \mu \text{m} \) if it is spun 4000 rpm and it has optimal sensitivity near the mercury g-line at 436nm. For samples with feature sizes in the 1.5 \( \mu \text{m} \) range, even thinner photoresists can be used. Usually, for a successful liftoff, the thickness of photoresist should at least be twice that of the metal layer to be evaporated or sputtered. If thinned photoresist (3:1) is used, it is exposed through a chrome mask for 12 seconds (with light power of 8.0 Watt/cm\(^2\)). Exposure time depends on the pattern; it can be up to 30 seconds.

There are different types of contacts made with the sample and the mask in MA6 mask aligner: these are hard, soft and vacuum contacts. In soft contact mode, there is an air gap between the sample and the mask. This exposure type does not give the best resolution but it is used if the mask is fragile, because this gives the least damage to the mask. Also, if the resolution is not critical soft contact is preferred. In hard contact mode, nitrogen gas is used to push the sample against the mask. In vacuum contact mode, the separation between the sample and the mask is under vacuum during the exposure; this is the closest the sample and the mask can get. This contact mode gives the best resolution. Sometimes there is a need to expose the sample without the mask; this is called flood exposure, and it is one of the modes of Karl Suss MA 6 mask aligner used in this study. Exposed photoresist developed for about 14 seconds with AZ 300 MIF (metal ion free) or MF 319 developers without dilution with DI water at room temperature. These two developers have the same chemistry, but are supplied by different companies. Samples are rinsed in DI water and nitrogen blown dried. Karl Suss MA 6 has another mercury light line, which is 365 nm (mercury i-line), which is preferred for other
photoresists. Channel 2 of MA6, with a light of wavelength 436nm (mercury g-line) is used for S1813. Photoresist is used without thinning but must be postbaked if the next step is an aggressive dry etching process. In some of applications Liftoff Resist B (LOR B)\textsuperscript{25} is used. When exposed photoresist on top of LOR is developed, LOR will give an undercut even though it is not UV sensitive. LOR gives an undercut, which helps liftoff process. It is baked at 200°C on hot plate for 2 minutes after it is spin coated usually with 3000 rpm. LOR is soluble in Remover PG by Microchem.\textsuperscript{25}

Metal-ion-free developers such as MIF 319 contain tetramethylammonium hydroxide (TMAH) as the active material. Its chemical formula is (CH₃)₄NOH and it is a strong base. MF 319 and MF 322 can be used for same purposes; difference comes from the amount of TMAH in their unit volume. MF 322 will develop faster but with less control over developing process.

The photoresist can be postbaked at 115°C for 3 minutes if the next step involves dry etching. For different applications different types of photoresists are employed. Photoresist is removed with acetone, but sometimes after dry etching, if it became carbonized, more aggressive methods are necessary to remove it from the surface like oxygen plasma or heating the Remover PG. Also there are special photoresist strippers from Microchem. If photoresist is used without thinning exposure time is about 28 seconds and developing is 30 seconds. To get slower developing, DI water can be mixed with the developer.

If there is no negative pattern on the chrome mask, it may be necessary to use a positive photoresist as a negative resist. The recipe for this is, using AZ 5214 E photoresist\textsuperscript{26}, it is an image reversal photoresist. The successful recipe used in this study is to expose for 10 seconds
with the mask and then bake 2 minutes on a hotplate at 110°C followed by a 30 second exposure without the mask (flood exposure).

3.1.3 Electron Beam Lithography

In Electron Beam Lithography (EBL) electrons are accelerated towards the sample, which is coated with a polymer, called resist that is sensitive to electron radiation. This process is similar to photolithography in the sense that it uses a coated layer, resist, to be patterned, from which the pattern will be transferred to the sample with metallization or etching. But they differ in the sense that, structures are patterned serially, one by one, whereas optical lithography is a parallel process. Polymethylmethacrylate (PMMA) with 950K molecular weight is the electron resist used. It is a positive electron beam resist. It has a long shelf life and has a resolution of 10 nm. It is spin coated on to the substrate with different spin speeds to obtain different thicknesses. After coating it is baked on a hotplate at 180°C for 1 minute. It could also be oven baked at the same temperature with longer time, which would yield a more uniform coating. In our experiments, most of the cases we have used insulator substrates to start with and then coated conducting layers to prevent charging during EBL, oven baking affected our conducting layers, so we used hotplate baking usually. This baking evaporates the solvent for the PMMA. There are two different solutions solid PMMA can be dissolved in: chlorobenzene or anisole. In our experiments we have used anisole as the solvent to dilute PMMA and obtain different concentrations for different thicknesses.

There are two types of electron beam lithography systems: commercial systems and systems modified from a Scanning Electron Microscope (SEM) using proprietary Nabity software. In this study, we have used both of them: Raith 150 as a commercially available EBL system and NPGS installed XL40 FEI SEM. Compared to the Raith 150, the Nabity system using FEI XL40 microscope is much faster, because the large field of view
1mm accommodates large exposure areas. But the uniformity all over the sample is an issue for such large areas. As a solution to this the Raith 150 writes the whole pattern by dividing it into smaller square-area writing fields. By doing so, it can produce same quality all over one writing field because electromagnets can focus the beam even at the edge of the field. But from write field to write field, even though stage position is controlled interferometrically, there are sometimes stitching field errors. The reasons for these errors arise from misalignment of writing fields (due to user) or shifts between consecutive write fields as the patterning proceeds (sometimes due to software/hardware).

The usual acceleration voltage is varied between 10 to 30 keV. Higher voltages give smaller spot size and better resolution but more energetic secondary electrons as well. Lower voltages give less contrast but less number of secondary electrons with lower energies. The number and the density of elements will determine the total time for exposure.

Samples are grounded with colloidal graphite or carbon tape or metal clamps. Focusing, stigmation, aperture alignment and write field alignment must be done reasonably well for a successful exposure. With good focusing, stigmation and aperture alignment, 20 nm diameter spots can be easily exposed. 30 μm aperture size is usually used with 10 keV it gives a current of about 0.18 nA. Bigger apertures will give higher currents (doubling the aperture will quadruple the current) so that exposure time can be significantly reduced. The dose is usually kept between 100 μC/cm² and 300 μC/cm².

Proximity effects occur if two elements are too close to each other. The smallest feature size needed in our experiment is a square array with 300 nm sides and 600 nm spacing in between them. In such a close and dense pattern, proximity effects start to become important.

For developing exposed PMMA, MIBK:IPA (3:1) mixture is used and the time of
development depends strongly on the metal layer underneath the PMMA and the density of elements in the pattern. If thick silver (100 nm or thicker) layer is underneath, 7 seconds is sufficient, whereas on silicon wafer with PMMA, 30 seconds is sufficient for development. Developed PMMA is rinsed with IPA and blown dried with nitrogen. Another mixture that is commonly used is MIBK:IPA (1:1), which develops faster and is very sensitive but gives less contrast. After developing, there may be some PMMA residue on the surface, which can be removed with oxygen plasma in a barrel asher with a power of 100 Watts. If the next step is metallization, the liftoff is done with a mixture of methylene chloride and acetone (9:1). PMMA has a natural undercut, which makes liftoff easier. But sometimes still copolymer might be needed underneath PMMA. Copolymer similar to LOR develops faster and gives an undercut. If it is coated on top of PMMA, after developing and metallization it gives mushroom-shaped T gates.28

To fabricate structures on insulating substrates, for example on fused silica, a very thin but conducting layer of silver (20 nm) is evaporated. This layer is enough to prevent charging in EBL. After exposure, silver metallization and liftoff, this thin layer is etched away with argon ion etch.

3.1.4 Focused Ion Beam Lithography

Focused Ion Beam (FIB) lithography similar to electron beam lithography, involves charged particles, this time ions instead of electrons. FIB processing does not require a mask or a resist as in EBL and photolithography even though masks could be used. It is real time processing of the sample. This removal of material has the disadvantage of being irreversible. In contrast, in EBL and photolithography the resist can be removed after an unsuccessful exposure, and then re-coated to start over. FIB does not offer this flexibility in most of the cases. FIB lithography systems are starting to be commercially available30 even though FIB was first
developed to be used a tool to detect and fixed broken microchips in semiconductor industry. Gallium is the common source of the ions in FIB systems because it is easy to extract ions from the low melting point liquid metal. The melting point of gallium is around 30 °C. Ga\(^{+}\) ions from liquid metal ion source are accelerated towards the sample. This charge must be grounded with a conductive layer on the sample. This conductive layer is usually grounded with colloidal graphite applied on the corner of the sample. Simultaneously there is gallium contamination as material is being removed from the sample. The acceleration energy of ions is 30 keV. The ion current used in the experiments is in the order of 50 pA. This is the lowest current that can be extracted from the system and has the least damage on the sample during imaging and aligning. An FEI DB 235 system is used in the experiments\(^{31}\). This system has dual beams: an electron beam (SEM) for imaging and positioning the sample and an ion beam for patterning. The etching time depends on the thickness of the layer, but it is usually in the orders of a few minutes. For metals focused ion beamed surfaces often become rough, because differently oriented grains have different etching yields.\(^{27}\) Fig. 3-1 shows this ion-beam induced roughness for a 100 nm thick silver film.

