Nanostructures for enhancing transmission and local field intensity in metal films

by

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Abstract

A new nanostructure for enhancing transmission and local field intensity in thin metal films is presented. The novel double-hole array design was numerically modelled using a finite-difference time-domain technique. Simulations were performed for different array periodicities and hole spacing to optimize the structure for maximum enhancement capabilities. An optimum double-hole array was able to produce simultaneous increase in transmission and near-field intensity. The local field enhancement was found to be 4 orders of magnitude greater than the incident field and strongly localized to a nanoscale area which is promising for a variety of applications. Arrays of the double-hole design were fabricated using a focussed-ion beam on a thin gold film. Linear measurements through the milled arrays showed the predicted enhancement in transmission for the optimum double-hole configuration. Finite-difference time-domain calculations were also done to study an isolated rectangular aperture to show the dependence of transmission on polarization of the incident beam and width of the aperture. Fabry-Pérot resonances were shown to exist for different film thicknesses and the phase of reflection was calculated from the transmission results. A microfluidic device with an embedded surface plasmon sensor was developed and its sensitivity to changes in refractive index was shown.
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Chapter 1

Introduction

Nanotechnology involves the creation and utilization of materials, devices, and systems involving ultra-small shapes on the order of one-billionth of a meter - one nanometer - to about 100 nanometers. Thus, it will invariably influence virtually all industries from computers to opto-electronic devices, energy, chemicals, and pharmaceuticals.

Plasmonics involves confining of light in structures in order to slow down, enhance and manipulate light. This technology has the potential to revolutionize telecommunications, computation and sensing as it can combine the high speed ability of photonics and miniaturization of electronics. Plasmonics makes use of surface waves traveling along the surface of a conducting medium like metals or semiconductors. Plasmons have the same frequencies and electromagnetic fields as light, but their sub-wavelength size results in taking up lesser space.

Surface plasmons (SP) in metals aid enhanced transmission through subwavelength holes. A subwavelength hole is one whose dimensions are smaller than half the wavelength of the incident light. This is of interest in creating compact nanometer-scale sensor devices with a high degree of sensitivity that can be used to detect chemical and biological substances.

Due to SPs' proximity to the surface and non-radiative nature they can help in field enhancement and localization. This is particularly important for nonlinear optical processes such as second harmonic generation (SHG), surface-enhanced Raman
scattering (SERS) and supercontinuum generation (SCG). Increased local field intensities can be achieved in metal nanostructures by harnessing surface plasmons.

Enhanced transmission through subwavelength holes in arrays was first shown in 1998 [1]. Since then various hole shapes have been proposed to provide a predictable increase in the transmission and local field intensity.

A new nanostructure design is presented in this work that is capable of simultaneously increasing the transmission and the local field intensity. The field enhancements are localized to well-defined periodic sites and are 4 orders of magnitude greater than the incident field. This structure is also amenable to currently available fabrication techniques.

1.1 Research objectives

The first aim of this thesis was to validate a finite-difference time-domain (FDTD) algorithm to be used, by studying the transmission through isolated nanoholes in metal films. FDTD was used to investigate and characterize resonances in transmission through isolated rectangular apertures in metal films.

The primary objective of this research was to design and develop a new nanostructure that was capable of significantly increasing the transmission and local field intensity compared to other available designs.

The next goal was to fabricate arrays of the proposed structure using the focused-ion beam (FIB) milling technique and measure the transmission through them to verify calculations.

The last objective was to fabricate arrays for a surface plasmon sensor to be used as a detection element in a lab-on-chip device.
1.2 Organization of the thesis

Chapter 2 consists of a review of surface plasmon theory, the phenomenon of enhanced transmission through nanohole arrays, the shape effect of apertures and previously proposed nanostructures for local field enhancement.

Chapter 3 outlines the key features of the FDTD algorithm used for the numerical modeling of subwavelength apertures.

Chapter 4 presents the results of the simulations done on an isolated rectangular aperture to characterize transmission through them. This is followed by simulation results of the double-hole array structure showing enhancement in transmission and local field intensity.

Chapter 5 is a description of the FIB fabrication method used for milling arrays of the proposed nanostructure. The different parameters used for milling, calibration runs performed, issues faced while milling and results of the fabrication are presented in that section.

Chapter 6 describes the experimental setup used for measuring linear transmission through the arrays. The experimental transmission results of the double-hole arrays are then shown, followed by non linear measurements that are currently being done on the arrays. The fabrication and proof-of-concept results of a surface plasmon resonance sensor based microfluidic device are also presented.

Chapter 7 summarizes the work done and outlines future initiatives that can be undertaken.
Chapter 2

Literary Review

Introduction

The transmission of light through a subwavelength hole in a metallic film was recently shown to increase by many orders of magnitude when compared to standard aperture theory. This counter-intuitive result sparked a great amount of interest in the study of enhanced field transmission and confinement in nanostructures on metallic surfaces. Since then several theories have been proposed to explain the physics behind this phenomenon. In addition, many nanostructure designs have spawned which make use of this effect for applications ranging from non-linear optics and nanolithography to sensors and lab-on-chip designs.

My research work involved the design and fabrication of a nanohole structure incorporated in an array for simultaneous increase in transmission and field enhancement. The following section will review past literature on different theories proposed to explain the enhanced transmission through nanohole arrays in metal films, effect of the nanohole's shape on field enhancement, application in various fields and other nanostructures that have been proposed.
2.1 Enhanced transmission

Hans A. Bethe proposed a theory in 1944 to explain the diffraction of light through subwavelength holes in a perfect electric conductor [2]. He found that the transmission efficiency of light through such a hole scaled as:

\[
\left( \frac{a}{\lambda} \right)^4
\]  

(2.1)

where 'a' is the radius of the hole and '\lambda' is the wavelength of the incident light. This was widely considered as the standard in aperture theory for understanding transmission through "small holes" [2].

In 1998 an experiment by Ebbesen and co-workers showed that the transmission through subwavelength holes in a metallic film was dramatically increased when they were placed in a two dimensional lattice structure [1]. This was in contrast to Bethe’s aperture theory as it showed that the transmission, when normalized to the hole density in the array, was nearly twice as much as the incident light. The increased transmission was attributed to the "coupling of light with surface plasmons on the surface of a periodically patterned metal film" [1].
Figure 2.1: Silver film of thickness 200 nm with a hole diameter of 150 nm and an array periodicity of 900 nm [1]. The periodicity of the array is indicated as $a_0$.

Figure 2.1 shows the zero-order transmission spectrum obtained by Ebbesen's group from a square array of cylindrical holes in a silver film with a quartz substrate. The peak at 326 nm is the bulk silver plasmon peak, the wavelength at which silver is transparent, which disappears as the film thickness increases. The maximum in transmission is seen at a wavelength that corresponds to the lattice constant of the array and was shown to vary correspondingly for different periodicities. The minima are a result of Wood's anomalies that arise when a diffracted order is tangent to the grating plane. This suppresses the transmission at specific wavelengths corresponding to the metal-dielectric interfaces [3]. It was stated that the enhancement in transmission was independent of the type of metal, hole diameter or film thickness.

A theoretical treatment of the transmission enhancement in arrays showed that it could be due to tunneling through surface plasmons generated on either side of the metal film [4]. This was experimentally verified by showing that the transmission decayed exponentially with increasing film thickness [5].
An alternative method of introducing a periodic corrugation was done by placing a series of concentric grooves around a single nanohole, called the bull’s-eye pattern [6]. The wavelength of the transmission resonance in this case depended on the spacing between these grooves and transmission enhancements 125 times greater than the incidence have been achieved [7].

2.2 The surface plasmon

Surface plasmons are longitudinal electron oscillations formed when light is incident on a metal surface and are characterized by an exponential decay away from the surface. The momentum of the surface plasmon, which comes from the interaction of the surface charge density with the incident electromagnetic field, is greater than that of the free space photon. This momentum mismatch is overcome by providing a periodic corrugation or grating on the surface in the form of nanohole arrays which diffracts the incident light [8]. When the momentum is matched the photons can be coupled with the surface plasmons.

The dispersion relation for the surface plasmon is given by;

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

(2.2)

where \( \frac{\omega}{c} \) is the free space wave vector, \( \varepsilon_i \) and \( \varepsilon_m \) are dielectric constants of the dielectric and metal respectively [8]. The decay length of the field is close to half the wavelength of incident light while the decay into the metal surface is dictated by the skin depth [9]. Hence the wave is bound to the metal and is non-radiative in nature resulting in concentration of the field on the surface.

The distance that the surface plasmon can propagate along a metal surface depends on the absorption in the metal which is characterized by the frequency dependent
complex dielectric constant of the metal. Silver has the lowest losses in the visible spectrum and was shown to allow for surface plasmon propagation ranging from 10-100 μm and close to 1 mm in the near-infrared wavelength region [9].

The role of the metal surface for surface plasmon generation using nanoholes was shown by Grupp and co-workers where transmission spectra were obtained from nanohole arrays in nickel and silver. The transmission obtained through a free-standing layer of nickel, an optically bad conductor, was enhanced when it was coated with a 20 nm layer of silver [10]. Typically, if the ratio of the real part of the dielectric function to the imaginary part is much greater than one, then the metal is capable of surface plasmon generation. This clearly demonstrated the dominant effect of surface plasmons in the enhancement of light transmission through thin metal films apart from the diffraction of the grating.

2.3 Enhancement through diffraction

Another school of thought attributes the enhanced transmission to diffraction of light through the grating. Treacy argued that surface plasmons coupled to evanescent diffraction modes which in turn coupled to propagating modes thereby being part of a "dynamical diffraction" process [11]. This theory was formulated in terms of Bloch waves which represented the modulation of the transmission spectrum as a function of periodic variation of the surface lattice. Cao's complementary study involving an array of slits does not regard surface plasmons as contributors to the transmission, but interprets the enhancement as a result of waveguide-mode resonance and diffraction [12].

In a recent paper Barnes' group suggested the idea of surface plasmon assisted diffraction [13]. The incident light is diffracted by the array and evanescent waves are produced. Since the subwavelength holes cannot support propagating modes, these evanescent waves couple into the surface plasmons on the metal surface and tunnel through the holes. At the exit side they scatter and decouple out of the surface plasmons to get transmitted as enhanced and wavelength-shifted light.
Although the physics concerning the functioning of this phenomenon is still being developed, the idea of increased light transmission and field confinement at the nanoscale level is exciting for a wide range of applications.

In this work, the enhancement in transmission and local field intensity will be treated as a surface plasmon effect.

2.4 Shape of the nanohole

In Martin-Moreno’s paper which presented a three dimensional theoretical treatment of nanohole arrays it was stated that the shape of the hole had little or no effect on the transmission properties [4].

Experimental work done on elliptical arrays showed that the enhancement in transmission had a strong polarization dependence on the aspect ratio and orientation of the hole in an array [14].

Koerkamp and co-workers examined the rectangular hole shape using arrays made in a gold film. They showed that the normalized transmission was increased by an order of magnitude despite a decrease in the hole-area when compared to circular apertures [15]. This was later verified by Degiron’s group who performed similar measurements on a single rectangular aperture in a free-standing silver film [16].
Figure 2.2: Transmission spectra for different hole widths from Degiron’s experiments done on a single rectangular aperture in a 300 nm thick free-standing silver film [16].

