Investigation into interferometric sub-wavelength periodic feature fabrication and their applications

Sidharthan Raghuraman

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**NANYANG
TECHNOLOGICAL
UNIVERSITY**

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2012

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Abstract

Recently, the interest in periodic structures has been increasing rapidly due to its potential application in many fields of nanotechnology and biotechnology. These application areas include photonic crystals, antireflective optical elements, thin film solar cells, biosensors and magneto-electronic devices etc. Fabrication of such periodic structures requires very precise methodologies such as advanced lithographic techniques so as to achieve uniform, large area patterns with high resolution and periodicity that scale down well beyond the employed wavelength range. In this context, this thesis investigate novel interference lithographic (IL) concepts/methodologies so as to fabricate 1D, 2D and 3D uniform large area sub-wavelength periodic features using simple experimental set ups.

One such methodology involves employing multifaceted prism based zero path interferometer for large area micron and sub-micron patterning. Employing different prisms like biprisms, tripism and pyramidal prisms, periodic grating features, hexagonal lattice features and square lattice features were fabricated using single exposure and multiple exposure approaches. Uniform features with pitch sizes ranging from $3.75\text{ }\mu\text{m}$ to 410 nm were fabricated using various prisms. Prism based solid immersion lithography was later investigated for patterning features with improved resolution and pitch size that scales down beyond $\lambda/2$ value. Two configurations for prism based solid immersion lithography were

employed. The first one uses a specialty custom fabricated grating to split the incoming beam, mirrors to guide them and a high index solid immersion prism, beneath which the beams are made to interfere. High resolution features with pitch size around 110 nm and sub-60 nm resolution were fabricated using a 364 nm laser source. The second configuration uses multiple converging lenses to direct the beam on to the high index prism. Here, the use of a mirror assembly is avoided and it is particularly advantageous when the number of beams involved is increased to more than two. Detailed theoretical investigation was performed and square lattice features with pitch sizes around 210 nm and 240 nm were fabricated using this double lens (biconvex)-prism based configuration.

It is to be mentioned that IL has the potential to fabricate complex patterns using multiple beams. However, polarization states of individual beams play a major role and help in broadening the range of structures that could be fabricated by interference lithography. In this perspective, a detailed theoretical analysis of two-beam, four-beam and five-beam interference for various polarization combinations involving TE, TM and circularly polarized beams is carried out and analysed in this thesis. Complex 3D periodic structures such as body centred tetragonal, woodpile, β -tin type and diamond cubic like structures were realized using five-beam interference employing a circularly polarized central beam with a phase shift of zero.

Sub-wavelength periodic features are found to be very useful in various areas of nano-optics and photonics. This thesis also explores the improvement of

propagation length of surface plasmon polariton in periodic mask based near field lithography by employing a dye based gain medium. Numerical investigations performed on the proposed configuration for surface plasmon based lithography show that the transmission through the sub-50 nm featured metal mask can be enhanced by a factor of 14.5 with the assistance of dye medium.

Furthermore, improving light absorption in thin film solar cell by employing periodic back structures and novel plasmonic concepts were also investigated in this thesis. Different silicon based thin film solar cell configurations employing periodic structures that take advantage of different phenomena such as SPP, gap mode effect, scattering etc. were introduced and studied. It was inferred that periodic back structures and front structures improves light trapping and hence absorption efficiency considerably. For example, one such proposed configuration, employing Ag spheres and Al back grating layer was found to improve absorption in silicon by a factor of 9% over planar metal layer-Ag sphere configuration.

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List of Symbols

λ	Wavelength
p	Pitch size, Period
n	Refractive index
$\alpha, \beta, \theta, \phi, \gamma$	Angle
ε	Dielectric constant
k	Wave vector
f	Focal length
u, v, d, h	Distance
$I(r)$	Intensity
E_i	Amplitude of electric field vector
ω	Angular frequency
t	Time
φ_i	Phase constant
$\hat{e}_i$	Polarization vector
$\hat{e}_i^*$	Conjugate of vector $\hat{e}_i$
$\hat{e}_i \cdot \hat{e}_j^*$	Dot product of vectors $\hat{e}_i$ and $\hat{e}_j^*$
$ z $	Magnitude or modulus of z
Σ	Summation
σ_e	Emission cross section of dye
τ	Decay rate

List of Abbreviations

1D	One dimensional
2D	Two dimensional
3D	Three dimensional
NA	Numerical Aperture
TE	Transverse Electric
TM	Transverse Magnetic
LC	Left Circular
RC	Right circular
FCC	Face Centred Cubic
FCT	Face Centred Tetragonal
BCT	Body centred Tetragonal
SP	Surface Plasmon
SPP	Surface Plasmon Polariton
LSP	Localized Surface Plasmon
SEM	Scanning Electron Microscopy
AFM	Atomic Force Microscopy
STM	Scanning Tunnelling Microscopy
LSP	Localized Surface Plasmon
LIL	Laser Interference Lithography
SIL	Solid Immersion Lithography
EBL	Electron Beam Lithography
FIB	Focused Ion Beam
PMMA	Poly Methyl methacrylate
Al	Aluminium
Ag	Silver
UV	Ultra Violet
DUV	Deep Ultra Violet
EUV	Extreme Ultra Violet

Chapter 1: Introduction

The chapter begins with the background and motivation for undertaking this research work. It discusses the scope and objectives of this thesis followed by a diagrammatic representation of the research roadmap planned for meeting the desired objectives of the thesis. The chapter concludes with the organization of the thesis.

1.1 Background and Motivation

Periodic structures are of increasing interest in many fields (e.g., nanotechnology and biotechnology) and are expected to provide the basis for new devices, such as photonic crystals, antireflective optical elements, biosensors and magneto-electronic devices [1-8]. A number of techniques such as electron-beam lithography, focussed ion beam lithography, nanoimprint lithography, photolithography etc. can currently be used to fabricate such periodic structures [9-13]. Micro and sub-micrometre patterning employing such methodologies promise potential periodic photonic crystal fabrication as well. However, most of these techniques have major limitations in throughput, patterning area, time consumption, cost of fabrication etc. For example, e-beam lithography (EBL) and focussed ion beam lithography (FIB) are time consuming, particularly when large area patterns are desired due to the sequential nature of writing [14, 15]. It would be highly advantageous to write the entire desired area in one step. Nanoimprint lithography on the other hand has less flexibility because of the

mechanical nature of imprint technology. It requires the fabrication of a hard mold with high resolution and temperature stability and also has issues due to stress induced during imprinting and heating [15, 34]. X-ray lithography (XRL) is another technique which has been used for high resolution patterning [16, 17] but require high resolution mask fabrication. Moreover X ray masks are very sensitive to stress, distortion and radiation. Stress and distortion issues also arise during fabrication of such masks. In contrast, optical lithography is a parallel writing technique and operates over large areas at low cost [18, 19]. Traditional optical lithography uses a mask-based approach to produce patterns. Once the mask is fabricated, all of the information on the mask is transferred onto the wafer in the lithography step. But the production costs including that of masks, makes this approach viable only for large volume production. Fortunately, the fabrication of periodic sub-wavelength structures could be achieved by a much simpler laboratory-scale laser interference lithographic (LIL) technique which requires no masks.