### 3.2 Characterization

After each process, quality of samples in terms of defects, surface roughness, and uniformity is examined using atomic force microscopy (AFM), optical microscopy, scanning electron microscopy, surface profilometer and Wycko optical profilometer.

### 3.3 Measurement

Our collaborators in Dr. Tanner's lab performed some of the measurements, mostly mid-infrared measurements. A tungsten bulb is used for near infrared (NIR) and deuterium bulb for UV and visible range of spectrum. Most of the measurements are performed with Fourier
Transform Infrared (FTIR) spectroscopy and optical microscope attached spectrometer. Reflection measurements are done with a conical angle of 8 degrees.
Figure 3-1. 100 nm Silver film during FIB, different grains have different etch rates.

Figure 3-2. AFM images of a dry etched Fused Silica substrate a. Dry etched Fused Silica with CF4 and O2, 150 nm deep, leaves a rough surface with photoresist mask. b. Same sample O2 plasma to smoothen the surface, 500 Watts, 20 minutes, 350 sccm of O2. c. Argon ion etch of the same sample, 10^-4 Torr, 15 minutes, surface gets better but there is lateral etching, depth is around 200 nm. Image is 5micron X 5micron.
CHAPTER 4
COMPUTATION

4.1 Introduction

To fully understand the physics of optical enhanced transmission in subwavelength structures, numerous experiments have to be done systematically. There are many parameters in the experiment to vary, including the metal film and its thickness, aperture size, shape and depth in the structures, periodicity, superstrate material, and the substrate. Along with the possible variations in the fabricated structures there are also variations in the measurement setup as well: polarization, angle of incidence and the measurement of reflection and transmission in higher orders. It is not possible to perform all these experiments because of limited resources. But help comes from the field of computational electromagnetics. In recent years with the advent of computers and with the availability of faster and more robust algorithms, computational electromagnetics has come a long way. There are different numerical methods applied to different electromagnetics problems each with unique advantages and disadvantages. Some of those methods are Rigorous Coupled Wave Analysis (RCWA),32 Finite Difference Time Domain Method (FDTD),33 Frequency Domain Method (FDM), Finite Element Method (FEM), Boundary Element Methods (BEM) and Method of Moments (MoM). This list is not complete. Here I will only review briefly the methods that we used in this study, RCWA and FDTD. These methods are often used in theoretical study of the enhanced optical transmission field.

When feature sizes in the structure become comparable with the wavelength, sometimes called rigorous domain, vectorial rather than scalar solutions of Maxwell equations are required, because effects like multiple scattering become important. RCWA and FDTD are implemented vectorially and thus are capable of solving rigorous domain problems.
4.2 Rigorous Coupled Wave Analysis

RCWA is a commonly used method for the analysis of diffraction gratings. It functions by dividing the structure into multiple layers and expressing dielectric permittivity and fields inside the layers with Fourier expansion in spatial coordinates in each layer. Eigenmodes in every layer are calculated independently from the other layers. These eigenmodes are the solutions of Maxwell's equation in each layer. At the end of the calculation, far field values of the electric field are calculated with the corresponding Fourier transforms giving transmittance and reflectance information. RCWA is claimed to be a fast method and is widely used for optimization of diffraction grating design. But it is a monochromatic method because fields are expressed in terms of the single wavelength of the interest.

Consider the one-dimensional lamellar diffraction grating in Fig. 4-1. To calculate the field intensities, reflected and transmitted, the field inside the grating has to be calculated. There are three regions of interest with index of refractions \( n_1, n_2 \) and \( n_3 \): region I is the superstrate, region II is the periodic rectangular grating (sometimes called binary or lamellar grating) region and region III is the substrate. Rectangular gratings are sometimes called binary or lamellar gratings.

In region I, there are two contributions to the total electric field: the incident electric field and the electric field due to reflected orders. The sum of these contributions, can be written in terms of traveling wave solutions,

\[
\tilde{E}_i = \tilde{E}_{inc} + \sum_j \tilde{R}^j \exp[-i(\tilde{k}_i)_j \cdot \tilde{r}]
\]

(4-1)

where \( \tilde{E}_{inc} \) is the incident electric field, \( \tilde{R}^j \) is the complex diffracted dielectric field of the \( j^{th} \) order, \( (\tilde{k}_i)_j \) is the wavevector of the reflected light and \( \tilde{r} \) is the displacement vector, which is,
\[ \vec{r} = x\hat{i} + z\hat{k} \] (4-2)

Inside region II, the total electric field can be expressed as,

\[ \vec{E}_2 = \sum_j \vec{S}_j(z) \exp(-ik_j \cdot \vec{r}) = \sum_j \vec{S}_j(z) \exp[-i(k_{sj}x + k_{s0}z)] \] (4-3)

where \( k_{sj} = k_{s0}jK \), \( k_{s0} \) is zero order wavevector, \( j \) is the diffraction order and \( K \) is the magnitude of grating vector, given as \( K = \frac{2\pi}{D_g} \).

Similarly the magnetic field can be expanded as a function of unknown vector field functions,

\[ \vec{H}_2 = \sum_j \vec{U}_j(z) \exp(-ik_j \cdot \vec{r}) \] (4-4)

and the dielectric function of Region II is expanded in terms of Fourier series,

\[ \varepsilon(x, z) = \varepsilon(x + D_g, z) = \sum_m \varepsilon_m(z) \exp(ikx) \] (4-5)

where \( \varepsilon_m(z) \) are the Fourier coefficients. The permittivity (dielectric function) is a periodic function of the lattice periodicity \( D_g \), which is inversely proportional to \( K \), magnitude of grating vector, as

\[ K = \frac{2\pi}{D_g} \] (4-6)

With the Maxwell equations,

\[ \nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} = -\mu \dot{\vec{H}} \] (4-7)

\[ \nabla \times \vec{B} = \frac{\partial \vec{E}}{\partial t} = \varepsilon \varepsilon_0 \dot{\vec{E}} \] (4-8)

and assuming \( \exp(-i\omega t) \) time dependence on the fields, the wave equation for Region II is of the form,

\[ \nabla^2 \vec{E}_2 + (k_z)^2 \varepsilon(x, z)\vec{E}_2 = 0 \] (4-9)
If equation 4-3 is substituted into equation 4-9, we get,

\[ \frac{d^2 S_j(z)}{dz^2} - 2i k_z \frac{dS_j(z)}{dz} = (k^2_{m_j} + k^2_{x_0})S_j(z) - k^2 \sum_m \epsilon_m(z) S_{j-m}(z) \]  \hspace{1cm} (4-10)

These are second order-coupled wave equations. Each term \( j \) in the expansion is coupled to the other \( j-p \) terms in the expansion. The solution of this large number of coupled equations can be obtained using a state variables method. Solutions are expressed in terms of eigenvalues and eigenvectors as,

\[ S_j(z) = \sum_m C_m \omega_m \exp(\lambda_m z) \]  \hspace{1cm} (4-11)

And finally similar to region I, fields in region III can be expressed as,

\[ \tilde{E}_j = \sum_j \tilde{T}_j \exp[-i(k_{\tilde{r}})_j \cdot (\tilde{r} - \tilde{d})] \]  \hspace{1cm} (4-12)

where we have assumed that the substrate is lossless.

These 3 equations (eq. 4-1, 4-3, and 4-12) for fields in regions 1, 2 and 3 are phase matched along the interfaces, thus providing solutions for far-field transmission and reflection data.

In this study, RCWA is used with commercially available software G Solver. In G Solver when light is incident on the surface, there are five input parameters of the numerical calculation: these are the wavelength, the two angles for incident and transmitted light and the two angles for polarization angle. In the simulation, the number of Fourier harmonics determines the resolution. If a sufficient number of Fourier components are involved in the solution, the results will converge. Usually increasing the number of Fourier harmonics in the expansion is a good check of convergence if the results are stable. The details of G Solver parameters that are used in this study will be given in Results and Discussion chapter.
4.2.1 Projection-slice theory

The bullseye structure fabricated in this work is analyzed as a one-dimensional grating. Even though the bullseye structure is circularly symmetric, its side view is a 1D grating. If bullseye structure's reflection and transmission is measured with polarized light, it can thus be comparable to a 1D grating. This idea is justified with the Projection-slice theorem, which states that one-dimensional Fourier transform of a parallel projection of an object is equal to a slice passing through origin of the two-dimensional Fourier transform of the same object.\(^{36}\)

In Chang et. al.\(^{37}\) it is shown that 1D gratings can be used as a model to simulate the bullseye structure. Similar conforming results are obtained in this study; and the details will be presented in Chapter 6.

4.3 Finite Difference Time Domain

In the FDTD method, Maxwell's equations are approximated by discretization of continuous field values into discrete values at certain points in space and time. To divide space and time into discrete values a mesh composed of cells is used. FDTD belongs to a more general class of Finite Difference Methods where discretization is used. The field progression and its values are calculated at every grid point. This provides a possibility of recording fields behavior as it propagates or as it interacts with the structures giving a possibility of animation of field as it propagates through the computational domain. Another advantage of FDTD is that structure can be excited with a broadband pulse, so that many wavelengths can be simulated simultaneously.