Figure 2.2 shows the transmission spectra for various aspect ratios of a single rectangular hole. It is seen that the maximum in transmission occurs for the rectangular aperture with the smallest width. By varying the incident polarization, it was seen that for a polarization perpendicular to the long edge of the rectangle the highest transmission was observed. These two effects were a result of excitation of surface plasmons on the upper and lower ridges of the aperture. A notable feature in the plot is the red-shift seen in the peak resonance wavelength as the width of the rectangle is reduced. It has been theoretically predicted that with further decrease in the width of the rectangular hole, the transmission will have a huge enhancement and undergo a pronounced red-shift [17].

These works showed that the hole-shape could be used as an efficient tool to tune the enhancement factor of nanostructures. As a result, many other innovative designs were proposed to provide a predictable increase in the local field intensity.
2.5 Applications for nanostructures

When surface plasmons are excited they produce a strong field concentration on the surface of a metal that are confined to nanometric areas. The periodic array structure provides a uniform distribution of “active sites” of field enhancements and hence is suitable for non-linear optics, biological sensors, lab-on-chip designs and nanomicroscopy.

Surface-enhanced Raman scattering (SERS) can be a highly sensitive method that is capable of detection at the single molecule level. It is capable of producing strong Raman signals with a very narrow bandwidth from molecules that are attached to nanoscale metallic structures [18]. Hence a periodic array structure can facilitate uniform distribution of field enhancements as opposed to random “hot spots” caused by rough surfaces.

One study used arrays of different periodicities and observed a shift in the Raman scattering signal corresponding to changes in the periodicity [19]. The measurements were done in transmission mode and surface plasmon resonances were used to detect the “molecular fingerprints” of different substances that were adsorbed on the surface of an array.

Since enhanced transmission through nanohole arrays produces intensity build up in apertures, a simulataneous enhancement in non-linear processes like second harmonic generation (SHG) can also be achieved. This process is sensitive to surface variations and is one of the most direct methods of estimating the magnitude of field intensity on a surface [20].

SHG from an electrolytically roughened surface was shown to be 4 orders of magnitude greater than from a smooth surface [21]. SHG done on metallic array structures in transmission mode for a periodic and random arrangement of holes showed
a greater nonlinear signal occurring for the periodic case [22]. Changes in the shape of the hole were also shown to affect the SHG signal from an array structure.

When an intense laser beam interacts with a material, its spectrum can be broadened dramatically due to the nonlinear interaction between light and matter. This process is called supercontinuum generation (SCG). With the use of a resonant optical antennae structure, white light supercontinuum generation (WLSCG) was generated in a recent work and 3 orders of magnitude field enhancement was shown to occur [23].

Nanohole arrays can be used as sensors as they are based on the resonant surface plasmon enhanced transmission through the holes. When a material is placed on a nanohole array the resonance peak shifts as a function of its refractive index. Since the resonance is very sharp a high degree of sensitivity can be achieved. A prototype sensor to monitor the binding of organic and biological molecules to the metallic surface has been shown to operate with this surface plasmon resonance concept [24]. This idea can also be extended to fabricate a microfluidic based lab-on-chip device for the real-time monitoring of chemical processes.

2.6 Structures for field enhancement

In the last few years a number of new structures and ideas have been put forth to provide a predictable increase in the local field intensity and for a high degree of field confinement. These have found use in many of the applications mentioned earlier.

One design consisted of a self-similar arrangement of metallic spheres which acted as a nanolens to enhance the local field [25]. The size and distance between these spheres varied linearly and each sphere focused the field onto the smaller adjacent one until a high field concentration was achieved. This setup was capable of producing enhancement in the field intensity by 6 orders of magnitude.
Another design involved a 3D metallic cone with a radius decreasing from 50 nm to 2 nm. This design proposed propagating surface plasmons along the length of the taper which would gradually slow down as it approached the tip. This would cause "rapid adiabatic slowing down and asymptotic stopping" of the surface plasmons resulting in field build up at the end of the taper with 3 orders of magnitude increase in the field intensity [26].

Reliable methods of fabricating the above two structures have not been proposed so far and therefore remain theoretical ideas.

A bowtie antenna design consisted of an array of triangular gold islands separated by nanometric gaps of about 16 nm [27]. This structure was also capable of producing field enhancement by a factor of 1500. Electron beam lithography was used for fabricating this structure.

The optical antenna consisted of two strips of gold that acted as two arms with a separation of nearly 20 nm [23]. When illuminated with a laser source this design was capable of generating 3 orders of magnitude field increase resulting in white light supercontinuum generation (WLSCG). This design was fabricated using a focused-ion beam (FIB) milling to create a two dimensional lattice of these antenna units.

2.7 Summary

This chapter gave an account of the concept of enhanced transmission through arrays of subwavelength holes. This was followed by a brief discussion of surface plasmon theory and the opposing theories in literature that attribute the enhanced transmission primarily to either surface waves or diffraction of the grating.

The importance of the shape of the nanohole was elaborated as it is one of the main reasons for designing alternate nanostructures. Different theoretical designs that
have been proposed and shapes that have been fabricated thus far were mentioned. Various applications that require increased transmission and enhancement in the near field were listed, which serve as the primary motivation for this work.

This thesis involves the study and modeling of surface plasmon effects in thin metal films. A novel double-hole array design that is capable of producing enhanced transmission and simultaneous field intensity enhancements 4 orders of magnitude greater than the conventional diffraction limit will be presented. The enhancement is achieved by incorporating a nanostructure with a new shape in an array configuration.
Chapter 3

The FDTD Technique

Introduction

Numerical simulations provide a suitable means of predicting the capabilities of a nanostructure design. This helps in quantifying the enhancement factors that can be achieved from a structure before it is fabricated. The finite-difference time-domain (FDTD) technique is used to calculate the electromagnetic response of a system. This method has been used previously to study the behaviour of light in isolated and arrays of nanoholes [28]. Using FDTD modeling a nanostructure has been shown to produce a high resolution light spot [29].

Vital aspects of the FDTD code that dictate its working are outlined in this section.

3.1 FDTD features

The FDTD method uses the approach of direct time-domain analysis of Maxwell’s differential equations on spatial grids or lattices. It is a marching-in-time procedure that simulates an electromagnetic wave in a finite spatial region by sampling
data and propagating the numerical analogs in a virtual computer grid. It is accurate, robust and an explicit computational method that involves no linear algebra which generally tend to limit the size of the frequency domain integral equations [30]. The various features of FDTD are briefly explained below.

### 3.1.1 Yee Algorithm

Kane Yee formulated a set of finite difference equations for Maxwell’s curl equations system for the lossless material case [31]. The Yee algorithm solves both the electric and magnetic fields in time and space using Maxwell’s curl equations rather than solving the fields one by one. Though many other grid configurations have come out subsequently, none have surpassed the efficiency and robust nature of the Yee grid.

![Yee cell schematic](image)

Figure 3.1: Schematic of Yee cell showing position of the electric and magnetic field vectors [32].
The Yee cell configuration centers the electric (E) field and magnetic (H) field in three dimensions in such a way so that each E field is surrounded by four circular H fields and vice versa as seen in figure 3.1.

The finite difference expressions used are central difference in nature and second order accurate. The continuity of the tangential E and H fields is maintained across an interface of dissimilar metals if it is parallel to either one of the lattice coordinate axes. The placement of the E and H fields in the Yee cell and the use of the central difference operation on these components implicitly enforce the two Gauss laws. Therefore the Yee mesh is divergence free in the absence of electric and magnetic charge.

Figure 3.2: Schematic of the leapfrog time stepping sequence [32].

A key concept used in this algorithm is the leap-frog time stepping process. All E fields are computed at a particular time point using the previous H field value that is known and stored in memory. Then, the H fields are computed using the E field value
found earlier. In this manner the cycle repeats until the end of time stepping. A diagram depicting this is shown in figure 3.2.

This method involves explicit calculations and hence the laborious matrix manipulations and solving of simultaneous equations are not involved. It is non-dissipative in nature i.e., the numerical waves do not decay due to numerical artifacts. Formulation of finite difference expressions from Maxwell’s curl equations that frame the Yee algorithm are given in Appendix A.

### 3.1.2 Courant Stability factor

The Courant stability factor is crucial to the stable working of an FDTD code. By keeping the Courant number below or equal to its limit numerical stability of the algorithm can be sustained, failing which errors in computed values will grow exponentially [30].

The Courant number is given by;

\[ S = \frac{c \Delta t}{\Delta x} \]  

(3.1)

where \( \Delta t \) is the time step and \( \Delta x \) is the grid size used.

To yield accurate results, the grid spacing \( \delta \) in the finite difference simulation must be less than the wavelength used, usually less than \( \lambda/10 \). The stability condition relating spatial and temporal step size is given by;

\[ v_{\text{max}} \Delta t = \left[ \frac{1}{\Delta x^2} + \frac{1}{\Delta y^2} + \frac{1}{\Delta z^2} \right]^{-1/2} \]  

(3.2)
where $v_{\text{max}}$ is the maximum velocity of the wave. When the step size $\delta$ is the same in all directions, the Courant stability condition is given by:

$$\frac{v_{\text{max}} \Delta t}{\delta} = \frac{1}{\sqrt{N}} \quad (3.3)$$

where $N$ is the number of spatial dimensions in the problem.

A typical finite difference grid contains regions of different dielectric properties representing cell components. By specifying the dielectric properties, $\varepsilon(i,j,k)$ and $\sigma(i,j,k)$, of each region, the fields within an enclosed region can be determined.

For example if the wavelength used is 500 nm, the maximum step size is given by;

$$\delta = \frac{\lambda}{10} = 50nm$$

For a two dimensional problem, the time step, based on the stability condition of equation 3.3, would be 0.117 fs. The formulation of the Courant stability number for one, two and three dimensions is detailed in Appendix B.

### 3.1.3 Boundary Conditions

When the need for simulating an infinite structure (such as a waveguide) arises, an Absorbing Boundary Condition (ABC) is used wherein the incoming waves from the structure are absorbed without any reflections. This replicates the condition of a wave propagating to infinity. The Perfectly Matched Layer (PML) is a type of absorbing boundary condition formulated by J. P. Berenger [33].

This PML simulates the infinity scenario by terminating the outer boundary of the lattice in an absorbing material medium. This is similar to the physical working of an anechoic chamber. A lossy and dispersionless absorbing medium can be used, but the
problem arises when obliquely incident waves hit the layer which cannot be matched. In the PML, Maxwell’s equations’ vector field components are split into two orthogonal components which are made to satisfy a coupled set of first order differential equations. By choosing loss parameters consistent with a dispersionless medium, a perfectly matched interface can be obtained.

In a lossy medium the reflection is zero only for normally (plane) incident waves. But for oblique waves reflection is still present, to avoid this, the layer has to be placed far enough from the source for waves to be planar. This is not advantageous as far as resolution of the structure is concerned. Hence a nonphysical absorber is used that can be matched independent of frequency and angle of incidence by making use of the additional degrees of freedom arising from a field-splitting strategy.

The PML can be used to simulate various microwave and optical behavior such as dispersion calculations in metallic and dielectric waveguides. This is possible because the PML uses a local time and space working method; hence no knowledge of the modal distribution and nature of dispersion present in the material is required. It has a high degree of accuracy and can operate over a wide range of group velocities.