LIL is a technique, where the standing wave patterns obtained by interference of coherent beams are recorded producing useful patterns over large areas and volumes with considerable pattern flexibility. Various setups have been used to generate uniform, sub-micrometre array structures over very large area for applications such as photonic crystals, bio sensing, high-density data storage [20, 21]. Maskless laser interference lithography employs several configurations, all of which can be divided into amplitude division or wavefront division approaches [22-25]. The former involves splitting the incoming coherent beam

into two or more components using either a beam splitter or grating arrangement and combining them on the surface of the wafer sample. These configurations are error prone owing to their complex setup and beam path sensitivity. The wavefront division method, on the other hand, involves splitting the incoming wave into wavefront's and superposing them to form periodic patterns. The paths of interfering waves are very small and so these can be termed as zero path interferometric techniques. Among different wavefront division techniques, the one employing a biprism is of particular interest, mainly because it further simplifies the experimental setup and has the ability to fabricate large area 2D periodic structures with sub-micron resolution. In this context, this thesis focuses on investigating the prism based zero path interferometric lithography.

As resolution requirement for most of the periodic features are increasing with respect to applications each day, it is also crucial to look into novel maskless configurations which offer better pitch size and resolution scaling below $\lambda/2$. From this perspective, this thesis also focuses on developing a methodology for periodic feature fabrication employing the immersion lithography concept. Polarization plays an important role in interference lithography. A broad range of intensity patterns can be produced by varying the polarization states of individual beams. In this context, this thesis also focuses on analysing the effect of polarization, in two-beam, four-beam and five-beam interference configurations, for producing 1D, 2D and 3D periodic structures respectively.

Recently, photolithographic techniques employing the concept of surface plasmon resonance has been widely studied to fabricate features with resolution scaling beyond the diffraction limit [26-29]. Surface plasmon based near field photolithography, using periodic metallic masks, has achieved much progress in the last decade [26, 30]. However, there exists a major limitation in the form of the short propagation length resulting from power dissipation in the metal. In this context, it is advantageous to investigate new approaches to surface plasmon lithography with the capability of compensating for the metal absorption losses. A dye based gain medium is investigated to compensate for these losses and to increase the propagation length of surface plasmon polariton, thus improving the depth of focus along with the resolution.

Photovoltaic's based on thin absorbing layer have invoked tremendous interest over the past decade, as a possible solution for future energy. Light trapping plays an important role in increasing the efficiencies of thin film solar cell by enhancing the optical absorption inside the thin active layer [31-33]. In silicon based solar cell, proper light trapping techniques can decrease in film thickness to less than 2 μm , without compromising the conversion efficiency. This helps in a reduction of material and hence a lowering of production cost. Light trapping by means of texturing front and back surface and by using various plasmonic concepts has been investigated in detail in recent times [34-37]. However, there is still a lot of scope for improvement in the efficiency level of thin film solar cells in order to meet the commercial requirements and so it is essential to look into novel concepts for light trapping and absorption

enhancement. In this thesis, the usage of periodic structures for light trapping, both at the front and back surface, along with the introduction of recent plasmonic concept of gap modes has also been investigated.

The specific objectives and scope of this thesis is outlined below.

1.2 Objectives

The main objective of the thesis is to investigate various interferometric lithographic concepts/methodologies for the fabrication of periodic structures, which provide resolution ranging from micron to sub-100 nm and to investigate their potential R&D applications. This can be subdivided as:

(a) Development of interference lithography techniques for the production of large area micron, sub-micron and nano-scale patterns which can be simply implemented.

(b) Investigating the effect of polarization in multiple-beam interference lithography and analysing the formation of 1D, 2D and 3D periodic structures.

(c) Investigation into near field lithography by employing periodic metallic structures and dye based resolution enhancement methodology.

(d) Investigation of the absorption enhancement in plasmonic thin film solar cells employing periodic structures.

1.3 Scope

In order to achieve the above objectives of the proposed research, a possible scope of study can be outlined as follows:

- Comprehensive literature review on various lithographic techniques for sub-wavelength periodic feature fabrication.
- Develop on interference lithography system for large area patterning based on multifacet prism; this will include theoretical formulation, simulation and experimental validation.
- Develop solid immersion interference lithography (SIL) system for the fabrication of high resolution features with period scaling beyond $\lambda/2$.
- Fabricate and characterize planar periodic structures using optical lithographic schemes.
- Theoretically investigate multiple-beam interference and formulate a methodology to visualize various 1D, 2D and 3D structures formed by the same.
- Develop a theory of a novel lithographic approach for resolution enhancement in dye based nanolithography; this will be supported by simulation.
- Develop novel approaches for the absorption enhancement in thin film solar cell and investigation of the same by Finite-Difference-Time-Domain (FDTD) simulations.

A block diagram, describing the research roadmap of the proposed investigation is given in Figure 1.1.

Figure 1.1 Research road map: Block diagram representation.

1.4 Organization of thesis

The entire thesis is divided into seven chapters. Each chapter starts with a short note reflecting the main contents of that chapter.

A comprehensive literature review of existing fabrication techniques for sub-wavelength periodic feature fabrication is given in chapter 2, underlying the importance of optical interferometric technique for the same. A review of the possible application of sub-wavelength periodic features in mask based surface plasmon lithography and plasmonic light trapping in thin film solar cell, is also given in a latter section of the chapter.

In chapter 3, investigations are made on wavefront division interferometer employing multi-facet prisms along with UV and DUV laser sources. Uniform 1D and 2D features with micron and sub-micron periodicity employing this simple technique has been fabricated and reported.

In chapter 4, two configurations for prism based solid immersion lithography are investigated. The first configuration uses a grating to split the beam, mirrors to guide it and a high index prism to make it interfere. And the second configuration uses an inverted prism to split the beam, and multiple converging lenses to direct the beam on to the high index prism. Uniform high resolution grating lines and square lattice features were shown to be patterned using these solid immersion techniques.

Chapter 5 details the study conducted with respect to the effect of polarization on the patterns written by two-beam, four-beam and five-beam interference. Variation in the contrast of the patterns in two-beam interference, formation of 2D square lattice patterns using four-beam interference and the formation of 3D periodic photonic crystal structures have been studied in detail using different polarization combinations involving TE, TM, LC and RC polarized light beams.

In chapter 6, the main focus was laid on investigating two different applications of periodic structures in the area of plasmonics; namely (i) dye based enhanced transmission through 2D metallic grating and (ii) light trapping in silicon based solar cell. Initially, a configuration for surface plasmon based lithography, employing a periodic metal array as mask and dye as a gain medium was proposed and numerically analysed for enhanced surface plasmon based transmission. Later, absorption enhancement using periodic structures in thin film silicon based solar cell was studied numerically for various configurations.

Chapter 7, final chapter of the thesis, gives the conclusion of the thesis along with guidelines for future work. The important contributions made in this thesis are explained in this chapter. Suggestions for future work are also briefly mentioned in this chapter.