The FDTD method originated with Yee's work.\(^{38}\) Maxwell's equations are expressed as difference equations. Once again, the Maxwell equations in SI units are, for an isotropic media,

\[
\frac{\partial \vec{B}}{\partial t} + \nabla \times \vec{E} = 0
\]  
(4-13)
\[
\frac{\partial \vec{B}}{\partial t} - \vec{\nabla} \times \vec{H} = \vec{J} \tag{4-14}
\]

\[
\vec{B} = \mu \dot{H} \tag{4-15}
\]

\[
\vec{D} = \varepsilon \dot{E} \tag{4-16}
\]

where \( J, \varepsilon \) and \( \mu \) are functions of time and space. The equations above can be expressed as coupled finite difference equations, as follows,

\[
-\frac{\partial B_x}{\partial t} = \frac{\partial E_z}{\partial y} - \frac{\partial E_y}{\partial z} \tag{4-17}
\]

\[
-\frac{\partial B_y}{\partial t} = \frac{\partial E_x}{\partial z} - \frac{\partial E_z}{\partial x} \tag{4-18}
\]

\[
\frac{\partial B_z}{\partial t} = \frac{\partial E_x}{\partial y} - \frac{\partial E_y}{\partial x} \tag{4-19}
\]

From here each of these equations can be expanded, or approximated if we replace their continuous values with discrete values taken at the midpoint of spacing that has been determined when the computational domain is defined. We can approximate field values as given by,

\[
\frac{1}{\Delta t} \left[ \frac{1}{B_x} \frac{1}{2} \left( i, j + \frac{1}{2}, k + \frac{1}{2} \right) - B_x^{n-1/2} \left( i, j + \frac{1}{2}, k + \frac{1}{2} \right) \right] = \frac{1}{\Delta z} \left[ E_y^n \left( i, j + \frac{1}{2}, k + 1 \right) - E_y^n \left( i, j + \frac{1}{2}, k \right) \right]
\]

\[
- \frac{1}{\Delta y} \left[ E_z^n \left( i, j + 1, k + \frac{1}{2} \right) - E_z^n \left( i, j, k + \frac{1}{2} \right) \right] \tag{4-20}
\]

Equation 4-20 states that, time derivatives can be approximated by differences between adjacent points. The working space is divided into grid cells and values are taken at the center of
the cell. This cell in FDTD is sometimes called the Yee Cell. All Yee Cells make up the lattice throughout the computational domain.

Any numerical solution of a partial differential equation requires that the equations should be transformed into algebraic equations similar to what we have seen in RCWA. And time intervals are expressed as finite time steps. From each step, or from each cell, the field values are evaluated and progressed with Leap Frog schemes.

The computational FDTD algorithm used in this study is the freely available Meep version 0.10. At the moment Steven G. Johnson of Massachusetts Institute of Technology is maintaining Meep. Usually 3D FDTD with dispersive materials needs about 8 processors and around 8 Gb memory to run about a few hours, so supercomputing facilities might be needed for such heavy computations. The FDTD code (Meep) is run in High Performance Computing Center of University of Florida (UF HPC). The author acknowledges the UF HPC for providing computational resources and support.
Figure 4-1. Diffraction grating with reflected and transmitted orders for a planar grating.
Figure 4-2. 3D Yee cell, the electric field values (blue) and magnetic field values (green) are calculated at the center of the cell.
CHAPTER 5
HOLE ARRAYS

5.1 Introduction

Bethe has calculated transmission of for a subwavelength hole with radius \( r \), on a perfectly conducting thin film as

\[
T \sim (r / \lambda)^4
\]

in 1944.\(^2\) But in a recent work it has been shown that the transmission per hole of an array of subwavelength holes in a metal film is higher than what Bethe has predicted and also bigger than the area occupied by the holes at certain wavelengths.\(^1\) Since then much work has been produced to understand this phenomenon. As discussed in the theory chapter (Chapter 2) many authors have suggested different models. The explanation of enhanced optical transmission through subwavelength holes remains controversial.

Here for simplicity, we will denote a specific hole array with parameters \( (a, D_g) \) where \( a \) is hole diameter (if it is a circular hole) or side of a square hole and \( D_g \) is the periodicity of the array. Another definition that we will be using is

\[
f = (a / D_g)^2
\]

where \( f \) is the open area fraction. We will use \( f \) to designate different hole arrays because we have observed that the difference in terms of peak amplitudes comes from the different open area fractions, \( f \), for our samples. Obviously peak amplitudes in transmittance depend strongly on the hole size and the peak positions depend on the lattice constant. A hole array is characterized by geometrical parameters, such as the hole size, the lattice (square, triangular, hexagonal, etc), the hole shape (rectangular\(^{42} \), circular\(^{42,43} \), annular\(^{44,45} \), elliptical\(^{46} \), the hole depth\(^{47} \) and the periodicity\(^{17,47} \). Also there are parameters that are coming from different materials used, like
different metals\textsuperscript{48,49} or the semiconductors like gallium arsenide\textsuperscript{50} and silicon carbide\textsuperscript{51} and the substrate or the superstrate and finally the materials filling inside the holes\textsuperscript{52}.

So far, in all the works produced since 1998 with different explanations, one thing that is certain is that the periodicity of the hole array is the most important parameter of all the listed parameters above in terms of determining the peak position in wavelengths.

Here our results, optical transmission, reflection and absorption of square hole arrays with periods ranging from 1 to 8 μm with different hole sizes, will be presented and comparison with numerical calculations will be given. We have used two different substrates, zinc selenide (ZnSe) and fused silica. Our numerical calculations use Finite Difference Time Domain (FDTD) techniques as well as frequency domain techniques, which include the calculation of the electromagnetic field distribution inside the holes. We will show scaling in the hole arrays, i.e. within the studied arrays, if we plot wavelength axis with respect to dimensionless scaling variable, the spectra of different samples with different holes sizes but with same periodicities will have similar features. Also we will show that enhanced optical transmission is due to diffraction and resonance of trapped modes. Trapped modes are the electromagnetic modes inside our structures. These modes have finite lifetimes and are solutions of Maxwell's equations with periodic boundary conditions.

A dip in R + T spectra will show existence of a trapped mode at the wavelength dip occurs. The resonance of the trapped modes happens close to the diffraction threshold. And depending on the hole size, the contributions due to the trapped modes and diffraction will be different. This is due to the fact that bigger holes interact strongly with the surrounding environment and due to this interaction they have a larger damping so they cannot support trapped modes. As the hole size gets smaller, this behavior changes, trapped electromagnetic
modes inside the holes have longer lifetimes and they do not decay as fast as the ones in the bigger holes.

5.2 Zinc Selenide

5.2.1 Transmission

Commercially available chemical vapor deposition (CVD) grown ZnSe substrates with 25 mm diameter and 2 mm thickness are used as the substrates in the infrared region experiments. As shown in Fig. 5-1, ZnSe is optically transparent between 2.5 μm and 20 μm as much as 60%.

The index of refraction, \( n_d \), of ZnSe is a function of wavelength. Its dependence on wavelength is given as,

\[
n_d^2 = A + \frac{B \lambda^2}{\lambda^2 - C^2}
\]

with the parameters, \( A=4.00 \), \( B=1.90 \) and \( C^2=0.113.53 \). As can be seen in Fig. 5-2, the index of refraction of ZnSe is decreasing for increasing wavelengths. \( n_d \) is 2.49 at 1 μm and 2.34 at 17 μm. In our study we have ignored this change and assumed a constant value of 2.4. We could have made a correction for the change in \( n_d \) of ZnSe, and we checked that for calculation of the peak positions and it did not make much difference. Also in this work mostly we will compare structures with the same periodicity but different filling fraction, which means that the change in \( n_d \) will be same for all. So it is reasonable to ignore the change in \( n \). To prevent the differences that might result from the processing of different samples, all hole arrays used in the transmission measurements are fabricated on the same substrate. For reflection experiments we needed a much larger area of the sample to get enough signal in measurement so each hole array is fabricated on a different ZnSe substrate. The fabrication of hole arrays is discussed in the experiment chapter (Chapter 3). But fabrication steps are patterning with electron beam
lithography or photolithography exposure and development, silver metallization and the liftoff in acetone or methylene chloride. Square hole arrays with squares of side $a$ and periodicity $D_g$ are fabricated. Silver thickness is around 100 nm.

The skin depth of any metal is given as

$$\delta = \frac{1}{\omega} \sqrt{\frac{2}{\varepsilon \mu} \left[ \sqrt{1 + \left( \frac{\sigma}{\varepsilon \omega} \right)^2} - 1 \right]}$$

where $\varepsilon$, which is a function of frequency $\omega$, is the dielectric function of the metal and $\sigma$ is the conductivity. At near infrared (NIR) frequencies, the skin depth of silver is around 20 nm. So our films are optically thick. The dependence of the dielectric function on the frequency is given by the Drude formula as,

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma}$$

where $\omega_p$ is the plasma frequency of silver, its value is 9.1 eV and $\varepsilon_\infty$ is 3.70 and $\Gamma$ is 18 meV. Using this equation we get at 1 µm (1.23 eV), $\varepsilon = -51$ and at 10 µm (123 meV) $\varepsilon = -5470$.