3.2 Advantages of the FDTD method

- It is an explicit and rigorous computation hence a high degree of accuracy can be achieved.
- The sources of errors are known and well documented; there is no possibility of errors due to unknowns, statistical or probabilistic errors, random walk and the like.
- Any change in shape or material of the structure is easily accounted for since it is eventually broken down into finite grid cells.
- Following the concept of mesh generation, the facility of using parallel computing is inherent to FDTD.
• Though it works on a nonphysical level, with development of visualization
techniques and related software, perception of a computed FDTD model is easier.

3.3 Advantages over other methods

• In asymptotic analyses there is difficulty when dealing with non metallic
compositions and volumetric complexity of structures. The concept of mesh
generation in FDTD tackles this issue.
• In methods that solve integral equations, apart from the calculations being
tedious, geometric details within a volume and size of the structure are a matter of
concern.
• In frequency domain finite element methods the incorporation of material
dispersion and nonlinearities is relatively difficult.

3.4 Disadvantages

• The biggest disadvantage of adopting the FDTD method is felt when there is a
lack of computational resources.
• The number of grid cells in the domain need to be finite and the boundary
conditions used are critical for accurate simulation of the structure.
• Since FDTD is a discretization process the step change in dielectric values at the
interface of two dissimilar media leads to a staircasing effect and yields a rough
(almost digitized) curve rather than a smooth one.

An FDTD solver developed by Lumerical Solutions Inc. [34] was used to model light
behavior in nanoscale holes in metal films. The transmission and field activity within
nanostructures was examined. Due to the large three dimensional domain sizes and fine
grid parameters (3 nm) used, multi-node parallel computing clusters were used to run the
simulations. The procedure to setup, queue and run parallel simulations on WestGrid [35]
using Lumerical’s FDTD is detailed in Appendix D.
3.5 Summary

Some of the key features of the FDTD code were discussed to illustrate its working principles. The FDTD method can be used to model light behaviour in nanostructures with good accuracy. Availability of computational resources in the form of parallel cluster systems makes numerical modeling using FDTD a helpful tool in testing new designs. Newer techniques to improve the existing algorithm are being developed with renewed interest. For example, working beyond the Courant limit [36] and development of a three dimensional unconditionally stable FDTD method [37] are some new developments.
Chapter 4

Simulation Results

A finite-difference time-domain (FDTD) method was adopted to study light behaviour through subwavelength apertures in thin metal films. Simulations of light transmission through an isolated rectangular aperture were done to test the working of the FDTD code used. Previously known dependence of transmission on polarization of the incident light and dimensions of the aperture were verified. Further investigation of this aperture led to the observation of Fabry-Pérot (FP) resonances appearing in the transmission for varying film thickness.

Once the FDTD code was validated, a new double-hole design incorporated in an array was modeled to gauge its enhancement capabilities. Simulations of the double-hole array structure showed that the transmission was greatly enhanced compared to an array of single holes. Also, a simultaneously increase in the local field intensity by 4 orders of magnitude greater than the incident field was observed.

4.1 Isolated rectangular aperture

A paper published by Degiron’s group showed the transmission of light through a single rectangular subwavelength aperture in a 300 nm thick free-standing silver film [16]. By varying the polarization of the incoming light they showed that transmission was maximum for a polarization aligned across the long edges of the aperture. They attributed
this to the excitation of localized surface plasmon (LSP) modes on the upper and lower aperture ridges which created optical tunneling of light through the metal film.

A single rectangular hole in a 300 nm thick silver film was simulated in three dimensions using *Lumerical's FDTD* software. Perfectly-matched boundary conditions were used along all three directions. The setup was illuminated by a plane wave with a wavelength range of 500 nm to 900 nm and a short pulse of 5 fs. At the exit side a frequency domain power monitor was placed to monitor the output light through the aperture. Another monitor was placed across the centre of the rectangular aperture to record field activity at the ridges of the hole.

![Figure 4.1: Plot of transmission through an isolated rectangular aperture. The 9-9-5 represents a grid size of 9 nm along the x and y direction and 5 nm in the z direction (propagation of the wave).](image)

The grid size of the FDTD domain was varied in order to achieve convergence. Figure 4.1 shows the transmission through a single rectangular aperture with dimensions of (270x105) nm in a 300 nm thick silver film. A grid size of 3 nm was chosen as it
showed good convergence and is a suitable value that can effectively capture surface plasmon (SP) effects.

### 4.1.1 Variation of hole width

![Graph showing normalized transmission vs. wavelength for different hole widths](image)

Figure 4.2: Transmission spectrum normalized to hole area for increasing hole widths of 105 nm, 185 nm, and 260 nm in the 300 nm thick silver film. (b)

The dimensions of an aperture play a crucial role in the enhanced transmission. As the hole size was decreased i.e., the long edges were brought closer together, transmission was dramatically increased with a pronounced red-shift seen in the peak resonance as seen in Figure 4.2. This is surprising considering that the transmission is increasing with decreasing hole area. This indicates that the region between the edges does not contribute significantly to the peak transmission but helps to allow the evanescent coupling of the plasmon modes on either side of the film. The red-shift is attributed to the effect of SP coupling between the upper and lower edges of the rectangular aperture [16].
4.1.2 Variation of incident polarization

![Transmission spectra graph](image)

Figure 4.3: Transmission spectra obtained from a 105x270 nm rectangular aperture for varying polarization angles. A schematic of the orientation of the incident beam’s electric field for 90 degree polarization is shown.

To further demonstrate the localization of SP activity at the edges of the aperture, polarization of the incident wave was varied from 0° to 90°. Figure 4.3 shows the transmission spectra for various polarizations. A distinct peak at longer wavelengths is evident for 90° polarization which gradually reduces and eventually disappears for 0° polarization. This indicates that the intensity of the peak is a function of the angle of polarization. The LSP modes are launched by the perpendicular components of the electric field. It is known that since LSPs are dipoles they oscillate along an axis normal to the polarization of the exciting wave.
Figure 4.4: Z-component of the electric on the exit side of the aperture clearly showing the localized field. Outline of the rectangular aperture is also shown.

Figure 4.4 shows the field localization on the edge normal to the electric field. The features observed in these spectra correspond well with the transmission results obtained from previous experiments [16].

4.1.3 Variation of film thickness

The enhancements shown experimentally by other groups were verified using FDTD simulations. Further study of the isolated rectangular aperture involved the variation of film thickness. The transmission spectra showed existence of Fabry-Pérot (FP) resonances. An FP resonator is a cavity with parallel reflecting surfaces in which a wave propagating in a direction normal to the surfaces resonates at a frequency determined by the distance between the surfaces.

Due to an impedance mismatch between modes in the rectangular aperture and free-space modes, there is a reflection of the mode when it approaches the surface interface of the film. Thus the aperture acts as a cavity with reflecting surfaces at the
ends. This problem is well studied for microwave waveguides, where the penetration of the field into the metal is negligible. It has been suggested that for a perfect electric conductor (PEC) waveguide, the reflection coefficient is relatively insensitive to the waveguide geometry [38], and found to be [39, 40];

\[ \Gamma = |r| \exp(j\varphi) = 0.25 \exp(-j0.42\pi) \]  \hspace{1cm} (4.1)

This shows that there is a negative phase-shift associated with the reflection. As a result a zeroth-order FP resonance (or displacement resonance) can exist for film-thicknesses that are even smaller than the half wavelength of light.

![Contour plot of transmission through a 270 nm by 105 nm rectangular hole in silver as a function of wavelength and film-thickness](image)

**Figure 4.5:** A contour plot of the transmission through a 270 nm by 105 nm rectangular hole in silver as a function of wavelength and film-thickness. The mode order are labelled \( m = 0, 1 \) and 2 and shown with dotted lines. The cut-off wavelength is shown with a dashed line.

The thickness of the silver film was varied from 200 nm to 1 µm and the corresponding transmissions at the exit side were recorded. Figure 4.5 shows the presence of the zeroth, first and second order FP resonances as peaks in transmission. The zeroth order mode exists at longer wavelengths and for very thin films, which is due to the negative phase of reflection between the mode in the hole and free space. For example, at
600 nm wavelength, the film thickness that provides a zeroth order FP peak is less than 200 nm. For thinner films, there is also considerable transmission past the cut-off wavelength due to evanescent coupling of the lowest order modes. The phase of reflection was calculated for the first order mode using the following equation:

\[ m\pi - \left( \frac{2m_{\text{eff}}L}{\lambda} \right) = \varphi \]  

(4.2)

where \( m_{\text{eff}} \) is the effective index obtained from a modal analysis of the structure.

Figure 4.6: Plot of phase of reflection as a function of wavelength as calculated from Figure 4.4 and Equation 4.2.

Figure 4.6 shows the phase of reflection calculated from Equation 4.2 and the first order resonance as shown in Figure 4.5. This phase shift is less than values found previously for microwaves [39, 40]. The phase shift becomes greater as the wavelength is increased and silver becomes a better conductor, thereby approaching the PEC case that is a valid approximation for the microwave regime.
The amplitude of reflection increases with the thickness of the film. The absolute value of the reflection coefficient, $|\Gamma|$, can be found from the following equation:

$$|\Gamma| = \sqrt{\frac{(a - 1)}{(a + 1)}}$$  \hspace{1cm} (4.3)

where $a$ is ratio of the maximum to the minimum value of the amplitude of the mode.

![Graph showing transmission as a function of wavelength for 600nm film thickness. The plot shows the extrapolated amplitude and minimum of the first order mode, m = 1.](image)

Figure 4.7: Plot of the transmission as a function of wavelength for 600nm film thickness. This plot shows the extrapolated amplitude and minimum of the first order mode, $m = 1$.

From Figure 4.7 which shows the transmission through a film of 600 nm thickness, the amplitude of the reflection coefficient was found to be 0.50. This calculation was taken from the FP resonance at 624 nm. A similar method was carried out for a 1μm thick film and the value of $|\Gamma|$ was found to be 0.47 for the FP resonance at 730 nm.
4.2 Double-hole arrays

A new double-hole design incorporated in an array was modeled to study the dynamics of the electric field within the structure. The calculations showed a significant increase in transmission and surface field intensity for an optimum double-hole configuration. These promising results led to the fabrication and optical testing of the double-hole arrays.

![Diagram of double-hole array](image)

Figure 4.8: Schematic of the overlapping double-hole showing the focusing of light at the apexes with arrows.

The double-hole structure is formed by gradually moving two holes closer to one another. They will eventually overlap and two apexes will be formed as seen in Figure 4.8. In this design, the apexes are much smaller than the size of the hole and hence the resolution at which the holes can be milled is not the limiting factor to fabrication. Rather, the precision of the centre-to-centre hole spacing and reproducibility of the hole diameter determine how reliably this structure can be fabricated. The holes serve to gather the incident light and channel it along the apexes as shown in Figure 4.8. As light slows down adiabatically in a tapered structure the field builds up at the tip [26]. Thus the built up field build at each apex interfere constructively at an optimum centre-to-centre
spacing of the holes and array periodicity to enhance transmission and the local field intensity.