Chapter 2: Literature Review

Over the last decade, the steady progress in the fabrication of photonic nanostructures has led to a variety of different one, two, and three-dimensional dielectric and/or metallic periodic structures. They exhibit qualitatively novel and fascinating linear-optical, nonlinear-optical and quantum-optical properties, providing an unprecedented control of light propagation and light-matter interaction. Such periodic structures have found tremendous applications in many areas including plasmonics and light trapping inside thin film solar cells. This chapter focuses on reviewing the work done in this area.

A large variety of techniques have been successfully employed for the fabrication of 2-D periodic features. It's important to identify the major constraints on these techniques. A large number of photonic applications employ periodic features and it is essential to investigate suitable simple fabrication techniques for fabrication of features with periodicity ranging from 100 nm to 1000nm. One such important application area of periodic features is the photonic crystal and it requires that periodicity be of the order of wavelength at which it is desired to perform. i.e., if the working wavelength falls in the communication (1.3-1.5 μ m) or visible (400-700nm) window, submicron periodicity is desired. This necessitates the need for a patterning technique which gives good quality, uniform submicron features over a large area. In this

chapter, various lithographic techniques for feature patterning which are commonly used will be discussed in detail.

2.1 Overview of sub-wavelength lithographic techniques

Moore's Law has been the most important benchmark for microelectronics development over the past four decades. It states that the number of transistor devices on a chip will double, approximately every two years [38]. The semiconductor industry has maintained this pace for more than 40 years and lithography along with other technological advancements has played a major part in keeping pace with this empirical rule. Roughly half of the density improvements have been derived from improvements in lithography. With the cost to fabricate a wafer remaining roughly constant, independent of size or content, this has resulted in the reduction in cost over this period. The chip-level integration of devices has been possible through improvement in lithographic resolution. Developing/improving lithographic techniques for mass production to go deeper into sub-micron and nanometre scale at a feasible cost has been and continues to be a challenge to researchers. Several types of lithographic techniques have been developed in order to achieve micron, sub-micron and nanometre scale patterns over the past several decades. All of these techniques have their own advantages and disadvantages and some of these are discussed in the following section.

2.1.1 Electron-beam lithography and focused ion beam lithography

Electron Beam Lithography is a lithographic process which uses a focused beam of accelerated electrons to form the circuit patterns on a wafer coated with electron beam sensitive material. Electron beam lithography, which uses a very small diameter focused electron beam, doesn't need masks to pattern the surface. Here, electron beam with spot size in the range of few nanometres, controlled by optics, scans the surface of the resist in a sequential order as determined by the input design. Resolution is not limited by diffraction limit; rather it depends on the diameter of the electron beam spot and scattering in the resist and can go as small as 10 nm [14, 39-42]. Electron-beam lithography can produce structures at smaller scales with almost complete pattern flexibility and therefore it is an attractive area of nanoscience research [43]. Nonetheless, e-beam lithography is not a viable approach as a result of its serial point-by-point writing nature which is time consuming, sometimes times taking few days to complete, especially when large patterning area is desired [44, 45]. Moreover, the resolution of an electron lithography system may be constrained by other factors, such as electron scattering in the resist and by various aberrations in its electron optics. These factors make it a complex, time-consuming and expensive process which restricts its use for mass fabrication.

Focused ion beam lithography on the other hand is very similar to e-beam lithography in that it uses an accelerated ion beam (mostly gallium ion) instead of e-beam to write the patterns [11, 15, 46]. Gallium ion is used to directly

punch metallic film on to the substrate. This is possible because of the heavy mass of ions and the generated patterns have very high resolution owing to shorter wavelengths of ion beam at high voltages. But because of its serial writing nature, it is time consuming and so the throughput is less, which means that it is not a good option for mass production. However, due to the technological advancement in e-beam and focused ion lithography over recent years and due to the fact that it provides very high resolution with high degree of accuracy, it is used for mask fabrication.

2.1.2 Nanoimprint lithography

Imprint lithography is a non-projection lithographic technology which uses a hard mold to imprint a polymeric film such as poly methyl methacrylate (PMMA), to generate nanoscale features, the process schematic of which is shown in Figure 2.1. Imprint lithography is a contact-patterning method. Although it was first introduced as hot embossing technique, later it was modified in 1996 by incorporating UV based curing (UV nanoimprint lithography) [9, 47, 48]. In the hot embossing method as illustrated in Figure 2.1(a), once the contact between hard mold and substrate is achieved, the thermoplastic polymer is heated above its glass transition temperature. This enables polymer flow and fills the structures of the mold. Later the temperature is lowered and the replicated structures are allowed to solidify, after which the mold is removed. Lateral resolutions of sub-10 nm are reported to be achieved by

this process [49]. However, one major issue with this technique is the stress induced during heating and cooling cycles.

In UV nanoimprint lithography, which is schematically shown in Figure 2.1(b), a UV curable monomer layer is employed. After imprinting the hard mold on this monomer, it is subjected to UV radiation through the transparent side of the mold. It results in monomer crosslink, forming a rigid polymer. Compared to hot embossing method it reduces imprint pressures and avoid stress induced during the high temperature cycle. Since no high-resolution lenses are required, imprint patterning tools are considerably cost-effective compared to high-performance step-and-scan exposure tools. However, the throughput and flexibility is lower because of the mechanical nature of imprint technology. Fabrication of the hard mold with high resolution and temperature stability, stress induced during imprinting and heating, availability of proper UV curable monomers etc are some of the areas which have to be explored and improved upon before it becomes a serious candidate for next generation lithography. However, imprint lithography is used in applications which are defect tolerant, have loose requirements for overlay, and low levels of integration. One example of the application of imprint lithography is the generation of patterned media for magnetic storage.

Figure 2.1 Schematic illustrations of (a) hot embossing nanoimprint lithography and (b) UV-nanoimprint lithography.

2.1.3 X-ray lithography

X-ray lithography (XRL) is another technique used for high resolution patterning and has been under development since the early 1980s [16, 17, 47, 50-54]. It uses x-rays of short wavelengths (0.1nm-10nm) to write features on x-ray sensitive resists and so overcomes the diffraction limits of optical lithography. It also employs a mask based fabrication approach similar to that of photolithography, where x-ray masks are employed to selectively absorb and transmit x-rays on to the resist, as shown in Figure 2.2. Mostly, synchrotrons are used as sources of x rays and the production cost is high due to the cost of the synchrotron source. To reduce cost per wafer, the number of aligners installed on a single synchrotron has to be increased. Also, accurate adjustment of the gap

between mask and resist is required for X-ray proximity lithography. For higher resolutions, the gap must also shrink, such that a 15 μm gap is expected to be needed for 100 nm ground rules, with a gap perhaps <10 μm for smaller dimensions. Therefore, so mask protection mechanisms should be employed.

X-ray masks consists of a very thin membrane (thickness < 2 μm) of low-atomic-number material, on which the patterns are placed in the form of high-atomic-number material. Low-atomic-number material transmits a large percentage of x-rays and the high-atomic-number material usually absorbs or scatters the rays, thus generating a pattern contrast. Silicon carbide is typically used as the membrane material and gold, TaN, TaSiN_x, Ta₄B etc are employed as the absorbing material. Key issues in XRL arise due to this thin membrane. Deformation under stress, distortion and radiation damage due to over exposure of the mask are some of the major challenges in XRL [17]. Stress and distortion issues can arise during fabrication of such masks too, if e-beam lithography is employed. Although X-ray lithography has been projected as a potential successor to optical mask based technology, a number of issues are yet to be sorted out for this to happen.