This change in $\varepsilon$ should be observed in the behavior of the hole arrays if their transmittance depends on the metal properties. To test this notion, we have fabricated different samples with different periodicities and different hole sizes such that the periodicities span from 1 µm up to 10 µm. Within this range the dielectric function of silver changes by a factor of about 100. In Fig. 5-3 the transmittance data for 9 different hole arrays are shown. The measurements are performed with a Bruker FTIR spectrometer. All of these 9 samples have different hole radii and/or periodicities, ($a$, $D_g$). In the figure around the highest peaks for each curve the corresponding $a$ and $D_g$ within parentheses are shown. Three different colors are used: blue, green and red for different periodicities of $D_g = 4$ µm, $D_g = 6$ µm and $D_g = 8$ µm respectively. Also for clarity each of the curves is labeled with a letter, just below the letter in the figure, in
parentheses \((a, D_g)\) is shown. Curves labeled with C and F are also shown with an arrow. The diffraction thresholds are indicated by vertical dashed lines. The positions of diffraction thresholds can be calculated as,

\[
\lambda_d = \frac{\lambda}{n_d D_g \sqrt{(i^2 + j^2)}}
\]  \hspace{1cm} (5-6)

where \(n_d\) is the index of the refraction of the substrate, \(D_g\) is the periodicity, \(i\) and \(j\) are indices of the diffraction order. All the blue curves have their highest peak at around 10 μm. The difference between these three blue curves is the hole size. The smallest amplitude blue curve (the one on the bottom) has a hole size of 4/3 μm. The open area fraction for this curve is \(f = 0.11\). As the hole size gets bigger, (as the open area fraction, \(f\) increases) the transmittance amplitude increases. The blue curve with transmittance amplitude of around 25 at around 10 μm has \(f=0.25\) with \(a=2\) μm. Finally the blue curve that is higher than the others is the one with the highest open area fraction \(f=0.44\). Three green curves, which have peaks at longer wavelengths than the blue curve, correspond to hole arrays with 6 μm periodicity. As the hole size gets bigger for these different arrays, the transmittance amplitude increases. The three green curves have open area fractions of \(f=0.11, 0.25\) and \(0.44\). All the red curves, \(D_g=8\) μm have their highest peak at around 20 μm.

The data set shown in Fig. 5-3 can be replotted if we define a dimensionless variable called scaling variable, \(\lambda_s\), as,

\[
\lambda_s = \lambda / n_d D_g
\]  \hspace{1cm} (5-7)

The result of plotting with respect to this scaling variable is shown in Fig. 5-4 with the corresponding diffraction thresholds shown by the dashed lines. The positions of diffraction thresholds for this curve is,
\[ \lambda_d = \frac{\lambda}{(i^2 + j^2)^{1/2}} \]  

(5-8)

where \(i\) and \(j\) are indices for the order of diffraction. In Fig. 5-4, in parentheses the indices are shown, the positions of thresholds are at 1, 0.7 and at 0.45. Every time diffraction threshold opens up, transmittance goes down, because light is scattered into higher diffraction orders and out of the normal direction. Here in this figure \(f=0.25\) samples are shown, with three different colors, the blue curve (C) is for (2,4), green (B) is for (3,6) and the red curve (A) is for (4,8). The importance of this figure is that the transmittance of different samples with same periodicity and with different hole sizes but with the same open area fraction, \(f\), when plotted against the scaling variable will have similar features in their spectra, i.e. peaks and dips, at similar wavelengths. This result is called scaling. Scaling means that enhanced optical transmission of hole arrays is independent of metal properties, and it implies that the enhanced optical transmission effect is mostly a geometrical effect.

5.2.2 Reflectance

In Fig. 5-5 the reflectance spectra for three arrays with different open area fractions are shown. The three arrays \((a, D_g)\) are \((8/3,8), (4,8)\) and \((16/3,8)\) with blue (A), green (B) and red (C) colors respectively. Also in the figure with the dashed lines, diffraction thresholds are shown for \((1,0)\) and \((1,1)\) orders. At the diffraction threshold the transmittance of the hole array in specular direction goes down, because light is scattered into higher orders. This also results in a drop in reflectance spectra for the corresponding hole array. In the figure this is best observed for the sample with \(f=0.25\) (the green curve). For these curves, the behaviors can be explained with two different phenomena, diffraction and the trapped modes. As will be discussed below, for different open area fractions, their contribution is different. As the open area fraction gets bigger, the diffraction dominates, the structure is not able to hold the trapped modes long enough, and
the trapped modes thus have a short lifetime and decay faster. As for the smaller open area fraction, for example for \( f=0.25 \), the contributions from both trapped modes and diffraction is observable. As the open area fraction gets even smaller, here for array shown with \((8/3,8)\) \( f=0.11 \) (blue, A curve), the diffraction does not play an important role as much as the trapped modes.

There is an angle of about 10 ± 2 degrees of incidence in the measurement setup; this makes the analysis of the reflection data more complicated. But even with this complication, two samples are shown in Fig. 5-6. In this figure, reflectance and transmittance data for two samples with \((3,6)\) (blue curves) and \((4,6)\) (red curves) are shown. In the figure with A and B letters reflectance and with C and D letters transmittance is shown. Instead of having a reflectance minimum where transmittance is a maximum, they are shifted with respect to each other. Right at \( \lambda_s=1.0 \) reflectance for the two curves goes down whereas transmittance decreases at longer wavelengths. This difference can be attributed to the above-mentioned angle of incidence in the reflectance measurement. At the moment, a new stage with capability of rotation is being built to measure transmittance at an angle of 10 degrees.

5.2.3 Absorption

With the reflectance together with transmittance measurement we can calculate the absorption in the structures. All of the absorbed light goes to ohmic losses. Absorption can be calculated from,

\[
A = 1 - T - R
\]

where \( A \) is absorption, \( T \) is transmittance and \( R \) is the reflectance amplitude. In Fig. 5-7, the result of such a calculation is shown for a sample with \((3,6)\) with \( f=0.25 \). The red curve (C) is for transmittance, the blue curve (B) is for reflectance and the black curve (A) is for absorbance. The dashed lines show the diffraction threshold and their orders are given in parentheses.
In Fig. 5-8 the numerical (blue curves or B, D & F) and the experimental (red curves or A, C & E) results are shown for different open area fractions, \( f \). There are two panels, in the upper panel \( f=0.25 \) and \( f=0.44 \) are shown, in the bottom panel \( f=0.11 \) is shown. The numerical calculations are performed by time dependent Lanczos-split algorithm.\(^{16}\) For silver the values to be used in numerical simulation are obtained from Ordal et. al.\(^{56}\) There is a very good match with the numerical and experimental results. In the numerical results the peaks are sharper and their amplitudes are little bit higher than the corresponding experimental results. The experimental results are broader due to imperfections, such as film roughness and hole size irregularities, in the samples. The diffraction thresholds are shown as vertical dashed lines.

In Fig. 5-9, electromagnetic field distribution of \( B_y \) is shown, magenta is for positive \( B_y \), and red is for negative \( B_y \) on the top, for \( f=0.11 \) and on the bottom it is for \( f=0.44 \). These figures are plotted as a function of \( z/D_g \) in x-axis and \( x/D_g \) in x-axis. The direction of the field is \( B_y \). The fields are localized in the vicinity of the holes. In Fig. 5-10, the electromagnetic energy (intensity \( E^2 + B^2 \)) is shown for different hole sizes (different open area fractions of 0.11, 0.25 and 0.44) for \( D_g=4 \) \( \mu \)m. In the figure, the leftmost panel represents the smallest holes with \( a=4/3 \) \( \mu \)m, the middle panel \( a=2 \) \( \mu \)m, and the rightmost panel \( a=8/3 \) \( \mu \)m. The color orange is for the highest field intensity. As can be seen from the figure field intensities are localized differently for different hole sizes. For the smallest hole, the fields are stronger on the metal circumference of the hole. For the hole size with \( f=0.25 \) (the middle panel in the figure) the modes are weaker, and some of the energy is localized in the center. Incoming light here is polarized in \( y \)-direction that results an asymmetry in the figures. In Fig. 5-10c, the field intensities are shown for the biggest hole. Again fields are localized around the circumference and in the center.
5.3 Fused Silica

Fused silica is used as a substrate for experiments in near infrared regions. It is index of refraction as a function of wavelength (in microns) given by

\[ n^2 - 1 = \frac{0.696 \lambda^2}{\lambda^2 - (0.0684)^2} + \frac{0.408 \lambda^2}{\lambda^2 - (0.116)^2} + \frac{0.897 \lambda^2}{\lambda^2 - (9.90)^2} \]  

(5-10)

In Fig. 5-11, transmittance of fused silica as a function of wavelength is shown. The transmittance is 80% up to wavelengths of 3.5 μm and at longer wavelengths transmittance decreases. Beyond 5 μm fused silica is opaque. The water and the metal content determine its optical transmission, which is usually comes from its production. But our purposes and over the wavelength region in which we are interested, the optical transmittance spectra are good enough.