The FDTD simulations were done for a 300 nm thick free-standing silver film with Drude parameters. The double-holes had diameters of 100 nm each that went through the entire metal film. Periodic boundary conditions were used to create the array in the x-y plane and 12 layers of perfectly-matched layer (PML) boundaries were placed normal to the surface of the film. The size of the grid along all three axes was chosen to be 3 nm to ensure that the SP effects were captured. A plane wave of normal incidence with a pulse of 30 fs and a centre frequency of 650 nm was used. The incident wave was polarized in the vertical direction (along y-axis) along the apexes of the double-hole structure. The total simulation time was set at 200 fs, which is beyond the time that the field took to decay to a negligible value.

Field monitors were placed on the entrance and exit sides with a spacing of one grid cell away from the metal surface. Another monitor was placed cutting through the apexes in the vertical direction to observe field activity in the apexes. A ‘movie’ monitor was placed on the top surface of the metal to observe the focusing effect of the apexes in the double-holes. Due to the large three dimensional domain regions and small grid size of the simulations, an 840 node parallel computer cluster facility (WestGrid) was used.

The distance between the double-holes was varied from a centre-to-centre spacing of 0 nm (single hole) to 150 nm (two single holes) to show the enhancement capability of the structure compared to single ordinary holes. The array periodicity was varied from 200 nm to 700 nm in steps of 50 nm to locate the configuration with the maximum field enhancement at the apexes. When the centre-to-centre hole spacing was 100 nm, the apexes had a single point of contact and numerical convergence could not be achieved. This issue was examined by running similar simulations on other FDTD solvers to confirm this anomaly.
Figure 4.9: Plot of transmission as a function of wavelength for the 100 nm hole spacing double-hole for different grid sizes.

Figure 4.9 shows the transmission through a double-hole array with a hole spacing of 100 nm for different grid sizes. It is seen that as the grid size is reduced, the transmission increases with a red-shift in the resonance peak and convergence is not achieved. A grid size of 3 nm was chosen so as to correspond with the value used for simulations in the previous section which showed good agreement with experiments.
4.2.1 Variations in the Centre-to-Centre Hole-Spacing

Figure 4.10 (a): Plot of transmission as a function of wavelength for a double-hole with centre-to-centre spacing of 90 nm and an array periodicity of 700 nm.

Figure 4.10 (b): Peak transmission intensity normalized to the incidence as a function of the centre-to-centre spacing between the holes. Diagrams of the double-holes are shown in grey to indicate different centre-to-centre hole-spacings.
Figure 4.10 (a) shows the transmission spectra through a double-hole with a centre-to-centre spacing of 90 nm and an array periodicity of 700 nm. Figure 4.10 (b) shows the peak transmission intensity as a function of the centre-to-centre hole-spacing in double-hole arrays with periodicity of 700 nm. From this curve, 90 nm was found to be the spacing that gave maximum transmission for the 100 nm hole diameter. The apex-to-apex gap was approximately 44 nm in this optimum structure. A more detailed study was performed close to the region of touching apexes for centre-to-centre spacing of 96 nm and 104 nm. For a closer apex gap of 28 nm, the transmission was reduced, which shows that merely reducing the apex gap does not necessarily produce the highest transmission. The 104 nm spacing, when the holes no longer overlap, had a reduction in the transmission, clearly showing the effect of apexes on transmission.
4.2.2 Field Enhancement with Array Periodicity

Figure 4.11: Peak electric field density as a function of the array periodicity. The incident field density at the source was 1 V/m².

Figure 4.11 shows the maximum field density of the 90 nm centre-to-centre hole spacing for varying periodicities of the array. The maximum enhancement is $1.75 \times 10^{4} \text{ V/m}^2$ with a uniform excitation of 1 V/m²; a four order of magnitude local field enhancement. The maximum field enhancement occurs in the vicinity of the apexes and at a wavelength of 490 nm. This wavelength does not match the Bragg resonances set forth by the periodicity of the array [1], hence it is the result of localized SP resonances of the double-hole structure. The periodicity of the array was varied and it was confirmed that the wavelength of maximum field enhancement did not follow the periodicity, but rather remained close to 490 nm (with a lesser field enhancement seen at 430 nm). This clearly indicates the dominant role played by the shape of the aperture on transmission and local field enhancement.
Figure 4.12: A logarithmic contour plot of the electric field density 3 nm away from the metal surface at a wavelength of 490 nm with centre-to-centre hole spacing of 90 nm and array periodicity of 450 nm.

Figure 4.12 shows a contour plot of the local field 3 nm above the surface of the metal film, where the measurements of Fig. 4.10 were recorded. The field is confined over the hole near the apexes with a full-width half-maximum of 3.3 nm measured in the y-direction. This shows that the double-hole structure is a good candidate for applications such as nonlinear spectroscopy on nanometric length scales, allowing for single molecule probing and detection. Unlike rough surfaces used for surface enhanced Raman scattering (SERS), the locations of field enhancement are well-specified in this double-hole array structure.
4.2.3 Simultaneous Field Localization and Transmission Enhancement

![Graph showing peak field density and transmission as a function of periodicity](image)

Figure 4.13: Peak electric field density (shown in grey) and transmission normalized to the hole density as a function of the array periodicity for the double-hole with 90 nm centre-to-centre hole spacing.

Figure 4.13 shows the local field enhancement and transmission intensity recorded at the transmission resonance of the periodic array. For comparison between different array periodicities, the enhanced transmission was normalized to the hole-density. The maximum in transmission occurred for a periodicity of 450 nm. This corresponds to a maximum in the local field intensity as found in Fig. 4.10. At this periodicity, the localized SP mode is coupled to the propagating SP modes, which leads to increased transmission and field density. Interestingly for the 500 nm array periodicity this is not the case and the local field density and transmission both experience a minimum. This occurs because the localized SP of the double-hole is off-resonance and interferes destructively with the propagating SP resonances of the array.
4.3 Summary

The ability of an FDTD code used to model light interaction in subwavelength apertures was demonstrated. The dependence of transmission of a rectangular aperture on polarization and hole dimensions was shown. For the single rectangular aperture FP resonances in transmission were shown for variation in the film thickness. The resonances were characterized and the phase of reflection of light at the interface was calculated.

Once the capability of the FDTD was established a new double-hole design was modeled. The design was optimized for hole spacing and array periodicity. The structure showed promising enhancement factors of 8 orders of magnitude in the local energy density. This design thus has good potential for many applications including nonlinear optics.
Chapter 5

Fabrication of arrays

Introduction

This section outlines the various steps involved in the fabrication of subwavelength arrays using the focused-ion beam (FIB) milling technique. This method allows for in situ high resolution fabrication of nanostructures in thin metal films on a glass substrate. The use of bottom-up methods to make arrays with specific hole shapes has not been perfected yet. Issues such as under-cutting of the metal and adhesion layer while employing such techniques pose a serious problem. Moreover, the use of the FIB method facilitates the simultaneous milling and imaging of the nanostructures being fabricated.

The equipment used was an FEI dual-beam Strata 235 focused-ion beam (FIB) and field-emission scanning electron microscope (SEM). It combines a SEM for high resolution imaging and a FIB with a metal ion source for nanoscale milling. The two columns placed 52° with respect to each other facilitate simultaneous fabrication and imaging of the milled structures. Figure 5.1 shows a picture of the FEI Strata DB 235 FIB SEM equipment.
5.1 Principle of working

In an SEM an electron gun is used to emit a beam of high energy electrons. These electrons are focused toward the anode using condenser lenses. The beam of electrons thus formed has a typical spot size of about 1 nm to 5 nm which is then deflected in a raster form over a rectangular field of view. As the primary electrons strike the surface they are inelastically scattered by the atoms and penetrate into the sample surface resulting in the emission of secondary electrons. These back-scattered electrons are then sensed by a detector to reconstruct the image. Hence the SEM is capable of giving depth of focus to the image being scanned.

The working principle of the FIB is similar to that of a SEM. Applying an electric field to the liquid gallium source results in the emission of ions. The use of a liquid metal ion source results in high intensity of emission over a small area. The stream of ions is focused using electrostatic lenses and the aperture size is controlled by varying the ion beam current. When the beam is rastered over a predefined area the accelerated ions
physically etch or sputter away the material in sub-micron geometries. Re-deposition of sputtered material can cause problems and must be taken into consideration when employing the ion milling procedure.

These methods are not restricted by diffraction which is the limiting factor of various lens-based spectroscopic methods. Since the resolution depends on the spot size of the beam, proper focusing of the beam without astigmatism is imperative for attaining good results. Astigmatism is an optical error occurring due to unequal magnification across different directions as a result of collimation errors. The stability of the beam is another important factor for achieving higher image clarity and milling accuracy. The system used here had a high degree of resolution and was capable of efficiently imaging 5 nm features. The milling, depending on beam stability, could be done with a spot size of nearly 7 nm. Since the deflection of the beam is computer controlled any desired shape can be fabricated accurately.

5.2 Stream Files

The FIB has a CAD based interface and provides a few predefined shapes which can be saved in the form of text files. Such a text file contains the various parameters required for milling the desired shape such as the x and y co-ordinates of the beam position, dwell time of the beam at each co-ordinate, number of times the milling needs to be repeated (looping parameter) and the total number of dwell points. Each co-ordinate corresponds to one pixel on the screen and its dimension depends on the magnification. For instance, at 5000× magnification one pixel translates to nearly 7 nm and covers a field of view of 30×25 μm².

To mill arrays of periodic nanostructures, text files in the form of stream files were generated using specific script files. Dimensions such as the diameter of the holes and array periodicity were given as input to the script files. The basis (circle, ellipse or double-hole) of the array was written as a separate module and was called upon at every
lattice point in the array. Each co-ordinate was then put into a matrix and rearranged sequentially before being written into a text file.

The script files were written in Fortran due to its modular nature and multi-platform portability. The script which generated stream files to pattern double-hole arrays is shown in Appendix E. Once the stream files were generated, they could be tested using the "Patterns" software package from FEI. This software translates the co-ordinates in the stream file to corresponding pixels on the screen and can thus be used for pattern verification.

5.3 Milling parameters

The various milling parameters need to be chosen judiciously in order to achieve the desired milling depth and accurate shape of the structure. The selection of these parameters varies depending on the type of material to be patterned and the thickness of the material. The software provided preset values for silicon and germanium but since the sample used was gold, suitable values had to be picked after a trial and error process which involved multiple calibration runs. The different milling parameters include:

- accelerating voltage of the source
- ion beam current
- magnification and field of view
- dwell time of the beam at each co-ordinate
- looping of the milling process

The first three parameters can be selected from the FIB software interface while the remaining parameters are to be incorporated into the stream files.

The accelerating voltage and ion beam current are proportional to the velocity and the amount of ions being discharged from the source. Hence these directly alter the amount
of material that is removed from the sample surface. A higher accelerating voltage and beam current will mill a deeper hole but at the expense of broadening its desired dimension. The dwell time affects the milling in a similar manner. Hence optimum values have to be found for these parameters after making acceptable trade-offs in terms of resolution (dimension) of the structure and the time taken to mill an array.

Alternatively, the beam can be made to revisit a co-ordinate any number of times by setting the looping parameter accordingly in the stream file. Thus by choosing a low beam current and making the beam revisit every co-ordinate repeatedly a specific number of times a better shape resolution can be attained.