Figure 2.2 X-ray proximity lithography with a collimated light source.

2.1.4 Scanning probe lithography

Scanning probe lithography is another interesting technique for high resolution patterning where a sharp atomic force microscope (AFM) probe is used to heat, write or transfer materials to the substrate surface. Among many techniques, Dip-Pen Nanolithography (DPN), is the most widely used method. In this technique, molecules are transferred to a surface via a water meniscus that forms between the tip and the surface by an AFM tip coated with a molecular ink [55-57]. Coating is performed by immersing the cantilever with AFM probe into a solution of interest or by evaporation. DPN lithography is schematically illustrated in Figure 2.3. Some of the key issues in this patterning technology are ink meniscus compatibility, initial surface quality (e.g., roughness and wettability), control over the speed of writing through change in the tip velocity, contact time, temperature and humidity conditions.

Figure 2.3 Schematic illustration of Dip-Pen nanolithographic process [57].

2.1.5 Optical Lithography

2.1.5.1 Mask Based Optical Lithography

Optical lithography is basically a photographic process where a light-sensitive polymer, called a photoresist, is exposed to visible or ultra-violet light and developed to form patterns on the substrate. In general, the ideal photoresist image will replicate the exact shape of the intended pattern in the plane of the substrate, with vertical walls through the thickness of the resist.

Traditional optical lithography uses a mask-based approach to produce patterns. Once the mask is fabricated, all of the information on the mask is transferred onto the wafer in the lithography step which is carried out as shown in Figure 2.4. The semiconductor surface is first cleaned using standard wafer cleaning method, which involves cleaning in piranha solution followed by rinsing in deionised water. The resist is then spin coated on to the cleaned substrate.

Resist can be either positive or negative tone. A positive tone resist is a type of photoresist in which the portion of the photoresist that is exposed to light becomes soluble to the photoresist developer. And in a negative tone resist, the portion that is exposed to light become insoluble to the photoresist developer and the unexposed portion of the photoresist is dissolved by the photoresist developer. Resists usually do not adhere properly to untreated surfaces of silicon. So, in order to ensure proper adhesion of the resist to the wafer surface, the wafer surfaces are treated prior to resist coating. Once the coating is done using proper recipe (resist volume, coating speed and time value) so as to obtain desired thickness, exposure is performed using a mask aligner system loaded with the desired mask. Exposure is followed by an optional post exposure step where the exposed wafer is heated to high temperatures (100–130 °C) to cause diffusion of the photoactive compound to smoothen out the standing wave ridges. This step is essential when a negative tone resist is employed to ensure polymer cross linking. After post baking step, the wafer is treated with developer to remove the portion exposed or unexposed, depending upon the resist employed. The developed wafer may then be subjected to etching, implantation, or metallisation and lift-off.

Figure 2.4 Lithographic processing steps.

Unlike e-beam and focussed ion beam lithography, optical lithography is a parallel writing technique and so is much less time consuming. And unlike the x-ray process, the mask preparation is much easier. The mask used in photolithography doesn't need to be as thin as in x-ray because of the use of UV light instead of x-ray which owing to its higher wavelength is not absorbed by most materials. Usually a quartz substrate and chrome metal are used as transparent and opaque medium respectively, for high resolution masks and the patterning is performed by other lithographic techniques. Because of the high throughput and the advancements made in developing lithographic systems and processes, it is widely used in the IC industry for patterning the circuits on silicon wafer. However, for high resolution feature fabrication in the range of sub-100 nm to $\leq 0.3 \mu\text{m}$, UV (i line source) mask based photolithography fails due to

the restriction imposed by the diffraction limit. To achieve this range in order to meet industrial standards, photolithographic systems employing deep ultraviolet (DUV) and extreme ultraviolet (EUV) sources are currently used commercially. Even though they promise better resolution, the system should be made of reflective optics because of the absorption at shorter wavelengths and new wafer processing steps have to be formulated, which increase the costs considerably.

2.1.5.2 Optical Interference Lithography

Fortunately, many applications such as those involving thin film solar cells, photonic crystals etc. require only a periodic or quasi-periodic pattern which could be achieved by a much simpler laboratory-scale technology, namely, interferometric lithography (IL). IL is a technique based on the interference of number of (most often two or four) coherent laser beams, which produces patterns over large areas and volumes, with considerable, but not total, pattern flexibility. The fundamental concepts of IL could be understood by considering the simple case of two-beam interference, which is depicted in Figure 2.5, where coherent laser beams are symmetrically incident from both sides. The period of the interference pattern is given by $\lambda/2\sin\theta$. Standing wave patterns exists throughout the overlap between the beams as long as this overlap distance is shorter than the longitudinal coherence length of the laser beams and the wafer can be placed anywhere inside this coherence volume.

Figure 2.5 Schematic representation of two-beam interference. (adapted from [58]).

IL is a conceptually simple process, which uses a small number of coherent optical beams which are incident from different directions on a photosensitive layer to produce an interference pattern whose intensity distribution is recorded in the photosensitive layer and is later transferred (developed) by thermal and chemical processes. The spatial-period of the features fabricated can be as low as half the wavelength of the interfering light, allowing for structures of the order of 100 nm from UV wavelengths [58]. In the literature, this concept has been variously referred to as holographic lithography, interference lithography, and IL. The beam division method in maskless laser interference photolithography can be divided into amplitude division and wavefront division. The former involves splitting the incoming coherent beam into two or more components using either a beam splitter or diffraction grating and later interfering them on the surface of the wafer sample, as shown in Figure 2.6. This so called conventional lithographic configuration, apart from having complex setup, are error prone, some of which includes polarization leakage leading to high periodic nonlinear error, beam path sensitivity etc [59]. The

wavefront division method, on the other hand, involves splitting the incoming wave into wavefront's and superimposing them to obtain periodic intensity variation at the substrate surface. A simple example is shown in Figure 2.7, where a Lloyd's mirror is employed for lithography [59-61]. However, in this setup the uniformity of the patterns fabricated depends on the quality of the mirrors. Dust particles on the mirror and any sharp edges give rise to scattered radiation which contributes noise, affecting the feature quality.

Figure 2.6 Conventional configuration for interference lithography [59].

Figure 2.7 Lloyd's mirror configuration for interference lithography [59].