In Fig. 5-12, the optical transmittances of two array structures fabricated on fused silica substrates are shown. The red curve (A) is for 0.5 μm side square holes in 70 nm thick silver film with 1 μm periodicity. The blue curve (B) is for (1,2). Diffraction thresholds are shown with dashed lines. This figure shows that scaling holds in the near IR region with good overlap of the scaled peak and dip positions. In Fig. 5-13, the data for Fig. 5-12 is scaled with the scaling parameter λs.

5.4 Annular Arrays

In Fig. 5-14 SEM pictures of two hole arrays are given. In Fig. 5-14a square arrays, in Fig. 5-14b square arrays with square holes filled with a metal annular structure are shown. The periodicity of the structures shown here is 4 μm, for the annular structure on the right, inside there is a silver square symmetrically spaced at the center of each hole.

It was first shown numerically by Labeke et. al. that the annular structures will lead to much higher transmission than a regular hole array.\textsuperscript{58,59} The stated reason for this enhanced transmission is that an annular structure will support the TEM mode of a waveguide, whereas a
regular hole will not. Accordingly, an enhancement is expected for annular arrays. With the following experimental works, this expected result is not observed. There are two reported experimental studies of annular arrays. The first study, taken in the visible region$^{60}$, there was not much a difference between the annular array and regular hole array. In the second work, a negligible difference in transmission amplitudes is observed, but the peak positions for the annular arrays were red shifted with the shift being attributed to cylindrical surface plasmons.$^{61}$ For us, when we have fabricated square annular structures, we also have not observed any further enhancement than that which occurs in a regular hole array. Our results are shown in Fig. 5-15 for regular hole arrays (the black curve, also labeled as A) and hole arrays with metal square (with inside square having a side of 1.0 μm) annular structures in the center (the red curve, B), both arrays with $D_g=4 \mu m$. Both structures have about 30 % transmission. So in our study, we also have not observed any enhancement or peak shifts due to annular arrays.
Figure 5-1. Optical transmittance of ZnSe substrate up to 25 μm.
Figure 5-2. Index of refraction of CVD grown ZnSe as a function of wavelength.
Figure 5-3. Transmittance of hole arrays with different $a$ and $D_g$ on ZnSe substrate. In parentheses ($a, D_g$) are shown. The blue curves are for $D_g=4\,\mu m$ (also labeled with A, B and C), the green curves for $D_g=6\,\mu m$ (D, E and F) and the red curves are for $D_g=8\,\mu m$ (G, H and I).
Figure 5-4. Transmittance of hole arrays with $f=0.25$ vs. scaling variable $\lambda_s$. Dashed lines show diffraction thresholds. The red curve (A) is (4,8), green (B) is for (3,6) and blue (C) is for (2,4).
Figure 5-5. Reflectance vs. scaling variable for 3 different hole arrays with (8/3,8) blue A with $f=0.11$, (4,8) green (B) with $f=0.25$ and (16/3,8) red (C) for $f=0.44$. These 3 arrays have different open area fractions but the same periodicity.
Figure 5-6. Transmittance and reflectance data for two hole arrays with (3,6) for blue curve, A is reflectance and D is for transmittance, and (4,6) the red curve, B is for reflectance and C is for transmittance. Dashed lines show diffraction thresholds.
Figure 5-7. Red curve (C) is for transmittance, the blue (B) reflectance, the black curve (A) is for reflectance and transmittance added together for an array of (3,6) (f=0.25).
Figure 5-8. Computation vs. experiment for hole arrays with different open area fractions, \( f \), are shown in the figure. The red curves (A, C & E) are for the experimental; the blue curves (B, D & F) are for computational results.
Figure 5-9. Electromagnetic field distribution for $E_y$ for $f=0.11$ (top figure) and for $f=0.44$ (bottom figure).
Figure 5-10. Energy density inside the holes with different open fractions, $f=0.11$ (left), $f=0.25$ (middle), and $f=0.44$ (right).
Figure 5-11. Transmittance of fused silica as a function of wavelength between 2.5 μm up to 5.0 μm.
Figure 5-12. The blue curve is for $a=0.5 \times 0.5 \, \mu m$ with $D_g=1 \, \mu m$ and the red curve is for $a=1 \times 1 \, \mu m$ with $D_g=2 \, \mu m$. 
Figure 5-13. Transmittance of two hole arrays with (0.5,1) blue (B) and (1,2) red (A) on fused silica substrate as a function of scaling variable, $\lambda_s$. Diffraction orders are shown with the dashed lines with the orders shown in the parenthesis.
Figure 5-14. SEM micrographs of two hole arrays with same periodicity ($D_g = 4 \, \mu m$) and same hole size ($a = 2 \, \mu m$). In the annular structure on the right, inside of the holes, there is a silver metal square thereby giving an annular shape to the aperture.
Figure 5-15. The red curve (B) is for the annular array with 2 μm hole size, inside 1 μm metal square and with $D_g=4\mu m$, the black curve (A) is for (2,4) hole array.
CHAPTER 6
BULLSEYE

6.1 Introduction

The bullseye structure is composed of concentric circular rings as shown in Fig. 6-1a. The rings, comprised of metal (Ag) and dielectric (PMMA) components, are periodically spaced. As can be seen in Fig. 6-2b, where the side view of a bullseye structure is shown, the structure is composed of mesas and steps with finite heights. Structures with this geometry can be considered as circular diffraction gratings. Their optical properties have been studied in the literature; enhanced optical transmission (EOT) is shown in the near infrared (NIR) and microwave regions. At terahertz frequencies the transmitted light amplitude depends on the phase between the light emitted from the central aperture and the light associated with the resonances (trapped modes) set up within the grooves. The net effect can be either enhancement (constructive interference) or suppression (destructive interference). In addition to enhancement, beaming is shown if the exit aperture is patterned concentric with the grooves. This opens a possibility of using a bullseye structure as a near field lens. Also it is shown that the surface waves are propagating towards to center. In another work, surface plasmons are excited with a near scanning optical microscope (NSOM) on a bullseye structure and interference effects with these surface waves are shown. According to Steele et. al. as the surface plasmon polaritons propagate across each groove they gain 0.07π phase shift. All these works mentioned above have used optically thick metal grooves. Recently a numerical study claimed that dielectric periodic structures on top of a thin flat silver film would lead to EOT as well.

In this work we have fabricated bullseye structures of silver metal films on quartz substrates with thick PMMA dielectric grooves. On top of the grooves we have evaporated a thin
metal silver layer. The silver metal is thick enough to reflect most of the light in the NIR, which is the region of interest for us in this study. Our initial intent was to show resonant light tunneling through optically thick metal films\textsuperscript{69-70-71} and associate this tunneling with the excitation of surface waves. In addition to that, we also wanted to understand the interaction between the central aperture and the surface waves in a circularly symmetric structure. Basically, in a bullseye structure, the incident light would have a direct path through the aperture and an indirect one through the generation and transmission of surface waves.

In our study, with the grating structure we are able to couple to trapped modes.\textsuperscript{5} Due to the presence of the trapped modes, our structures have EOT even without apertures. The peaks in transmittance spectra of our structures are very close to the diffraction thresholds and their positions are determined primarily by the periodicity of the bullseye structure, but as the thickness of the dielectric is increased secondary peaks due to cavity modes are observed. It is shown by other authors that thick dielectric grooves surrounded by metal will support cavity modes.\textsuperscript{72} When the grooves are thick enough, we clearly observe these cavity modes as well at longer wavelengths. We emphasize in this chapter the distinction between the trapped modes and their associated diffraction thresholds, which are sensitive to lattice periodicities, and the cavity modes, which are associated with the dielectric-filled grooves and are sensitive to changes in geometrical parameters like the dielectric thickness and the ratio of dielectric width to periodicity. We also observe that when the central apertures are drilled, the transmittance amplitude of the peaks that are due to trapped modes increases but the peak amplitude due to cavity resonance does not change. In addition, if some of the rings are removed from the structure, transmittance amplitude is decreased. These experiments, which are described below, thus help distinguish the relative contributions of the trapped and cavity modes.
To numerically analyze our structures, we have used rigorous coupled wave analysis (RCWA) implementing software GSolver demo. This software can numerically simulate the behavior of 1D subwavelength diffraction gratings. The justification for the use of 1D gratings to simulate bullseye structures is discussed in Chapter 4, in accordance with the Projection-slice theorem.

6.2 Results

When the PMMA is thick enough (around 200 nm) EOT is observed. If the PMMA is even thicker (400nm), a 2nd peak is observed. We attributed the first peak, which is in close proximity to the diffraction threshold, to trapped modes and diffraction as discussed in the previous chapter for hole arrays, and the second peak to cavity resonances that take place within the metal cavity surrounding the thick PMMA dielectric. To understand the interactions between different electromagnetic modes, the band gap of the structure has to be measured or calculated; this can be done if we can measure or calculate reflection as a function of angle of incidence.