Due to memory constraints of the equipment's data acquisition system the number of dwell points was limited to one million. This prevents milling of large arrays which have a high hole density. Since the magnification defines the field of view and corresponding pixel dimension, this constraint can be overcome by increasing the magnification. This would automatically translate to a larger dimension of each co-ordinate. Another method to get around this issue is to interleave the co-ordinates when writing into the matrix and using an increased beam current or dwell time to accommodate for the skipped co-ordinates.

The sample used was a 100 nm thick gold film on a glass substrate that was 3 mm thick. A 5 nm layer of chromium was used to bond the gold layer with the glass substrate. Since chromium is an optically bad conductor, the nanoholes were milled through both the gold and chromium films by choosing appropriate milling parameters.
5.4 Calibration of stream files

In order to obtain the optimum milling parameters for the sample under consideration, a calibration run was done by varying the dwell times. Figure 5.2 shows one such calibration run that had varying dwell times from 0 ns to 3000 ns. It can be seen that for the last line (3000 ns) the milling is complete without any imperfections. There was also good correspondence between the input and the milled dimensions of the line.

![Figure 5.2: Calibration run for varying dwell times of the beam for each milled line. The scale bar corresponds to a length of 5 μm.](image)

Numerous such calibration runs were performed before settling for an accelerating voltage of 30 keV and a beam current of 100 pA. The dwell time of the beam at one pixel was chosen to be 3000 ns. A lower beam current and longer dwell time was chosen to mill through the 100 nm gold film and chromium layer while allowing for a high resolution of the double-hole structure’s dimensions. The magnification was chosen to be 5000× and each milled array covered an area of 30×25 μm².
5.5 Issues with the FIB

A number of issues affected proper milling of the nanostructured arrays. Some of the problems encountered and solutions for them have been outlined below.

As mentioned earlier, the astigmatism and instability of the beam play a detrimental role in achieving the desired dimensions and shape of the structure. Figure 5.3 (a) shows the image of an isolated double-hole milled with a high degree of astigmatism in the x direction. Figure 5.3 (b) shows a double-hole array milled when the beam was highly unstable clearly indicating the extreme effect of instability.

![Image of SEM images](image)

Figure 5.3: SEM images of (a) an isolated double-hole structure milled with astigmatism resulting in a double-ellipse structure and (b) an array of double-holes with beam instability.

The astigmatism should be removed by systematically stigmating the beam in the x and y directions for increasing steps of magnification up to ten times higher than the final magnification to be used. For this milling the beam was focused and stigmated at a magnification of 50000× for favourable results. Beam instability may arise due to insufficient heating of the source or lack of proper beam alignment.
5.6 Images of patterned double-hole arrays

Once the beam is aligned, focused and stigmated to an optimum condition the milling of a typical array having close to 3000 holes will take an average time of 120 s. Figure 5.4 (a) shows an array of double-holes with a diameter of approximately 210 nm and an array periodicity of 750 nm. Figure 5.4 (b) is a tilted image of another array of double holes images at an angle of 52° which clearly shows the complete milling of the holes through the gold film.

![Figure 5.4: SEM image of (a) double-hole array showing consistent and good shape of the nanostructures and (b) tilted image showing the depth of milling.](image)

Figure 5.5 shows isolated images of individual double-holes milled with an average diameter of 250 nm and an array periodicity of 750 nm.
Figure 5.5: SEM images of isolated double-holes with varying centre-to-centre spacing, from two separated single hole array (1) to a single hole array (12). The magnification of each image is not the same.

5.7 Other issues

Some additional precautions that needed to be taken while working with this specific FEI Strata DB 235 SEM FIB at Simon Fraser University were;

- In certain cases when the stream file is loaded, the screen might display co-ordinates that are out of the field of view, the software then displays an error message – “Not all patterns are within the field of view”. This can be overcome by using a program called “runscript.exe” to run a secondary script file called “millstream.psc”. This script will override the software and force the FIB to continue milling despite the co-ordinates being out of the field of view.
• The door of the vacuum chamber needed to be held closed while the chamber was being pumped up.

• When performing a single scan of an image the "Scan" button had to be clicked only once to prevent exiting of the FIB software.

• When some patterns were loaded the "Time" tab on the patterning window, which indicates the milling time, would display "00:00:00". To overcome this an appropriate time had to be entered to continue milling.

5.8 Summary

The various steps involved in the fabrication of the double-hole arrays using the FIB method and the issues related with it were discussed. SEM images of double-hole arrays milled for different hole spacing and periodicities were presented. The FIB milling technique is very useful as it allows for fabrication and imaging of the arrays simultaneously.

Although this method was capable of achieving the expected results, other methods to produce such arrays with high repeatability and yield are being investigated, including self-assembly techniques.
Chapter 6

Experimental Setup and Transmission Results

Introduction

The fabricated double-hole arrays were tested to verify the enhanced transmission obtained from numerical simulations. The experimental setup used for the optical testing of the various double-hole arrays is discussed in this section.

This chapter presents the results of transmission through double-hole arrays for varying hole spacing. As the holes moved closer and overlapped, an optimum centre-to-centre distance between the holes produced the highest transmission. This peak transmission was greater than an array of single holes and was greater than the maximum that could be achieved with a pair of single holes. The effect of the apexes was clearly demonstrated by the increased transmission values. Moreover, as the distance between the apexes was reduced, a red-shift was observed in the peak resonance.

Advanced optical testing currently being done on the double-hole arrays show a significant enhancement in the second harmonic signal for the optimum centre-to-centre hole spacing. This demonstrates the field enhancement capability of the double-hole structure and serves as a good validation of the design.

The other results include proof-of-concept transmission spectra obtained from a FIB-fabricated surface-plasmon resonance (SPR) sensor. The SPR is to be used in a
microfluid-based lab-on-chip device as a sensor for real time monitoring of chemical processes and characterization of protein and DNA.

6.1 Experimental Setup

The transmission spectra from the subwavelength arrays were obtained using the experimental setup shown in figure 6.1.

![Experimental Setup Diagram]

Figure 6.1: Schematic of the experimental setup used for linear measurements in the Chemistry department at University of Victoria.

The arrays were illuminated with white light from a halogen source from the gold side using an Olympus BHSM metallurgical microscope. The incident beam was collimated using a pair of lenses and focused down to a spot using a 100× microscope objective. The arrays were spaced 100 μm apart to allow the focused beam, covering an area of approximately 2800 μm², to illuminate the entire area of each individual array. The transmission through the arrays was measured using an Ocean Optics USB-2000
spectrometer coupled with a 400 µm core broad-area optic fiber. The acquisition time when recording the spectrum was set at 100 ms for a source voltage of 12 V.

Prior to measurements the sample was cleaned using an oxygen-plasma cleaner and an ultrasound bath. The procedure involved cleaning the gold surface in the plasma oven for 30 min followed by sonicating the sample in ultra-pure water.

![Transmission spectrum](image)

**Figure 6.2:** Transmission spectrum recorded through a double-hole array with 195 nm centre-to-centre hole spacing and an array periodicity of 750 nm.

Figure 6.2 shows the vertical components of the spectrum obtained from a double-hole array with 195 nm centre-to-centre hole spacing. The spectrum obtained was normalized to the incident light and compensated for the dark current of the detector. The spectrum clearly shows the peak at the transmission resonance of around 800 nm which follows the periodicity of the array. The dip in the transmission due to the Wood’s anomaly is also seen at nearly 750 nm [1].

### 6.2 Experimental transmission results through double-hole arrays
The sample used was a 100 nm thick gold film on a glass substrate with a 5 nm chromium layer used for adhesion. The holes had an average diameter of 200 nm and were milled through the entire gold film. The transmission was collected 3 cm away from the sample on the glass side and obtained using normally incident unpolarized light. A polarizer was placed between the sample and the fiber to obtain the s-polarization transmission through the arrays.

6.2.1 Transmission Spectra

![Graph showing transmission spectra through 200 nm diameter double-hole arrays for different centre-to-centre hole-separation of 0 nm (single hole), 195 nm (overlapping holes) and 250 nm (separated holes).](image)

Figure 6.3: Transmission spectra through 200 nm diameter double-hole arrays for different centre-to-centre hole-separation of 0 nm (single hole), 195 nm (overlapping holes) and 250 nm (separated holes).

Figure 6.3 shows the measured transmission spectra for the array of single holes and two different arrays of double-holes where the centre-to-centre separation is 195 nm and 250 nm. The recorded transmission was normalized to the incident spectrum of the halogen source. For the 195 nm double-hole array, the holes were overlapping to produce apexes with a gap of 35 nm. The 250 nm holes were non-overlapping.
The single-hole array and the 250 nm double-hole array show similar spectral features to past experiments [2, 42]. There are Bragg resonance peaks in transmission due to the array for the (1,0) resonance at 770 nm and for the (1,1) resonance at 600 nm. As expected, the 250 nm double-hole array has an overall larger transmission with respect to the single-hole array due to the increased number of holes.

Different characteristics are observed for the 195 nm double-hole array, when there are apexes present in the structure. The transmission peak is red-shifted and enhanced; for example, the 770 nm peak shifts to 800 nm and the magnitude of the transmission peak is more than doubled with the introduction of the apexes. The enhancement is not as strong for the (1,1) resonance, which has a polarization at $45^0$ with respect to the axis through the apexes.

The observed doubling in transmission is for the same overall hole-area, so this effect is expected to arise entirely due to the apexes. Therefore, this reiterates that there is significant modification to the local field in the region of the apexes, which is promising for further investigations of enhancement to the local field intensity in the double-hole apex structures.
6.2.2 Peak Transmission vs. Hole-Separation

Figure 6.4: Peak transmission normalized to the incidence as a function of the centre-to-centre hole-separation. The diagrams of the double-holes are shown in grey to illustrate the different hole-separation.

Figure 6.4 shows the normalized (1,0) peak intensity for different hole-separation. The intensity increases monotonically and abruptly decreases when the holes are no longer overlapping, beyond 200 nm separation. For the separated double-holes, the peak transmission reduces, but is less than double the single-hole value. The closest apex gap of 35 nm does not give the highest transmission; although the transmission values of the 190 nm and 195 nm hole separation are similar, the 65 nm apex gap has a higher transmission. This clearly indicates the contribution of the apexes in the double-holes to the transmission.
6.2.3 Peak Wavelength vs. Hole-Separation

Figure 6.5: Peak wavelength as a function of the centre-to-centre hole-separation. The array periodicity of all the arrays was 750 nm.

Figure 6.5 shows the peak wavelength of the (1,0) transmission resonance for varying hole separation. As the hole separation increases from zero, the increase in wavelength is greater as the apexes approach closer to each other. For a 35 nm apex gap in the 195 nm hole-separation the wavelength red-shifts to a value close to 800 nm. It can be seen that even for the 190 nm case when the apex gap is 65 nm; the wavelength still has a pronounced red-shift.

This red-shift in the wavelength of the (1,0) resonance can be partially explained by the fact that the double-hole structure is being made wider, which increases the cut-off wavelength of the hole. Past work on rectangular holes, however, showed that reducing the aspect ratio of the holes, without increasing the width, also increases the cut-off wavelength [15-17]. Therefore, it is expected that bringing the apexes closer together also increases the cut-off wavelength. When the double-holes are separated, the wavelength of the transmission peaks drops to near the single-hole value.
6.3 SHG from double-hole arrays

The double-hole array structure has been shown to produce highly localized enhanced electric fields (see Figure 4.12). Thus the structure would be capable of second-harmonic generation (SHG) since the efficiency of SHG is proportional to the square of the local field intensity. It is one of the most direct methods of estimating the magnitude of field intensity on a surface. This process is sensitive to surface variations and can hence serve the purpose of investigating the effect of apexes in the double-hole structure.