The resolution of a lithography system is usually expressed in terms of its wavelength (λ), a constant depending on the lithographic process condition, and the numerical aperture (NA) as,

$$Resolution = \frac{\lambda K}{NA} \quad 2.1$$

In IC manufacturing employing mask based photolithography, typically, projection optics and wafer stage are placed in air or in vacuum, to reduce the light absorption by surrounding medium. Which means, the refractive index (n) is unity and $NA = \sin \theta$. The numerical aperture therefore cannot be greater than 1, and more practically, takes on values in the range from 0.5 to 0.8, with a higher number reflecting a less stringent process. The NA of optical lithography tools ranges from about 0.5 to 0.6. Immersion technology provides another simple way to increase numerical aperture: by making $n > 1$. This could be achieved by using liquid (all liquids have a refractive index greater than unity) between the projection optics and writing medium as shown in Figure 2.8 [62, 63]. Typically the index matching liquid used has a refractive index ranging from 1 to 2. It should be noted that the high index liquids should have sufficient transmission at the desired wavelength, which restricts the increase in index far greater than 2.

Now the same concept could be applied to interferometric lithography as well. All classical optical interference lithographic techniques are diffraction limited to writing features of size $\lambda/2$, where λ is the optical wavelength [64,

65]. In case of simple two-beam interference in air or vacuum, the period (p) of the patterns is given by,

$$p = \frac{\lambda}{2 \sin \theta} \quad 2.2$$

Where, λ is the wavelength of the laser beam and θ is the half angle of intersection of the two beams at the sample. Using an immersion concept, employing a liquid of index $n > 1$, the period can be written as,

$$p = \frac{\lambda}{2 n \sin \theta} \quad 2.3$$

Figure 2.8 Schematic diagram of mask based projection lithography system using immersion liquid.

But the feature size can be reduced further by using the concept of immersion lithography where the refractive index of the surrounding medium is changed to increase the numerical aperture to $NA = n \sin \theta$. For example, if θ is fixed at 45° , and purified water ($n=1.33$) is employed as liquid medium, NA increases from 0.707 to 1.01. And if a liquid of $n=1.8$ is employed, NA becomes

1.27. Various configurations have been investigated to date to fabricate features using immersion interference lithography [58, 60, 66, 67]. Most of the reported work explores the implementation of immersion concept using a deep ultraviolet (DUV) light source [60, 63, 66]. In one such system developed by IBM research, 193 nm light enters the vacuumed interferometric body from the top and is split into two beams by diffraction from a fused silica grating [68]. These beams overlap at the wafer surface after reflecting from two mirrors and passing through a custom fused- silica prism. Even though this configuration provides better resolution, it requires deep UV optics for its implementation, which is very expensive. Moreover, most configurations require larger number of optical components like beam splitters, mirrors etc.

Figure 2.9 Schematic of a interferometric system developed by IBM for 193 nm interference lithography [68].

Comparisons of various lithographic techniques discussed in the above sections are made and is shown in Table 2.1 .

Table 2.1 Comparison between various high resolution lithographic techniques.

Lithography Technique	E-Beam	X-ray	Nanoimprint	Dip Pen	Mask based Photolithography (i-line)	Laser Interference Lithography (i- line)
Resolution →	~15nm	~10nm	~10nm	~14nm	~100nm	Sub 100nm
Speed →	Slow	Very Fast	Fast	Slow	Very Fast	Very Fast
Writing nature →	Serial	Parallel	Parallel	Serial or Parallel	Parallel	Parallel
Material Flexibility →	No	No	Yes	Yes	No	No
Mask requirement →	No	Yes	Yes	No	Yes	No
Periodic feature patterning without mask →	Yes	No	No	Yes	No	Yes
3D patterning →	No	No	No	No	No	Yes

2.2 Periodic Structures: Applications in Nanophotonics

Nanophotonics is an appropriate application area for sub-wavelength structures fabricated using IL. Nanophotonics is that area of nanoscience which is defined as the science and engineering of light-matter interactions that take place on wavelength and sub-wavelength scales. At such small scales, the physical, chemical and the structural nature of the natural or artificial nanostructured matter controls the interactions [69]. Nanostructures can have unique, controllable, and tunable optical properties that arise from the fact that they are smaller than the wavelength of light used to observe them. The optical properties of nanostructures can be tailored for various applications, such as

compact photoelectric power sources, tunable light sources, detectors, filters, waveguides, high-speed optical switches, sensors, and biophotonic medical diagnostics and therapeutics. Both the properties of the nanostructures and their organization into large-scale materials are important in modifying the optical response to meet the demands of various applications. Often the requirement is for large areas of periodic or quasi-periodic structures. Some of the immediate application areas include 1-D, 2-D, and 3-D photonic crystals. One-dimensional periodic nanoscale structures, such as multilayer optical coatings and distributed Bragg reflectors, have long been used in optical design and engineering. Controlling the periodicity of structures at the nanoscale in multiple dimensions (two- and three-dimensional photonic crystals), enables a control of both the magnetic and the electrical response of materials for photonics crystal and metamaterial applications or to manipulate nanoscale structures for enhanced field concentration for plasmonic applications.

2.2.1 Periodic Structures for Surface Plasmon Lithography

Recent research suggests that it is possible to develop a new photolithographic technique beyond the diffraction limit using the concept of surface plasmon resonance [70-72]. Surface plasmons are collective electron oscillation propagating at the interface between a metal and a dielectric [73-75]. Surface plasmon resonances in metallic films are of interest for a variety of applications due to the large enhancement of the evanescent field at the metal/dielectric interface. Researchers observed extraordinary transmission

through perforated metal films and it showed that surface plasmons on the metal film can greatly enhance the electric field distribution and redistribute the electromagnetic field at the nanometre scale. Using such sub-wavelength metallic masks, the transmission of normally incident light is enhanced at the wavelengths that satisfy the surface plasmon resonance conditions given by [76],

$$\lambda_{SP(i,j)} = \frac{p}{\sqrt{i^2 + j^2}} \sqrt{\frac{\epsilon_m \cdot \epsilon_d}{\epsilon_m + \epsilon_d}} \quad 2.4$$

Where p is the periodicity of the array, i and j are integers, ϵ_d is the effective dielectric constant at the metal-dielectric interface, and ϵ_m is the dielectric constant of the metal. The surface plasmon (SP)-mediated transmission is several orders of magnitude higher than expected from Bethe's law for the transmission of light through sub-wavelength apertures [77]. The enhanced transmission is accompanied by strong field localization and has potential in several applications involving nano-optics, such as nanolithography and near-field optical microscopy probes. There are many reports of extraordinary transmission in sub-wavelength metal hole arrays metal due to surface plasmon resonance. One such attempt employing a sub-wavelength gold hole array for extraordinary transmission is depicted in Figure 2.10 [78]. A normalized transmission spectra of white light through three arrays of sub-wavelength holes of periodicities 618, 545, and 590 nm and diameter 200 nm and thickness 100 nm is shown in Figure 2.10(c). Peaks due to surface plasmon excitation are clearly visible in these plots. To generate sub-wavelength features from apertures much smaller than the exposing wavelength, the critical concerns are extremely low

transmission through the apertures, limitation of the exposing distance away from the mask, and poor contrast. They suffer from strong damping of surface Plasmon (SP) due to the scattering from the rough metallic surfaces, propagation loss due to the surrounding dielectrics and the inherent loss resulting from the complex dielectric constant of the metal. This energy dissipation limits the effective penetration depth, thereby creating an obvious obstacle in utilizing them in practical optical devices and circuits. Several attempts have been made to increase the transmission of the metal with perforated sub-wavelength features. One such attempt includes employment of structures with various shapes and sizes [78]. Among the various proposed structures, a coaxial aperture is found to have more light transmission than a single hole aperture due to the excitation of a cavity resonance mode [79]. About 90% extraordinary optical transmissions in the visible range through annular aperture metallic arrays have been obtained [80]. Bortchagovsky et al have studied transmission through small triangular apertures by confining the electric field at one edge with the help of an obliquely incident light beam [81]. Huizhong Xu et al. have studied the transmission of light through sub-wavelength apertures with dielectric filling [82]. They have reported an increase in transmission due to excitation of Fabry-Perot like resonant modes inside the cavity. Przybilla et al [83] obtained increased enhancement for array's larger than the SP propagation length. The achievement of high aspect ratio masks is a major goal for all these investigations. In this context, this thesis investigates dye

based plasmon enhancement in an embedded periodic metallic mask based near field lithographic configuration for high aspect ratio feature fabrication.