Fabrication steps are shown in Fig. 6-1; the details of the processes are discussed in Chapter 4. Briefly, a very thin, 3 nm thick, InOx layer is sputtered onto quartz substrates; this film has an optical transmission of about 75 % starting from 800 nm towards longer wavelengths as shown in Fig. 6-3. The electron resist (PMMA) is applied by spin coating and electron beam lithography (EBL) is used to pattern the bullseye structure. A silver film is then thermally evaporated onto the developed bullseye structures. Fig. 6-2a shows an SEM of a typical structure. After the optical transmission and reflection of this structure is measured, the central aperture is drilled through using a focused ion beam (FIB). The optical transmission and reflection can then be again measured and changes associated with the aperture noted.

Flat (surface roughness is around 5-10Å by AFM) silver films of 35 nm thick are optically opaque at 800 nm and longer wavelengths as shown in Fig. 6-4. In Fig. 6-4 the bulk
surface plasmon mode, at 323 nm (3.84 eV) is giving the transmittance maxima shown. In the literature the bulk plasma of silver is stated to be 3.80 eV \footnote{55} and 3.78 eV \footnote{75}. From the plasma frequency, the number of free electrons can be calculated as,

\[
n = \frac{\omega_p^2 m}{4\pi e^2}
\]

where \( m \) is electron mass and \( n \) is volume number density of free carriers. From this formula, which is in CGS units, \( n \) is calculated to be 1.06x10\(^{22} \) cm\(^{-3} \), which is smaller than the reference value of 5.85x10\(^{22} \) cm\(^{-3} \). \footnote{76}

### 6.2.1 Peak Positions

Opening of a diffraction threshold leads to a drop in transmittance of the structure, giving a transmission minimum at the threshold. The diffraction threshold is defined in the previous chapter in the discussion about hole arrays. The transmission maximum is always on the longer wavelength side of the threshold. Opening a diffraction threshold, basically, enables the structure to couple to higher diffraction orders or surface modes leading to a sharp drop in transmission in the direction normal to the film. With this the peak positions lie right next to the diffraction threshold, just on the longer wavelength side. Scaled diffraction threshold positions are given as,

\[
\lambda_{\text{threshold}} = \frac{\lambda}{n_d D_g \left(i^2 + j^2\right)^{1/2}}
\]

where the integers \( i \) and \( j \) denote the diffraction order, \( n_d \) is the refractive index of the substrate and \( D_g \) is the periodicity of the structure.

Equation 6-2 basically determines the peak positions. We have fabricated bullseye structures with different periodicities; \( D_g = 2 \mu m \) is shown in the Figure 6-5 and \( D_g = 1 \), \( D_g = 1.2 \mu m \) are shown in Figure 6-6. The dimensionless thresholds are shown with a dashed line in Fig. 6-5 and 6-6 and in parentheses the orders \( (i,j) \) are marked. The longest wavelength threshold
corresponds to (1,0). In these figures, the transmittance data are plotted against a dimensionless scaling variable, $\lambda_s$, which is defined as,

$$\lambda_s = \lambda / D_g n_d \quad (6-3)$$

The peak positions, which depend on periodicity of the structure, are then calculated by the equation,

$$\lambda_{peak} = \lambda_s D_g n_d \quad (6-4)$$

where $\lambda_{peak}$ is the wavelength of the peak.

In Fig. 6-5, there are two peaks; corresponding $\lambda_{peak}$'s are approximately 2400 nm and 3275 nm with $\lambda_s = 0.845$ for the first peak ($i=1,j=0$) and $\lambda_s = 1.137$ for the second peak ($i=1,j=1$). We can compare peak positions ratio (ratio of first peak position to the second peak) with the ratio of positions of corresponding diffraction thresholds. It is difficult to identify the exact location of the peaks, because the peaks are very broad. We estimate a ratio of $1.14/0.85 = 1.35$. Using the corresponding diffraction thresholds at $\lambda_s = 1$, 0.7 and 0.45 we calculate the ratio of the position of the first diffraction threshold to second threshold to be 1.4, in good agreement with the observations.

In Fig. 6-6, the red (A) curve corresponds to a bullseye with $D_g = 1 \mu m$, the peak positions are approximately at $\lambda_s = 0.9$ and $\lambda_s = 1.25$. Their ratio gives 1.39. From the same figure, for $D_g = 1.2 \mu m$ (the blue curve (B)), the peak positions are 0.80 and 1.15, and their ratio gives 1.44. These two spectra when plotted as a function of the scaling variable, $\lambda_s$, will have similar features in their spectra; this is called scaling. Basically this means, the physics of these two structures is only dependent on geometry but not on the other parameters like metal properties.

The peaks in our experimental data are very broad compared to calculations. This is in all likelihood due to irregularities of the sample like grating height not being constant all over the
total sample area and the presence of imperfections in the metal films. Also simulation assumes normal incident plane waves (zero incident angle) at a single wavelength. In the measurement the light beam has an angular cone of incidence, about 10 degrees, which may couple to higher order surface modes. In Fig. 6-6 the peak positions are marked with arrows. There are two distinct peaks in each curve.

Beyond 3.3μm the peaks are beyond our spectrometer's range. Also at 5μm there is a cutoff due to the fused silica substrate. In all our measurements the structures are illuminated from the quartz side.

6.2.2 Dielectric Thickness

Dielectric thickness is an important parameter in our experiments. As can be seen in Fig. 6-7, which compares experiment with simulation, for a sufficiently thick enough dielectric (300 nm) shown with light green color (also labeled with letter A), trapped modes are excited and enhanced transmission is observed. In the figure dark green color (label C) is for simulation for a 300 nm thick PMMA dielectric. When dielectric thickness is a factor of three thinner (100 nm), there is no peak observed as shown in the figure where the cyan curve (label B) is experimental and the blue curve (label D) is for numerical result. Here the results are shown for a bullseye with $D_g=1.0$ μm fabricated with a 35 nm thick silver film. Dielectric thickness is measured with AFM (atomic force microscope) and surface profilometer. The AFM gives a result around 350 nm. The PMMA datasheet for the spin speed used for this sample gives the thickness to be 300 nm. AFM scanning is done with a very slow scan speed giving a high resolution with good reproducibility.

The metal layer on top of the grooves provides coupling as well as it might be supporting surface waves. If the metal layer is made too thick (100 nm or so) the peaks disappear. We have also observed this in simulations.
In Fig. 6-8 numerical results for different PMMA thickness are shown. For the 300 nm thick PMMA sample there is only one peak formed around 1 μm, which is very close to the diffraction threshold, shown as a dashed line in the figure. As the dielectric thickness is increased, there is a second peak formed as seen in the figure with magenta color (the curve denoted with letter C). And as the thickness of the dielectric increases further there is a red shift in second peak's position but first peak's position does not change much with the thickness. The first peak's position is determined by the diffraction threshold and is independent of the dielectric thickness. The strong dependence of position of the second peak with respect to dielectric thickness suggests that this peak is due to a cavity mode inside the dielectric. As the cavity increases in size the mode it supports moves to longer wavelengths as would be expected.

6.2.3 Central Aperture

In Fig. 6-9 the transmission of $D_g = 1.2$ μm bullseye after drilling the central hole with different diameters, is shown. The central hole will contribute to transmission with zero phase shift. If the two peaks result from different processes, and any of those processes would add a phase to the transmitted light, they will interfere with the light coming out of the central hole. In our experiments, with the central hole, transmission always goes up for both of the peaks, thus implying a constructive interference. As shown in Fig. 6-1, the central aperture is circular. Opening the central hole, increases the overall transmission more than the relative area increase associated with the hole opening. Accordingly, the light coming out of the bullseye structure and the light coming out of the central hole interfere constructively.

To make sure that results are repeatable and to minimize concerns about the effect of the focused ion beam on sample quality, at least four of the same bullseyes with same hole size are fabricated. Bare holes have a very low transmittance as shown in Fig. 6-10. Cao et al.\textsuperscript{64} showed the position of the central hole with respect to grooves can lead to enhancement as well as
suppression of the transmitted light. In our experiments we have not observed this effect even though we have changed the relative position of the hole with respect to the grooves.

In Fig. 6-9, optical transmission of a single bullseye structure with different diameter center holes is shown. The same hole is widened with the FIB after each measurement. Here, as can be seen from the figure, as the hole size increases, the transmission goes up. The dielectric thickness of this sample is around 350 nm. In this figure the green curve is for the case without any holes, the cyan curve (also labeled as E) is for 2 μm, the black curve (label F) is for 5 μm, blue (D) is for 7 μm, magenta (C) is for 10 μm, yellow (B) is for 12 μm hole and red curve (A) is for 16 μm diameter hole. Bullseyes with 2, 5 and 7 μm central holes have the same transmittance amplitude. The first peak, peak around 1200 nm shows the largest relative increase compared to second peak (peak around 2000 nm) for all hole sizes. As can be seen in Fig. 6-9 the transmittance of the first peak increases with the widening of the central hole whereas the second peak amplitude does not change. These observations suggest that the second peak is a cavity mode, which is only dependent on the dielectric thickness and thus does not shift in position since the dielectric thickness remains constant.