The double-hole arrays that were fabricated are currently being tested for SHG (by Dr. Antoine Lesuffleur) to experimentally gauge the electric field intensity in the apexes. Enhanced SHG measured in transmission was found to be dependent on the incident light polarization and the hole-spacing. SHG is not possible in centro-symmetric materials like bulk gold, but it may be observed at the surface where the symmetry is broken. The SHG from arrays of single holes have been shown to depend on the angle of incidence [22]. Hence the angle of the incident beam was set at $10^0$ to break the centro-symmetry of the double-hole design.

For the SHG measurements, a Ti:sapphire 30 fs pulsed laser with center wavelength at 800 nm was used. Due to the operating wavelength of the laser, the (1,0) resonance was examined for the SHG experiments, which has a similar resonant wavelength. The SHG was measured by a streak camera operating in synchroscan photon-counting mode. Due to fabrication irregularities more arrays were milled close to a hole spacing of 200 nm. The SHG signal from the arrays reflects the resolution of the apexes, which plays a crucial role in field confinement and enhancement.
Figure 6.6 shows the measured SHG in transmission geometry. There is a strong dependence on polarization; the vertical polarization SHG signal was up to 30 times the SHG signal from horizontal polarization. This is consistent with the linear transmission measurements. Under the same conditions for an array of single holes (not shown) with the same periodicity, only 4 counts were recorded.

A strong peak in the SHG was observed for the optimum centre-to-centre hole spacing of 195 nm with 5600 counts. The SHG signal from this array is enhanced by 10 times with respect to the 130 nm hole-spacing value. For the linear measurements, the transmission was only 1.5 times greater between the 130 nm and 195 nm hole-spacing arrays. A simple consideration of the squared dependence of SHG on the intensity would suggest that the SHG signal should be increased by only 2.25 times between the two arrays. Therefore, an additional enhancement of 4.44 times is seen for the 195 nm hole-spacing. Other hole-spacings of do not show similar enhancements; therefore the proximity and sharpness of the apexes are important to this phenomenon.
The observed SHG is a good measure of the double-hole design's field enhancement capability. This is promising because the double-hole structure can be readily fabricated and the same design may be applied to SERS and other demonstrations of nonlinear optics.

### 6.4 Microfluidics

A microfluid-based device with an embedded surface-plasmon resonance (SPR) sensor is currently being developed as a collaborative effort by groups from the Chemistry, Mechanical and Electrical Engineering Departments at University of Victoria. This device will be capable of real-time monitoring of on-chip chemical processes. When chemical analysis is done at the micro-scale the system has greater speed, low cost and reagent consumption, high sensitivity, efficiency, safety and automation [43, 44].

In an SPR process the resonance is sensitive to the adsorption of species at the metal surface [8]. Hence nanoscale biosensing on a microfluidic chip is possible using this phenomenon. In the prototype made, the sensor contained arrays of single nanoholes with varying periodicities. Each array had its characteristic narrow resonance at the corresponding Bragg resonance wavelength. The introduction of a substance with a different refractive index will cause the resonance to shift, since it is very sensitive to the surface, thereby detecting the particular substance.
Figure 6.7: Image of the microfluid-based SPR sensor with a PDMS layer on a gold film.

Figure 6.7 shows an image of a preliminary lab-on-chip prototype that was developed. The outlets and the inlet can be seen at one end of the device and the arrays are situated at the opposite end to provide greater clearance for the input and output tubes that carry the solution. Channel structures are used to flow solution through the arrays and transmission through the arrays are measured by shining light from the top. The arrays in the SPR sensor were fabricated by me and the channel structures were made by Angela DeLeebeek at the Microfluidics laboratory.

The transmission spectra through these arrays are to be recorded using a setup similar to the arrangement used for the double-hole arrays (see Figure 6.1). Single hole arrays with varying array periodicities were fabricated using the focused-ion beam (FIB) method. Initially, to allow for greater clearance between the sample and objective lens multiple arrays were “stitched” together to increase the size of an array.
Figure 6.8: A 3x3 set of arrays with a periodicity 525 nm with hole diameter of 110 nm. The bar indicates a length of 20 μm.

Figure 6.8 shows nine arrays stitched together to cover a large area of 90×90 μm² with nanoholes. It is seen in the figure that improper stitching led to overlap in certain zones and empty areas when there was an offset in the milling. This affected the resonance of the arrays due to errors caused by phase-mismatch of the coupling between adjacent arrays. To provide for greater clearance, a long-range microscope was used to overcome this difficulty. The iris of the microscope was closed down to a level where only one array was in focus. Subsequently, isolated single hole arrays with different periodicities were milled and the transmission through them was measured.
Figure 6.9: Transmission spectra through single hole arrays recorded in air with different periodicities.

Figure 6.9 shows the non-normalized transmission spectra through the arrays for three different periodicities in air without any channel structure. The shift in resonance for the change in periodicity is clearly seen. The magnitude of transmission through the arrays decreases with increase in periodicity due to the lesser number of holes available for transmission.
Figure 6.10: Transmission spectra through single hole arrays with a periodicity of 450 nm. These measurements were done by Angela DeLeebeek.

When a substance with a certain refractive index is placed on the arrays a shift in the resonance will be noticed. A shift of up to 5 nm can be typically sensed thereby providing an excellent means of high sensitivity detection. Figure 6.10 shows the shift in peak resonance for an array with periodicity of 450 nm. Plain water had a refractive index of 1.332 and that of the sucrose solution was 1.356.

The resonance peak for the 450 nm array is characteristically narrow and placed at a wavelength close to that of an IR laser. The sensitivity of the SPR can be observed by flowing liquids of varying refractive indices over the arrays and then re-measuring the transmission. The light intensity will thus vary depending on which fluid flows over the nanoholes.

Once the sensitivity of the SPR is verified, detection of different chemical and biological substances adsorbed at the nanoholes will be attempted. Preliminary
measurements have shown an appreciable shift in the resonance when transmission through solutions of different refractive indices was measured.

6.5 Summary

Optical testing of linear transmission through a novel double-hole array structure was done. Increased transmission was observed for the arrays that had double-holes with apexes and was shown to correspond well with the numerical calculations. The experimental setup used for the measurements was outlined.

Non linear measurements have shown enhanced SHG signal for the optimum double-hole array thereby validating the double-hole design’s capabilities. Further systematic studies of different angles of incidence and different hole-shapes are currently underway to maximize this effect. The observed SHG is promising because the overlapping double-hole structure is readily fabricated and the same design may be applied to SERS and other demonstrations in nonlinear optics.

The fabrication of an SPR sensor for a microfluid-based on-chip device was shown. The working principle and steps in progress for the realization of such a device were outlined. Results of preliminary transmission measurements done to detect changes in refractive index of substances were shown to validate the sensitivity of the SPR sensor. The next step would be to adsorb chemical or biological species on the array surface and characterize the substances by spectroscopic methods.

The double-hole arrays used for SHG were milled by me using the FIB method and the nonlinear measurements were done by Dr. Antoine Lesuffleur. My contribution to the microfluid-based device involved fabrication of single-hole arrays for the SPR sensor and performing preliminary proof-of-concept linear transmission measurements.
Chapter 7

Conclusions

A new nanostructure was presented which incorporated a novel double-hole design in an array. Using a finite-difference time-domain (FDTD) method the transmission and field enhancement abilities of the structure were examined. An optimum hole spacing and array periodicity was shown to produce simultaneous increase in the transmission and local field intensity. The field was highly localized to a nanoscale region with an increase of 8 orders of magnitude in the local energy density.

A focused-ion beam (FIB) was successfully operated to fabricate arrays of the double-hole design. Several arrays of double-holes with a fixed periodicity for varying hole spacing were milled. Linear transmission measurements through these arrays were done using spectroscopic methods which showed enhancement in transmission and pronounced red-shift in the resonance peak for the arrays with apexes. This qualitatively corresponded with a similar trend found in the simulations that showed the effect of apexes on transmission.

Ongoing second-harmonic measurements show a significant increase in the SHG signal for the optimum double-hole structure which is further proof of the enhancement capabilities of the design. These results are promising for nanophotonic applications that require strong local field intensities confined to nanoscopic regions. Such applications include nonlinear processes such as surface-enhanced Raman scattering (SERS) and supercontinuum generation (SCG). Due to the nanometric field confinement,
spectroscopy at the single molecule level may also be attainable by using the proposed double-hole array structure.

This thesis also presented the study done on an isolated rectangular aperture in a metal film to characterize the enhanced transmission resonances using the FDTD technique. The existence of Fabry-Pérot resonances in transmission was shown for variation in the film thickness. The FDTD algorithm used was validated by performing first-time numerical calculations of the single rectangular hole that agreed well with experiments done previously by others.

In addition, a surface plasmon sensor for a microfluid-based lab-on-chip device was fabricated. Linear transmission measurements through the single-hole arrays were done to show the working and sensitivity of the sensor. The final goal of this device would be to monitor real time chemical and biological processes.

The results of this thesis have been submitted for publication elsewhere as follows:

2005. (Presenting results of transmission measurements of double-hole arrays from Chapter 5)

- L. Kiran Swaroop Kumar and R. Gordon, "Localized field enhancement in metal films using an overlapping double-hole nanostructure," CLEO conference, 2006. (Presenting results on double-hole arrays from Chapter 4 and 6)


Future work would involve non-linear measurements of the double-hole arrays in transmission and reflection to comprehensively study the structure and determine its field enhancement capability. This would help quantify the amount of field intensity that a double-hole structure is capable of concentrating in its apexes. The promising SERS capability of this structure is to be investigated as a logical step forward.

The shape of the structure plays a key role in the enhancement ability of a nanostructure. At the nanoscale level even small defects that occur during the fabrication process can be detrimental to the ultimate performance of the nanostructure. Hence it is imperative to develop suitable methods for achieving a high degree of resolution of the designed structure. A self-assembly technique involving colloidal spheres would prove a good option for better shape resolution [45]. This would also be less time-consuming and laborious compared to the FIB method of fabrication and would facilitate a convenient method of achieving high through-put.
The double-hole design offers a straightforward method of fabricating apexes in arrays for high field localization and enhancement. The apexes are responsible for significant enhancements and their sharpness and resolution are crucial. Alternate designs that incorporate the apex element such as double-ellipses and single peak structures are to be developed. The double-ellipses would have longer apexes and can be expected to achieve greater local field intensities. Single apex structures would provide a means of breaking the centro-symmetry even for normal incidence for nonlinear applications.

The development of the mirofluid-sensor device is to be done to eventually enable protein and DNA characterization. An additional initiative would be to incorporate the double-hole design in the SPR arrays for greater sensitivity.
Bibliography


Appendix

A. Yee Algorithm:

Maxwell's curl equations for an isotropic medium are;

\[
\nabla \times \vec{E} = -\mu \frac{\partial \vec{H}}{\partial t} \\
\nabla \times \vec{H} = \sigma \vec{E} + \varepsilon \frac{\partial \vec{E}}{\partial t}
\]

(A.1)  (A.2)

The above equations can be written as six scalar equations in Cartesian coordinates.