Figure 2.10 Scanning electron microscopy (SEM) images of an array of sub-wavelength holes in gold. The image in part B is an enlargement of the array presented in part A. (C) Normalized transmission spectra of white light through three arrays of sub-wavelength holes. All arrays were patterned in a 100-nm-thick gold film deposited on the glass slide, and the diameters of the holes were about 200 nm. The lattice parameters (periodicities) of arrays a-c were 618 nm, 545 nm, and 590 nm, respectively [78].

2.2.2 Periodic structures and plasmonics for enhanced light trapping

Photovoltaics have invoked tremendous interest among researchers over the past decade as a suitable solution for energy problems in the future. Various materials such as quantum dots, semiconducting nanostructures, and conjugated polymers for next generation photovoltaics have been extensively researched in

recent times. However, crystalline silicon remains the basic proven material for high-efficiency stable photovoltaics. However, the prices of currently available solar cells are high due to the higher costs of silicon material and processing techniques. Therefore, thin film solar cells are of great interest because of their smaller film thickness. Usually, polycrystalline or amorphous silicon are used as active materials in thin film solar cells because of their lower cost, nontoxicity, abundance and advanced processing technologies. But, in thin film solar cells, carrier diffusion length is very short and the absorbance of near bandgap light is small, resulting in lower absorption and conversion efficiency. Increasing the film thickness by few micrometers is not feasible as recombination rate would be very high. Hence, thin film solar cells need to be structured properly to trap more light in order to increase absorption. Various configurations to increase the optical path length inside the active silicon layer have been investigated and reported [84-89]. Most of the concepts rely on surface modification to realize scattering and enhanced reflection from back surface. Enhanced reflection by back surface texturing has also been widely reported in the literature [84-86]. One such reported geometry employing periodic Ag ridges of period 325 nm at the silicon back surface is shown in Figure 2.11(a) [84]. And the total absorption spectrum for models with rectangular and semicircular ridges at the back surface is compared with the reference cell and is shown in Figure 2.11(b). It clearly shows improvement in the IR and near IR range when periodic ridges were employed. Research has also been made documenting front surface texturing either randomly or with patterning using metal or dielectric structures [87-89].

One such configuration employing Ag strips of period 295 nm on front surface is shown in Figure 2.12(a) [89]. Enhanced current density due to plasmon resonance of the metal strips is clearly visible in Figure 2.12(b). Out of these, the ones employing periodic metallic back structures are of interest here. Most of such configurations, apart from taking advantage of scattering principle and enhancing reflection, also introduce surface plasmon modes that enhance absorption further. Some of the other major plasmonic light trapping geometries for thin film solar cells used at present is discussed in the following paragraph.

Figure 2.11 (a) Schematic of solar cell with Ag ridges at the back surface with period of 325 nm at the a-Si/Ag interface. (b) Incident solar spectrum (AM1.5) and total absorption spectra of the cell shown in (a) with rectangular and semicircular ridge back structure models is compared with the reference [84].

Figure 2.12 (a) Thin film solar cell using periodic array of metallic nanostructures; (b) Spectral short-circuit current density for the solar cell with and without Ag stripe array of periodicity 295 nm, showing the surface plasmon resonance [89].

At present, plasmonics concepts are widely used in thin film solar cells to achieve enhanced light localization and hence improved absorption [90-93]. Two structures utilising this concepts have already been discussed. Light can be concentrated and folded into a thin semiconductor layer, by proper engineering of metallic nanostructures that support the surface plasmons. In thin film solar cell, to allow complete light absorption and photocarrier current collection, the absorbing layer should be optically thick. Recently, three plasmonic concepts have been reported where physical thickness of absorbing layer is reduced while keeping their optical thickness constant [90]. The configurations are shown in Figure 2.13 . In the first method, freely propagating plane waves from the sun were coupled and trapped in the absorbing layer, by employing nanoparticles as sub-wavelength scattering elements. In the second reported work, localized

surface plasmons (LSP) are excited in nanoparticles. Here, the resonant plasmon excitation enhances the strong local field around the nanoparticle and thereby increase the absorption in the surrounding semiconductor medium. Light localization using surface plasmon polaritons (SPP) is the third reported concept. Light can be efficiently trapped and guided in the absorbing layer when SPPs are excited at the metal/semiconductor interface. In this context, in this thesis, various configurations utilizing periodic back and front structures, which takes advantage of scattering, reflection, guided modes and surface plasmon polariton principle are investigated.

Figure 2.13(a) Light trapping by scattering by metal nanoparticles at the top surface of the solar cell, (b) Light trapping by the excitation of localized surface plasmons in metal nanoparticles embedded inside the solar cell & (c) Light trapping by surface plasmon polariton(SPP) excitation at the interface of metal and semiconductor [90].

Apart from various principles discussed, gap modes which exists in metal-particle systems is also of interest. Gap modes are the localized electromagnetic normal modes that exist in the space between the metal nanoparticle and the surface. The excitation of gap modes strongly depends on the distance of the nanoparticle from the surface [94, 95]. When the particle-surface distance is sufficiently small, the system can support a series of gap modes and the electric

field becomes more and more localized at the gap between the particle and the surface. Furthermore, the modes responsible for the field distribution could be weakly coupled gap modes of single particle-film systems. The metal particle-surface system is expected to find a variety of potential applications in near field optics, although the roles played by gap modes have not yet been fully explored. One of the promising applications is in scanning tunnelling microscopy (STM) in which the tunnelling current is excited by gap modes [96]. In another reported work, excitation of gap modes in a metal particle-surface system for patterning periodic nanostructure were proposed and numerically demonstrated using the finite-difference time-domain method [97]. The Ag-Al layer based lithographic configuration is shown in Figure 2.14(a) and the enhancement in electric field using the same is shown in Figure 2.14(b). The simulation results show that this configuration provides a strong enhanced field to give shorter wavelengths of surface plasmons to fabricate sub-30 nm periodic structures.

Introduction of this gap mode principle into thin film solar cell will possibly enhance the absorption inside active silicon layer and provide better solar cell conversion efficiency. In this context, this thesis also investigates further absorption enhancement by employing the gap mode concept and periodic metallic structure.