Fig. 6-10 shows the transmission of corresponding bare holes on the same 35 nm-thick flat silver film. The coloring is the same as used in Fig. 6-9. The same behavior can be observed here as well; at longer wavelengths, the 16 μm diameter hole has the same transmission as much as 2 μm diameter hole. All of the curves in Figure 6-10 collapse to the same curve for wavelengths greater than 1800 nm.

6.2.4 Sidewalls

We presented our results for specular transmission measurements. Light coming out of the sidewalls will be contributing more to higher orders. It's hard to say how thick the sidewalls are. Grooves being very tall, silver film being very thin, it might be safe to assume there are no
sidewalls, or they are very thin and very leaky. In simulation electric field is perpendicular to slits. More work and analysis might be needed to assess the role of sidewalls on overall optical behavior of our structures.

6.2.5 Silver Thickness

During fabrication of the bullseye structure silver is evaporated to cover everywhere on the sample uniformly. This coating goes on top of the grooves and onto the mesa between them and has the same thickness for both. The amount of this silver is varied and optical transmission measurements are performed for different thicknesses. Experimental results are shown in Fig.6-11. As can be seen for the blue curve with silver thickness of 17 nm, a peak does not form. This film is too thin and the structure transmits without a well-defined diffraction threshold. When the silver thickness is doubled (the red curve for 35 nm thick silver) the diffraction threshold peaks are observable. This is the standard thickness used in all other bullseye experiments where thickness of silver was not a parameter, i.e. when for example thickness of the dielectric is varied for different samples, a 35 nm-thick silver film is used for all the samples. In Fig. 6-11, yellow and navy curves correspond to thicker silver films, 50 nm and 65 nm respectively. In other words the diffraction threshold does not change. After the peaks are formed, we did not observe much of a change in the peak positions within the thickness values varied here. In the computation without the metal on top, light would not be coupled to the dielectric layer and if the metal layer is made too thick, enhanced transmission will not be observed. In Fig. 6-12 the numerical results for the silver thicknesses used in the experiment are shown. In the simulation program thicknesses can be made multiples of 20 nm. The results shown in Fig. 6-12 are for 20, 40 and 60 nm thick silver film with the colors of blue, red and navy respectively.
The thickness of the silver has to be right to couple the light into the dielectric PMMA, but if it is too thick, then light will not penetrate through this metal layer and cavity modes will not be observed. The ideal thickness of the silver film is found to be between 30 nm to 70 nm.

6.2.6 The Role of Missing Rings

If some of the rings are missing in a bullseye structure, its overall transmission will go down as shown in Fig. 6-13. In Fig. 6-13, green curve (A) is for a bullseye with all the rings. Starting from the 10th ring from the center, we have removed some of the rings. The red curve (C) is for one ring (eleventh) missing, blue (D) is for 3 rings (11-13th rings), cyan (E) is for 5 rings (11-15th rings) and yellow (B) is for 10 (11-20th) rings missing. Magenta (F) represents the case when the first ten rings are missing. As can be seen from the figure, as we take out some of the rings the overall transmission goes down. The overall area occupied by the rings is $100 \times 100 \, \mu m$ and it contains 50 rings. From the data it seems that the biggest contribution to transmittance comes from the first 20 rings. Also the effect is most drastic if the rings are taken from the center. This experimental result justifies in our simulation of FDTD where we can use a finite number of rings to simulate this structure. The details of this will be discussed in Results and Discussion chapter. When a ring is removed, it is replaced with a flat silver film. Transmission drop shows that light only passes through a part where there are grooves. In a recently published paper it is shown that a hole array composed of $9 \times 9$ holes will be enough to saturate the transmittance enhancement.4

6.2.7 Different Open Area Fractions

Bullseye with different ratios of groove width to periodicity other than 0.5 can be fabricated to understand more about the nature of resonances inside the dielectric. The calculated behavior for different fractions of $a/D_g$ is shown in the Fig. 6-14 where $a$ is the width of the dielectric PMMA. Different ratios of $a$ to $D_g$ (for $D_g=1 \, \mu m$), $a=0.3 \, \mu m$ red (A), $a=0.4 \, \mu m$ blue
Dielectric thickness is 300 nm. The highest transmission is observed for $a=0.3 \, \mu m$. For the computed groove widths here, only 0.5 \mu m ones are fabricated and measured. To further progress this work, some of the different groove widths should be fabricated to confirm the simulation results. Also in this figure the 40 nm-thick flat silver film's transmittance at these wavelengths is shown with the green curve.

### 6.2.8 Polarization

The optical measurements shown up to this point are performed with unpolarized light. For the bullseye structures, since they are circularly symmetric, the polarization will not make any difference. In Fig. 6-15 two curves are shown, the blue curve corresponds to case with NIR polarizer whereas the red curve corresponds to unpolarized light. As can be seen from the figure, the blue curve is very noisy because with the polarizer, the light intensity gets lower. The peaks are at the same position in both of the curves for polarized and unpolarized light. Since polarized and unpolarized light in the experiment (the blue and the red) have the same spectral shape we conclude that we can use unpolarized light in our measurements and can compare results with numerical calculations performed with polarization. All the numerical results are shown up to now only for TE modes. TE (also labeled as s) polarization is defined as the case where electric field is parallel to slits, perpendicular to plane of incidence. Within the polarization choices of TE and TM, TE polarization fits better to our experimental results. This is shown in Fig. 6-16 where there is a comparison of two different polarizations and the experimental result. The black and the yellows curves are for the experiment with unpolarized light for two different samples with same dielectric thickness, the blue (E) curve is for TE polarization, the red (A) curve is for TM polarization and finally the green (B) curve is the average of the cases TM and TE polarizations. (the average of the blue and the red curve). The average is shown here because for
a similar structure to ours in their work, Chang et. al. showed a comparison of average of two polarizations for unpolarized light incident on a circularly symmetrical structure.\textsuperscript{37}

With TM polarization computation there seems a numerical instability because the transmission calculation results are growing up to 0.8 after 1600 nm and remain as high. Our conclusion out of this is that, in the bullseye structures TE polarization will couple and dominate the transmission characteristics.

\textbf{6.2.9 Separation of bullseye into two components}

Can bullseye structure be considered to be composed of two components, i.e. as the sum of two different diffraction gratings?

In this section we will decompose bullseye structure into two diffraction gratings and investigate the behavior of these separate structures, and compare them with the bullseye structure. Separation into these two components is shown in Fig. 6-17. In the figure a sideview of bullseye structure is shown which can be divided into two components. Corresponding transmittance data for these two diffraction gratings is given in Fig. 6-18 and Fig. 6-19 with their structure shown just above the figures. In Fig. 6-18 optical transmittance for the structure shown above the graph is shown for self-standing (the black curve) and structures with substrates (the green curve is for 400 nm thick PMMA and the blue curve is for 600 nm PMMA).

In Fig. 6-19, the transmittance of the second diffraction grating with 35 nm thick silver is shown. The black curve is for self-standing, and the blue curve is for structures with the substrate (fused silica, n=1.44) is shown. In the blue curve there is an additional peak at 1440 nm as a difference from the case of self-standing metal films. For each interface there is a diffraction threshold. But in the previous structure of Fig. 6-18, there is no peak formation at 1440 nm, but there is a small kink in the spectra. The sum structure transmittance of separate diffraction gratings is shown in Fig. 6-20 with comparison of the transmittance data for bullseye structure.
In Fig. 6-20, with the red curve (A) the summation of transmittance of two individual gratings is shown, with the black curve (B) transmittance of bullseye structure is shown. And the blue curve (C) is the average of sum of transmittance of two individual diffraction gratings. Even though individual behavior of two diffraction gratings is not able to predict the peak positions, the sum of their transmittance or average of transmittances is able to predict peak positions as can be seen from Fig. 6-20.
Figure 6-1. SEM pictures of bullseye structures. a. (top left) Bullseye structure with $D_r = 1 \mu m$ before a central hole is drilled. b. (top right) Bullseye structure with the central hole. c. (bottom) Bare hole in silver film. Silver thickness is about 100 nm.
Figure 6-2. Fabrication steps, sideview of bullseye structure, figures are not to scale, they are exaggerated for illustration purposes. 

a. (top left) InOx (purple color) and PMMA (green) are coated onto the fused silica substrate. 

b. (top right) After EBL, development and silver liftoff. 