The grid coordinates are defined in the Yee algorithm as;

\[(i, j, k) = (i\Delta x, j\Delta y, k\Delta z)\]  \hspace{1cm} \text{(A.3)}

where \(\Delta x, \Delta y\) and \(\Delta z\) are the grid size in the domain.

As a function of space and time, it can be represented as;

\[F'(i, j, k) = F(i\Delta x, j\Delta y, k\Delta z, n\Delta t)\]  \hspace{1cm} \text{(A.4)}

where \(\Delta t\) is the time increment, \(n\) is the time index and \(\delta = \Delta x = \Delta y = \Delta z\). The spatial and temporal derivatives of \(F\) are written using central finite difference approximations as;
\[ \frac{\partial F^n(i, j, k)}{\partial x} = \frac{F^n(i + 1/2, j, k) - F^n(i - 1/2, j, k)}{\delta} \] (A.5)

\[ \frac{\partial F^n(i, j, k)}{\partial t} = \frac{F^{n+1/2}(i, j, k) - F^{n-1/2}(i, j, k)}{\delta} \] (A.6)

The central differences are taken to the right and left of the observation by \( \Delta x / 2 \). This was done to interleave the E and H fields in the space lattice at intervals of \( \Delta x / 2 \). Similarly, the time finite difference is \( \pm 1/2 \Delta t \) to accommodate for the leapfrog algorithm.

Applying equations A.5 and A.6 to the six scalar equations these finite difference expressions in a space region can be formulated. They have a continuous variation of material properties contained in coefficients which are updated for every field vector component before the time stepping starts. In this manner the variation in material properties is accounted for. The three dimensional equations can be further broken down into two dimensions i.e., the TE and TM wave equations by assuming that all the partial derivatives of the fields with respect to \( z \) are zero. The equations for the TMz mode are given below;

\[ E_z^{n+1/2}(i, j + 1/2, k) = A_{i,j+1/2,k} E_y^n(i, j + 1/2, k) + B_{i,j+1/2,k}[H_x^{n+1/2}(i, j + 1/2, k + 1/2) \]
\[ - H_x^{n+1/2}(i, j + 1/2, k - 1/2)] + H_z^{n+1/2}(i - 1/2, j + 1/2, k) - H_z^{n+1/2}(i + 1/2, j + 1/2, k) \]

\[ H_x^{n+1/2}(i, j + 1/2, k + 1/2) = H_x^{n-1/2}(i, j + 1/2, k + 1/2) \]
\[ + \frac{\Delta t}{\mu \delta}[E_y^n(i, j + 1/2, k + 1) - E_y^n(i, j + 1/2, k) + E_z^n(i, j + k + 1/2) - E_z^n(i, j + 1, k + 1/2)] \]
\[ H_y^{n+1/2}(i+1/2, j, k+1/2) = H_y^{n-1/2}(i+1/2, j, k+1/2) + \frac{\Delta t}{\mu \delta} [E_z^n(i+1, j, k+1/2) - E_z^n(i, j, k+1/2) + E_x^n(i+1/2, j, k) - E_x^n(i+1/2, j, k+1)] \]

where the terms \( A \) and \( B \) are given by;

\[ A_{i,j,k} = 1 - \frac{\sigma(i, j, k)}{\varepsilon(i, j, k)} \] (A.7)

\[ B_{i,j,k} = 1 - \frac{\Delta t}{\varepsilon(i, j, k) \delta} \] (A.8)

In all of the finite difference equations the components of \( \vec{E} \) and \( \vec{H} \) are located within a single unit cell in the three-dimensional lattice.

**B. Courant Stability Condition:**

The one dimensional scalar wave equation is given by;

\[ \frac{\partial^2 u}{\partial t^2} = c^2 \frac{\partial^2 u}{\partial x^2} \] (B.1)

Dispersion relation of the one dimensional wave equation is given by;

\[ k = \pm \omega / c \]

Phase velocity, \( v_p = \omega / k = \pm c \)

Group velocity, \( v_g = \frac{\partial \omega}{\partial k} = \pm c \)
Finite Differences:

By considering Taylor’s series expansion of \( u(x, t_n) \) about the space point \( x_i \), to \( x_i + \Delta x \) and keeping \( t_n \) fixed, by rearranging we get:

\[
\frac{\partial^2 u}{\partial x^2} \bigg|_{x_i,m} = \frac{u''_{i+1} - 2u''_i + u''_{i-1}}{(\Delta x)^2} + O[(\Delta x)^2]
\] (B.2)

This is the second order accurate central difference approximation to the second partial space derivative of \( u \).

Similarly,

\[
\frac{\partial^2 u}{\partial t^2} \bigg|_{x_i,m} = \frac{u''^n_{i+1} - 2u''^n_i + u''^n_{i-1}}{(\Delta t)^2} + O[(\Delta t)^2]
\] (B.3)

Substituting B.2 and B.3 in B.1, neglecting Taylor’s series remainder terms and solving for the latest value of \( u \) at the grid point \( i \) we get:

\[
u^{n+1}_i \equiv (c\Delta t^2)\left[\frac{u''_{i-1}}{(\Delta x)^2} - 2u''_i + u''_{i+1}ight] + 2u''_i - u''_{i-1}
\] (B.4)

In the above expression it is clear that all RHS terms are obtained during previous time steps which are stored in the computer memory. Hence the next time step \( u^{n+2}_i \) can be found and thereby the next time step and so on and so forth.

Put \( c \Delta t = \Delta x \) in (B.4)

\[
u^{n+1}_i = u''_{i+1} + u''_{i-1} - u''_{i-1}
\] (B.5)

Here, the LHS terms are exactly equal to the RHS terms and this is called the magic time step. The solution is exact in spite of the Taylor’s series approximation. Another
interesting point is that all the LHS terms are known for a certain time point and hence the value for the future time point can be found. Proceeding in this manner gives the method to resolve the entire structure numerically.

**Numerical Dispersion relation:**

Let \( \tilde{k} = \tilde{k}_{\text{real}} + j \tilde{k}_{\text{imag}} \)

Hence, \( u^n_i = \exp(\tilde{k}_{\text{imag}} i \Delta x).\exp(j(\omega_n \Delta t - \tilde{k}_{\text{real}} i \Delta x)) \)

Substituting \( u^n_i \) in B.4, factoring out \( \exp[j(\omega_n \Delta t - ki\Delta x)] \) and using Euler’s identity to complex exponentials we get;

\[
\tilde{k} = \frac{1}{\Delta x} Cos^{-1} \{1 + \left(\frac{\Delta x}{c\Delta t}\right)^2 [Cos(\omega \Delta t) - 1]\} \quad (B.6)
\]

**Dispersive Wave propagation:**

The phase velocity of a sinusoidal numerical wave within the grid is determined by the grid’s sampling resolution.

Considering \( c \Delta t = \Delta x / 2 \) and assuming \( \Delta x = \lambda_0 / 10 \), B.6 becomes;

\[
\tilde{k} = \frac{1}{\Delta x} Cos^{-1} \{1 + 4[Cos(\frac{k\Delta x}{2}) - 1]\} = \frac{0.63642}{\Delta x}
\]

Hence, \( v_p = \frac{2\pi f}{0.63642 / \Delta x} = 0.9873c \)

For our assumptions it is seen that a numerical phase error of 45.72° results. Hence this indicates the dependence of \( v_p \) on the sampling resolution of the grid.
Numerical Stability:

To ensure numerical stability $\Delta t$ should be bounded for a given bounded input. We follow the approach of complex-frequency analysis to study the numerical stability. Let $\omega$ be the complex value

$$\omega = \omega_{\text{real}} + j\omega_{\text{imag}}$$

Hence, $u_i^n = \exp(-\omega_{\text{imag}} n\Delta t) \cdot \exp(j(\omega_{\text{real}} n\Delta t - k_i \Delta x))$

The numerical dispersion relation is given by;

$$\cos(\omega \Delta t) = \left(\frac{c\Delta t}{\Delta x}\right)^2 [\cos(k\Delta x) - 1] + 1$$

Defining the Courant number, $S = c\Delta t / \Delta x$

$$\omega = \frac{1}{\Delta t} \cos^{-1}(\xi)$$

$$\xi = S^2 [\cos(k\Delta x) - 1] + 1$$

(B.7)

(i) For $-1 \leq \xi < 1$, $\sin^{-1}(\xi)$ is real valued and hence has a constant amplitude with time.

(ii) For $1 - 2S^2 \leq \xi < -1$, $\xi_{\text{lowerbound}}$ occurs for $\cos(k\Delta x) = -1$. This is given by;

$$\xi_{\text{lowerbound}} = 1 - 2S^2 \} \text{ for } k\Delta x = \pi$$

(B.8)
The complex-valued arc-sine function is;

\[ \sin^{-1}(\xi) = -j \ln(j \xi + \sqrt{1 - \xi^2}) \]

Substituting this in B.7, factoring out \( j = \exp(j\pi/2) \) and taking natural logarithm;

\[ \omega = \frac{1}{\Delta t} \{ \pi + j \ln(-\xi - \sqrt{\xi^2 - 1}) \} \]

\[ \omega_{real} = \frac{\pi}{\Delta t} \]

\[ \omega_{imag} = \frac{1}{\Delta t} \ln(-\xi - \sqrt{\xi^2 - 1}) \]

\[ uni = (\frac{1}{-\xi - \sqrt{\xi^2 - 1}})^{\text{**}n} \exp(j[(\pi / \Delta t)(n\Delta t) - k\Delta x]) \]

\[ **n \Rightarrow n^{th} \text{ power} \]

The multiplicative factor is given by;

\[ q_{growth} = \frac{1}{-\xi - \sqrt{\xi^2 - 1}} = -\xi - \sqrt{\xi^2 - 1} \]

Substituting (7) in the expression above gives;

\[ q_{growth} = (S + \sqrt{S^2 - 1})^2 \]

This clearly indicates the growth factor increase for even very small increase in the stability factor beyond 1 resulting in exponential growth and rapid oscillations of the wave in the grid. This exponentially increasing wave has a temporal frequency;

\[ f_o = \frac{\omega_{real}}{2\pi} = \frac{1}{2\Delta t} \]

The velocity of this wave is given by;
\[ v_p = \frac{w_{\text{real}}}{k} = \Delta x / \Delta t = c / S \]

Therefore for numerical stability for a one dimensional scalar wave equation the upperbound on \( \Delta t \) relative to \( \Delta x \) is given by:

\[ S = \frac{c \Delta t}{\Delta x} \leq 1 \]

\[ S_{\text{stability limit}} = 1 \]

For the two dimensional wave equation:

\[ \Delta t \leq \frac{1}{c \sqrt{\frac{1}{(\Delta x)^2} + \frac{1}{(\Delta y)^2}}} \]

for \( \Delta x = \Delta y = \Delta \)

\[ \Rightarrow \frac{1}{c \sqrt{\frac{1}{(\Delta)^2} + \frac{1}{(\Delta)^2}}} = \frac{1}{c \sqrt{\frac{2}{(\Delta)^2}}} = \frac{\Delta}{c \sqrt{2}} \]

Hence, \( \Delta t = \frac{\Delta}{c \sqrt{2}} \)

\[ \frac{c \Delta t}{\Delta} = \frac{1}{\sqrt{2}} \]

\[ S_{\text{stability limit}} = \frac{1}{\sqrt{2}} \]
Similarly for three dimensions;

\[ S_{\text{stability limit}} = \frac{1}{\sqrt{3}} \]

**C. Perfectly Matched Layer:**

Maxwell’s curl equations are modified and expressed in their time-dependent form as;

\[ \varepsilon_2 \frac{\partial E_x}{\partial t} + \sigma_y E_x = \frac{\partial H_z}{\partial y} \]

\[ \varepsilon_2 \frac{\partial E_y}{\partial t} + \sigma_x E_y = -\frac{\partial H_z}{\partial x} \]

\[ \mu_2 \frac{\partial H_{zx}}{\partial t} + \sigma_x^* H_{zx} = -\frac{\partial E_y}{\partial x} \]

\[ \mu_2 \frac{\partial H_{zy}}{\partial t} + \sigma_y^* H_{zy} = \frac{\partial E_x}{\partial y} \]

The \( H_z \) component is split into two additive subcomponents;

\[ H_z = H_{zx} + H_{zy} \]

\( \sigma_x \) and \( \sigma_y \) are the electric conductivities and \( \sigma_x^* \) and \( \sigma_y^* \) are the magnetic losses.