Figure 2.14 (a) Ag-Al (Nanoparticle-planar layer) configuration for gap mode based lithography (b) Electric field enhancement using the metal-particle system [97].

2.3 Outcome of Literature Review

In semiconductor fabrication, lithography is the step which defines the pattern and its resolution. A large variety of techniques such as electron-beam lithography, X-ray lithography, focussed ion beam lithography, nanoimprint lithography, photolithography etc. could be employed to fabricate periodic photonic nanostructures, which in recent times have found tremendous applications in many areas such as plasmonics and thin film solar cells. However, all of these techniques are not best suited for periodic 2D and 3D feature patterning and has its own advantages and disadvantage. For example, e-beam lithography (EBL) and focussed ion beam lithography (FIB) are time consuming processes because of their sequential nature of writing, especially when large patterning area is desired. Nanoimprint lithography on the other hand has less flexibility because of the mechanical nature of imprint technology. It also

requires the fabrication of hard mold with high resolution and temperature stability using other lithographic techniques. Moreover, it also has issues due to stress induced during imprinting and heating. X-ray lithography (XRL) is another highly developed patterning technique but it also requires high resolution mask fabrication. X-ray lithography requires masks that are very thin and made of high and low atomic number materials, which makes mask production more complex. Moreover, x-ray masks are sensitive to stress, distortion and radiation. Compared to all the above mentioned techniques, traditional mask based optical lithography gives high throughput and is the lithographic technique which is widely used in industries. But it still requires masks and can be replaced by maskless laser interference lithography (LIL) when periodic feature fabrication is desired. Moreover, interference lithography is best suited for 3D periodic feature fabrication which is very difficult to achieve using other techniques. Various interference lithographic configurations exist which generate uniform, sub-micrometre array structures over large area. These can be classified into two major groups: amplitude division and wavefront division techniques. In amplitude division methods, the incoming coherent beam is split and is made to interfere on the recording surface. However, most of these configurations are error prone owing to complex setup and beam path sensitivity. Wavefront division method on the other hand involves splitting the incoming wave into wave fronts and superposing them to form periodic patterns. The path length traversed by interfering wave fronts is very small. Among different wavefront division techniques, the one employing biprism has the ability to

fabricate large area periodic structures with sub-micron resolution using minimum number of optical components. In this context, this thesis initially focuses on investigating this prism based zero path interferometric lithography. Later, novel prism based maskless configurations employing the immersion lithography concept are also examined to fabricate periodic features with improved pitch. The polarization state of individual interfering beams also plays an important role in interference lithography. A broad range of intensity patterns can be produced by varying the polarization states of individual beams. In this context, this thesis also focuses on analysing the effect of polarization, in two-beam, four-beam and five-beam interference configurations, for producing 1D, 2D and 3D periodic structures respectively.

Periodic sub-wavelength features are widely used in recent times in various areas of nanophotonics. Lithography employing periodic 2D metallic masks and surface plasmon phenomena has been researched recently to pattern features with resolution scaling beyond the diffraction limit. However, in such surface plasmon lithographic techniques, the propagation length of surface plasmon polariton (SPP) is very small because of the high power dissipation in the metal. In this context, it is advantageous to investigate new approaches for surface plasmon lithography with the capability of compensating for these losses. Employing a dye based gain medium around the metallic mask is expected to compensate these losses and increase the propagation length of surface plasmon polariton. This thesis investigates such a lithographic technique which utilizes

periodic metallic mask and dye to generate SPP and enhances its propagation length.

Photovoltaic's is one area of nanophotonics which utilizes periodic structures to improve efficiency. In silicon based thin film solar cell, light trapping techniques need to be employed to enhance optical absorption inside the thin active layer. This helps in reduction of material thickness and hence a corresponding reduction in production cost. Light trapping by means of texturing front and back surface and by using various plasmonic concepts has been investigated in detail. However, there is still a lot of scope for improvement in the efficiency level of thin film solar cell in order to meet the commercial requirements and so it is essential to look into novel concepts for light trapping and absorption enhancement. In this thesis, the usage of periodic structures for light trapping, both at the front and back surface along with introduction of recent plasmonic concept of gap modes has also been investigated.

Chapter 3: Multi-Facet Prism based Laser

Interference Lithography

This chapter illustrates periodic feature patterning using laser interference lithography based on multi facet prisms. Theoretical analysis of pitch dependency on various factors for a prism based interferometer and experimental analysis of periodic patterning using biprism, trip Prism and pyramidal prism based configurations have been demonstrated in detail. Highly uniform patterns with micron and sub-micron periodicity was fabricated using this relatively simple whole field patterning technique using both UV and DUV laser sources.

3.1 Introduction

Laser interference lithography (LIL) is a relatively simple process where the standing wave patterns obtained by the interference of two or more coherent beams, are recorded in a photoresist medium. The configurations used for LIL in general can be divided into two: amplitude division and wavefront division techniques [98-100]. In amplitude division configurations, the incoming coherent beam is split into two or more beams using either beam splitter or grating arrangement and it is later interfered on the surface of the wafer sample. These configurations are more error prone owing to the complex setup employing large number of optical components and large beam path. Wavefront division configurations on the other hand use a single optical element to split the

incoming wave into wave fronts and these wavefront's are later superposed near to the optical element to form periodic patterns. Among different wavefront division techniques, the one employing biprism is of particular interest, mainly because of its ability to fabricate large area 2D periodic structures with sub-micron resolution, employing a relatively simple setup which uses minimum number of optical components. A prism based method is a zero path UV laser interference lithographic technique where the wavefront's are made to interfere immediately outside the prism surface thus reducing the beam travel distance in air after splitting. In this chapter, such a prism based interferometric technique, which employs various facet prisms along with ultraviolet (UV) and deep ultraviolet (DUV) laser sources, is investigated for the fabrication of two dimensional structures using single and double exposure technique.

3.2 Theory

3.2.1 Biprism theory using ray optics

To understand the basic theory behind multi facet prism based interferometric technique, a biprism is chosen to be the basic element. A biprism consists of two thin prisms joined at their bases to form an isosceles triangle [101]. In a biprism interferometer employing a point source, a single wavefront impinges on both the thin prisms. The top portion of the wavefront is refracted downward and the bottom portion is refracted upward. Interference occurs in the region of superposition producing alternate dark and bright fringes

corresponding to destructive and constructive interference respectively. Fringe pitch (p) is given by following Eq. (3.1).

$$p = \frac{\lambda D}{d}; \quad \text{for } D \gg d \quad 3.1$$

Where, λ is the wavelength of the light used, D is the separation between source and sample and d is the distance between two virtual coherent sources obtained by tracing back the refracted rays from biprism as given in the following Eq. (3.2).

$$d \cong 2\alpha(n - 1)a \quad 3.2$$

Where α is the biprism angle which is generally very small and ‘ a ’ is the distance between the source and biprism. As seen in Eq. (3.1) and Eq. (3.2), the fringe width depends on the wavelength of the light, the biprism angle, the refractive index of the biprism material, the distance of the sample from the biprism and that of the source from the biprism.