c. (bottom) After a central hole is drilled with FIB.
Figure 6-3. Optical transmission of Indium Oxide (InO$_x$) 3 nm thick flat film on fused silica. (experiment)
Figure 6-4. Optical transmission of a flat 35nm-thick silver film on fused silica. (experiment)
Figure 6-5. Optical transmittance of the bullseye structure ($D_g = 2 \, \mu m$) versus scaling parameter, $\lambda_s$. Dashed lines show diffraction thresholds. In parentheses indices $(i,j)$ corresponding to diffraction threshold are shown. $(1,0)$ threshold is at $1 \, \mu m$, $(1,1)$ threshold is at $0.7 \, \mu m$, and $(1,1)$ is at $0.447 \, \mu m$. (experiment)
Figure 6-6. Different periodicities of bullseye, red curve is for $D_g = 1 \mu m$ and blue is $D_g = 1.2 \mu m$. Arrows indicate peak positions for the corresponding data with the same colors. (experiment)
Figure 6-7. Transmittance vs. wavelength for bullseyes with different dielectric thicknesses. If the dielectric thickness is on the order of 100 nm, there is no peak around 1200 nm. Light blue curve (B) is for the experimental result, blue (D) is for computation of 100 nm thick gratings. But if the dielectric is made thick enough (400 nm) as can be seen from light green curve (A)(experiment) and dark green curve (C) (computation) the peaks associated with the diffractive threshold (vertical dashed line) are observed and have approximately the same transmittance. (experiment + computation)
Figure 6-8. Transmittance vs. wavelength for bullseyes with different dielectric thicknesses. The simulations are done for 1D slits with $D_g = 1 \mu m$, $a = 0.5 \mu m$. The respective PMMA (dielectric thickness) thicknesses are 200 nm (green) (E), 300 nm (red) (D), 400 nm (magenta) (C), 500 nm (blue) (B) and 600 nm (yellow) (A). Numerical results are for zero order transmission with TE polarization. The vertical dashed lines represent the first and second diffraction thresholds. (computation)
Figure 6-9. Dg=1.2 \( \mu \text{m} \) bullseye with A6 PMMA spun with 3000 rpm. The green curve (label G) for no central hole case, the cyan curve (label E) 2 \( \mu \text{m} \), the black curve 5 \( \mu \text{m} \) (F), the blue curve 7 \( \mu \text{m} \) (D), the magenta curve (C) 10 \( \mu \text{m} \), the yellow curve 12 \( \mu \text{m} \) (B) and the red curve (A) is for 16\( \mu \text{m} \) diameter hole in the center. Because of the space limitation only labels A, B and G are shown in the figure, labeling starts from top with A and goes down to G. The first order diffraction threshold is shown by the dashed line. (experiment)
Figure 6-10. Bare circular holes with diameters 16 (red), 12 (yellow), 10 (magenta), 7 (blue) and 5\,\mu m (black) on a 35nm flat silver film. (experiment)
Figure 6-11. Transmittance vs. wavelength for a bullseye structure with 300 nm dielectric thickness, with $D_g=1\mu$m. Different curves represent different silver thicknesses. Blue curve is for 17 nm, red is for 35 nm, yellow is 50 nm and navy is for 65 nm. Two bottom ones are, green 35 nm flat silver film and cyan for 70 nm flat silver films. (experiment)
Figure 6-12. Transmittance vs. wavelength of 1D grating for different silver thicknesses. The blue curve is for 20 nm (A), red 40 nm (B) and the navy curve (C) is for 60 nm thick silver film. Above in Fig. 6-11 experimental results for similar thickness bullseye structures are shown. (computation)
Figure 6-13. Bullseye with defects, green (A) with highest transmission is for the case when all the rings are present, i.e., no ring defects. The other curves represent cases (see text) when some rings are missing. Note that the lowest transmission (magenta, F) occurs when the innermost ten rings are removed. A6 PMMA (300 nm thick), 3000 rpm, Dg=1μm. (experiment)
Figure 6-14. Different ratios of a to Dg (its value (periodicity) is fixed, Dg=1 μm), a=0.3 μm red (A), a=0.4 μm blue (B), a=0.2 μm cyan (C), a=0.5 μm black (D) and green is 40 nm thick silver film (E). Dielectric thickness is 300 nm. (computation)
Figure 6-15. Optical transmittance of the bullseye structure as function of wavelength. The blue (upper) curve is for polarized light. Red (lower) curve is unpolarized light. (experiment)
Figure 6-16. The blue curve (E) is for TE polarization, the red curve (A) is for TM polarization, green (B) is for average of TE and TM. And the black curve (D) and the yellow curve (C) is for the experiment for 300 nm thick dielectric. (experiment + computation)
Figure 6-17. Separation of bullseye structure into two components.
Figure 6-18. First grating with its optical transmission. The black curve (A) is for the self-standing (without substrate), green (C) is for 600nm thick PMMA and the blue curve (B) is for 400 nm thick PMMA case.
Figure 6-19. The grating only with metal structure on top of it. The black curve (A) is for self-standing (without substrate) and the blue curve (B) is with the substrate. The metal thickness is 35nm thick silver.
Figure 6-20. The transmittance of bullseye structure (400 nm PMMA on a substrate) is shown with the black curve (B). The red curve (A) is for sum of transmittance of two individual gratings (of Fig. 6-18 (B) and Fig. 6-19 (B)). And the blue curve (C) is for the average of transmittance of two individual gratings (of Fig. 6-18 (B) and Fig. 6-19 (B)).
CHAPTER 7
CONCLUSIONS AND FUTURE WORK

7.1 Conclusions

In this thesis we have presented the results of our work on subwavelength optical structures. Enhanced optical transmission is shown for two different systems, hole arrays and bullseye structures. By varying the hole size and periodicity of the hole arrays, it has been shown that enhanced transmission can be explained without surface plasmon polariton excitations. By scaling geometrical parameters of the system it is shown that transmission spectra depend only on geometry but not metal properties. This excludes the role of surface plasmons. The enhanced transmission is attributed to diffraction and trapped modes. The resonant scattering due to trapped modes and its interference with diffracted modes leads to extraordinary optical transmission. Also in bullseye structures apertureless enhanced transmission due to the presence of trapped modes and cavity resonances is shown.

7.2 Future Work

To our knowledge enhanced optical transmission of periodic arrays of dielectric structures, hole arrays or cylinders have not been shown experimentally. This kind of work will further assure that trapped modes do not require metallic structures. Also another work will be on differentiating material resonances in materials (like SiC, GaAs, GaP) from trapped modes by characterizing hole arrays made in these polar materials.\textsuperscript{16} For the bullseye structures some of the results we have shown are only for numerical calculations. To further check validity of our numerical results, additional experiments, such as varying the width of the grooves to the periodicity, need to be performed. Also nonlinear effects using trapped modes have been predicted.\textsuperscript{77} We have shown that electric field strengths can be significantly enhanced in
subwavelength structures. Accordingly, nonlinear materials can be strategically placed within the structure for second harmonic generation and possible use in applications.
APPENDIX
GALLIUM ARSENIDE EXPERIMENT

In this section, briefly failed experiments on Gallium Arsenide (GaAs) wafers will be given. We tried to fabricate GaAs free standing hole arrays. We have used three different wafers, GaAs wafers, GaAs on Silicon wafers and GaAs on AlGaAs/GaAs wafers.

Initial works are performed with GaAs wafers. In Fig. A-1 reflectance of a square hole array on GaAs wafer as a function of wavenumbers is shown. The range of wavenumbers are from 100 cm\(^{-1}\) to 600 cm\(^{-1}\). The red curve is only for GaAs without any pattern and the blue curve is for 18 \(\mu\)m periodicity (a=9 \(\mu\)m) square hole arrays. The peak around 300 wavenumbers is due to ionic material resonance of GaAs due to its polar character. In these structures we were not able to observe diffraction effects. These wafers were not backside etched and were not free standing. In the second part, we have used GaAs (2\(\mu\)m) thick grown on Silicon wafers for our measurements. The transmittance as a function of wavelength for these type of wafers are shown in Fig. A-2. As a comparison with the red curve in this figure transmittance of GaAs wafers is shown. And with the black curve the transmittance for silicon wafers are shown. As can be seen from the figure silicon is not transparent between 5 and 20 \(\mu\)m range. The reflectance curve as a function wavelength for GaAs on Silicon wafers is shown in Fig. A-3. The pattern is 18 \(\mu\)m periodicity (a=9\(\mu\)m) hole arrays. All these three curves, red, green and black correspond to same structure measured for different times. Also for the red curve a larger shutter is used in the measurement to increase the amount of incident light on the sample. But these samples were not self-standing, so we have tried to etch this silicon layer using different wet and dry etches. This part of the experiment did not work well. So we have used another wafer, which is GaAs on AlGaAs/GaAs. The concentration of Al in AlGaAs is 0.6. This work is still in progress.
Figure. A-1. GaAs reflectance as a function of wavenumbers. The red curve is for GaAs substrates and the blue curve is for 18 μm periodicity hole array with a=9μm.
Figure A-2. Transmittance of GaAs and GaAs on Silicon. The red curve is for GaAs wafers and the blue curve is the transmittance of GaAs on silicon wafers.
Figure A-3. Reflectance of a=9, Dg=18\(\mu\) hole array on GaAs on Silicon wafer. The black and the green curve is for smaller shutter in the measurement setup and the red curve is for a larger shutter.
LIST OF REFERENCES


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BIOGRAPHICAL SKETCH

I was born in Ankara, Turkey. I lived there for 26 years. I have graduated from Mimar Sinan High School in Demetevler. I have attended Middle East Technical University in Ankara. Initially I have studied geology for one year before I have realized that I enjoyed physics more. In 2000, I have graduated with B.Sc. degree in physics. My interests were experimental condensed matter physics so I have attended to Bilkent University in Ankara for 2 years to get master's degree but before finishing that I moved to Florida to start Ph.D. studies at the University of Florida. I graduate from University of Florida in May 2008.