By letting the electric conductivities and magnetic losses equal to zero we see that the above equations reduce to the Maxwell’s equations. If the electric conductivities equal to \( \sigma \) and the magnetic losses are zero then it is an electrically conductive medium. If we assume that the electric permittivities and the magnetic permeabilities are equal to one
another, the conductivities are equal to \( \sigma \), the magnetic losses are equal to \( \sigma^* \) and the following condition is satisfied;

\[
\frac{\sigma^*}{\mu_1} = \frac{\sigma}{\varepsilon_1}
\]  (C.1)

Then the above equations describe an absorbing medium for normally incident plan waves.

Also, if \( \sigma_y^* = \sigma_y = 0 \) the medium can absorb a plane wave propagating along \( x \), but not along \( y \). The converse is true if \( \sigma_x^* = \sigma_x = 0 \). These properties are characterized by the parameters \((\sigma_x^*, \sigma_x, 0, 0)\) and \((0, 0, \sigma_x^*, \sigma_x)\). Thus if these pair parameters satisfy condition (C.1) then they can act as an absorbing medium of electromagnetic waves.

By expressing the above four time dependent wave equations in their time harmonic form and obtaining the plane wave solution within such a medium we can get transmitted fields within the PML medium as follows;

\[
\dot{H}_z = \text{Ho} \exp(-j\beta_1 x - j\beta_1 y) \exp(-\sigma_x x \eta_1 \cos \theta)
\]

\[
\left\{
\begin{align*}
\dot{E}_x &= -\text{Ho} \eta_1 \sin \theta \exp(-j\beta_1 x - j\beta_1 y) \exp(-\sigma_x x \eta_1 \cos \theta) \\
\dot{E}_y &= \text{Ho} \eta_1 \cos \theta \exp(-j\beta_1 x - j\beta_1 y) \exp(-\sigma_x x \eta_1 \cos \theta)
\end{align*}
\right.
\]

Within the PML the transmitted wave travels with the same speed as the incident wave and also undergoes exponential decay along the \( x \) axis. The \( \sigma_x \eta_1 \cos \theta \) is the attenuation factor which is independent of the frequency. Also, this applies to all incident angles irrespective of their angle of incidence. Similar medium can be drawn up for the attenuation along the \( y \) axis. By combining the two we can formulate a 2D PML as shown in figure C.1.
Figure C.1 Schematic of the placement of PML boundaries for a 2D system [30].

This depicts a PML having a Perfect Electric Conductor (PEC) at its outer boundary. The losses on each side of the grid are matched according to the relation (C.1) and at the corners where there is an overlap of two PMLs the losses are set equal to those of the adjacent PMLs.

This kind of a setup gives an excellent absorbing and reflectionless outer boundary layer that can be used to simulate an infinite structure. Though the numerical phase velocity dispersion may hinder the accuracy of the PML, reflections are very small compared to the measured phase velocity variations. The step discontinuity in $\sigma^*_x$ and $\sigma_x$ gives rise to numerical artifacts due to the finite spatial sampling leading to reflections. This can be reduced by grading of the loss parameters either by polynomial or geometric grading methods.
D. Setting up FDTD simulations on WestGrid:

- Open simulation file “filename.fsp”
- Note the memory requirements from the “Check memory requirement” option from the “Simulate” menu.
- Select “Parallel FDTD Options” from the “Simulate” menu in the layout editor.
- Select a “Shell/batch file type” of “qsub (WestGrid)” with file extension “.sh (Linux)”
- Check the box to “Create a parallel shell/batch file when saving fsp file”. This will automatically create a script file “filename.sh” when simulation is saved.
- Set the executable directory to “/global/software/lumerical-3.1/bin/”.
- Set the “current directory” to the location of the file “filename.fsp” in the WestGrid directory.
- Set “No. of processors” to “1” and “No. of nodes” to “16”. This will give a better chance of getting off the queue and beginning execution of the simulation sooner, since there is lesser memory requested per processor.
- Set the memory per processor in accordance with the memory requirements of the particular simulation. For faster completion of the simulation, keep memory requirements below 2 Gb by exploiting symmetry of the problem.
- Open “filename.sh” and add the following commands;

```
#PBS –m yourmail@address.com
#PBS –M bea
```

These commands alert the user by sending an e-mail when execution of the simulation begins and at the end.
E. Script for generating stream file of double-hole array:

character*30 ifile
dimension isheet(4096,4096)

common ixpmax, iypmax, ix2pmax, iy2pmax

xpmax = sheet x pixel dimension
ypmax = sheet y pixel dimension
idt = threshold resolution for beam

ixpmax = 4096
iypmax = 4096

ix2pmax = ixpmax/2
iy2pmax = iypmax/2

nanometers to pixel conversion factor
fna2pix = 7.14

Parameter Input Section

Number of rings
inr = 15
Spacing between rings in nanometers
rs = 710.
Width of rings in nanometers
rw = 100.
Pix Resolution for skipping
idt = 2
Burn time
itime = 100

Diameter of the inner solid circles in nanometers

irdia = 200

Distance between the inner solid circles in nanometers

irdis = 100

WRITE(*,*),
WRITE(*,*)'READING PARAMETER FILE'
WRITE(*,*),

open(20, file='input.par')

read(20,'(a30)')iﬁle
read(20,*),irdia
read(20,*),irdis
read(20,*),idt
read(20,*),iptime
read(20,*),ivspac
read(20,*),ihspac

open(10, file='iﬁle//.log.txt')

write(10,'(a30)')iﬁle

write(10,*),'Diameter of the inner solid circles in
&nanometers = ', irdia
write(10,*),'Distance between the inner solid circles in
&nanometers = ', irdis
write(10,*),'Pix Resolution for skipping = ', idt
write(10,*),'Inner Pattern Burn time = ', iptime
write(10,*),'Vertical spacing = ', ivspac
write(10,*),'Horizontal spacing = ', ihspac

write(10,*),

----------------------------------------------------------------------------------

convert from nanometers to pixels

----------------------------------------------------------------------------------
c Diameter of the inner solid circles in pix
irdiap = irdia/fna2pix

c Distance between the inner solid circles in pix
irdisp = irdis/fna2pix

c Vertical spacing in pix
ivspacp = ivspac/fna2pix

c Horizontal spacing in pix
ihspacp = ihspac/fna2pix

c-----------------------------------------------
c Initialise the isheet array to 0
WRITE(*,*)''
write(*,*)'INITIALISING SHEET ARRAY''
WRITE(*,*)''
do i = 1, ixpmx
   do j = 1, iymax
      isheet(i,j)=0
   enddo
enddo

c-----------------------------------------------

c Inner solid circles
WRITE(*,*)''
write(*,*)'DRAWING INNER SOLID CIRCLES''
WRITE(*,*)''

c Compute position of the center of the circles
ixir = 1
ixor = irdiap/2

ixmin = ixor+10 + irdiap/2
ixmax = ixpmx - ixor - 10 - irdiap/2 - irdisp
iymin = ixor + 10 + irdiap/2
iymax = iypmax - ixor - 10 - irdiap/2

do ix = ixmin, ixmax, ihspacp

do iy = iymin, iymax, ivspacp

c  write(*,*) ix, iy

c  left solidring

  ixp = ix

  call rings(ixir, ixor, isheet, idt, ixp, iy)

c  right solidring

  ixp = ix + irdisp
  call rings(ixir, ixor, isheet, idt, ixp, iy)

  enddo

  enddo

 -------------------------------------------------------------

  c  Data output to file

  -------------------------------------------------------------

  WRITE(*,*)''
  write(*,*)'COUNTING NUMBER OF DATA POINTS'
  WRITE(*,*)''

c  count the number of Isheet valid pixel nodes

  irec = 0
  irec1 = 0
  irec2 = 0
  do i = 1, iypmax
    do j = 1, ixmax
      if(isheet(i,j).eq.1.or.isheet(i,j).eq.2 ) irec=irec+1
      if(isheet(i,j).eq.1) irec1=irec1+1
      if(isheet(i,j).eq.2) irec2=irec2+1
    enddo
enddo

write(10,'(i)'),
write(10,'(i)') Total Number of Records = ', irec
write(10,'(i)') Total Number of Records (Outer) = ', irec1
write(10,'(i)') Total Number of Records (Inner) = ', irec2

Write the headers for the stream data file

WRITE(*,*)''
write(*,*)'WRITING OUTPUT FILE'
WRITE(*,*)''

open(11,file=ifile)
write(11,*),s'
write(11,*),l'
write(11,'(i7)')irec

Write the Isheet valid pixel nodes
iwrite=0
 do i = 1, ixpmax
   do j = 1, iypmax

     if(isheet(i,j).eq.1) then
       write(11,'(315)')iptime, i, j
     iwrite=iwrite+1
   endif

   enddo
 enddo

write(10,'(i)') Total Number of Records written = ', iwrite
200 stop
end

c+++++++++++++++++++++++++++++++++++++++++++++++++++++++++++

subroutine rings(ixir, ixor, isheet, idt, ix, iy)
DOUBLE PRECISION  ast,xpirad
dimension isheet(4096,4096)

xpirad=(22./7.)/180.
c common ixpmax, iypmax, ix2pmax, iy2pmax

c xpmx = sheet x pixel dimension
c ypmx = sheet y pixel dimension

ixpmax = 4096
iypmax = 4096

c compute the extreme points (iexlx, iexrx) along x-axis for the ring

do iradius = ixir, ixor, idt

aradius = float(iradius)
WRITE(10,*)''
write(10,*)radius = ',', aradius

do j=1,21600
do j=1,14400
do j=1,7200

c compute theta

c ast = float(j)/60.
c ast = float(j)/40.
c ast = float(j)/20.

radtheta = xpirad * ast
xpos = ix + aradius * cos(radtheta)
ypos = iy + aradius * sin(radtheta)

c write(*,*)xpos, ypos
ixpos = xpos
iypos = ypos

isheet(ixpos, iypos)=1

enddo

enddo

return

END
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**Author**  Kiran Kumar

**Signature**  

**Dated:** 23rd Aug '06

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