But if the incoming beam is considered to be collimated, the number of parameters on which the fringe width depends would be reduced. This can be understood considering the standard case of two-beam interference where the pitch p is given by,

$$p = \frac{\lambda}{2\sin\theta} \quad 3.3$$

Where, θ is the half angle of intersection of the two interfering beam's.

When the incoming beam is collimated, all the rays incident on the biprism suffers equal amount of refraction. These refracted rays interfere at the output at an angle θ , which is the same over the entire region of interference. This interference phenomena as shown in Figure 3.1 can now be characterized by Eq. (3.3), where the half angle of intersection (θ) becomes equal to the deviation angle ϕ of the biprism, as given by,

$$\theta = \phi = \arcsin(n \sin \alpha) - \alpha \quad 3.4$$

And the pitch of the interference fringes is given by,

$$p = \frac{\lambda}{2 \sin[\arcsin(n \sin \alpha) - \alpha]} \quad 3.5$$

For small α Eq. (3.4) can be approximated as,

$$\theta = \phi \cong (n - 1)\alpha \quad 3.6$$

And the pitch for small α is given by,

$$p \cong \frac{\lambda}{2 \sin [(n - 1)\alpha]} \quad 3.7$$

Thus by employing collimated beam, the fringe pitch given by Eq. (3.5) and Eq. (3.7), depends only on wavelength (λ), biprism angle (α) and the refractive index of the biprism material (n). This equation can be applied not just to biprism but also for other multi facet prism based interferometer too. The interference area could be changed by varying the position of the sample holder. As shown in Figure 3.1, the area of the interference is at its maximum when the recording plane is at a distance X from the biprism given by,

$$X = \frac{r}{2} \frac{(1 - \tan \theta \tan \alpha)}{\tan \theta} \quad 3.8$$

And the width h at this plane, as indicated in Figure 3.1, is given by,

$$h = r(1 - \tan \theta \tan \alpha) \quad 3.9$$

Where θ is obtained using Eq. (3.4).

Figure 3.1 Biprism illuminated with collimated beam.

The dependence of the half angle of intersection θ over the biprism angle α for four different commercially available glass materials such as N-LASF ($n=1.93$), F2 ($n=1.66$), BK7 ($n=1.53$) and Lithotec CAF2 ($n=1.44$) are given in Figure 3.2 and the dependence of the normalized pitch (p/λ) over half angle of intersection θ at an operating wavelength of $\lambda = 363.8nm$ is shown Figure 3.3. It should be noted that the linear dependence as per Eq. (3.6) is given by the dashed line in Figure 3.2 for a refractive index of 1.44. It is evident from these graphs that if the biprism angle is increased, the pitch decreases. This happens

until the biprism angle limit is reached. The total internal reflection limit is shown by the dotted line in Figure 3.2, corresponding to the equation $\theta = 90 - \alpha$. At lower values of biprism angle, half angle of intersection θ is linearly dependent on the biprism angle α as given by Eq. (3.6). Correlating both graphs in Figure 3.2 and Figure 3.3, it is evident that the pitch size decreases when the biprism angle is increased. The refractive index of the biprism material has a significant effect on the resulting pitch. With higher refractive index material and with a higher biprism angle as permitted by TIR limit, a lower pitch can be attained. The material of interest here is BK7 which has a transmittance of 90% at an operating wavelength of 363.8 nm. With a BK7 biprism, which has a refractive index of 1.53 at 363.8 nm, a minimum pitch of 248 nm can be obtained when a biprism of angle 40.8° is used. This is shown by point ‘a’ in Figure 3.2 and Figure 3.3. As discussed, pitch can be further reduced by employing higher refractive index material. Using a N-LASF31A biprism, which has a higher refractive index of 1.93 at the operating wavelength, the minimum estimated pitch is 212.6 nm, as given by point ‘b’ in Figure 3.2 and Figure 3.3. For reducing the pitch further, the operating wavelength needs to be reduced. It can also be noted from Figure 3.3 that for small angles, the angular dependence is larger.

Figure 3.2 Dependence of half angle of intersection θ over biprism angle α for four different glass materials.

Figure 3.3 Dependence of normalized pitch (p/λ) over half angle of intersection θ at operating wavelength of $\lambda = 363.8$ nm.

3.2.2 Biprism interferometry using deep UV laser source

From Eq. (3.5), it is evident that the pitch size is directly proportional to operating wavelength. To reduce pitch, a light source of lower wavelength needs to be used and it does not hamper the area of interference if the same beam diameter is employed. As evident from Figure 3.1, the width h at the position X where you have largest interference area is given by Eq. (3.9), and it depends on the beam diameter and biprism angle. So a variation in wavelength only results in a change of pitch, as defined by Eq. (3.5), and not a change of area of interference.

Figure 3.4 gives a comparative analysis of the achievable pitch when two different wavelengths: $\lambda_1 = 266 \text{ nm}$ and $\lambda_2 = 364 \text{ nm}$, are used. A pitch reduction with 266 nm source is apparent from the graphs. For example, when the half angle of intersection is 20° , the ‘i’ line source gives a pitch of 532nm, were as a 266 nm source reduces it to 389nm. In this work, as will be discussed in detailed in the later part of this chapter, a DUV source of wavelength 266 nm is employed to fabricate patterns with sub-500 nm periodicity. Biprism and pyramidal prisms made of fused silica with an angle of 30.4° has been employed for this purpose. Now, fused silica material has a refractive index of 1.49968 at 266nm. And according to Eq. (3.5), the theoretically achievable fringe pitch with a 30.4° biprism and a 266 nm collimated light beam is 418.3nm. When the same biprism is illuminated with a 364 nm source, the theoretically obtained fringe pitch was 572.5nm, which is much higher compared to former case.

Figure 3.4 Dependence of pitch over half angle of intersection at operating wavelength of $\lambda_1 = 266 \text{ nm}$ and $\lambda_2 = 364 \text{ nm}$ (with reference to Eq. (3.1)). It can be seen that at the same half angle of intersection, λ_1 provides much smaller pitch compared to λ_2 .

3.3 Experiments and Results

3.3.1 Patterning using Biprism Based Two-beam Interference Lithography and 364 nm UV source

The schematic diagram of the experimental setup used is given in Figure 3.5. Initially, a continuous wave UV Argon Ion laser (Spectra Physics, 363.8 nm wavelength) is employed as the source for the interferometer. The high energy spatially filtered laser beam is expanded by a factor of ten and collimated using a 10X beam expander/collimator. This collimated beam then illuminates the biprism to achieve wavefront splitting. A photoresist coated silicon wafer is mounted onto the rotatable sample holder. An electronic shutter placed in front

of the laser head provides control over the exposure time. The biprism is positioned in such a way that half of the expanded beam falls on the upper part of the prism and half on the lower part. The entire set up is placed as close to the laser head as possible in order to minimize the effect of air circulation in the experimental room on laser beams.

Figure 3.5 Experimental setup of biprism based two-beam interference lithography.

Biprism based lithography is used to produce grating structures employing single exposure and square lattice structures employing double exposure [102]. Single exposure lithography technique using positive photoresist is expected to produce periodic array of line pattern as in Figure 3.6(a), under optimized dose condition. On exposure and subsequent development, areas where dark fringes are formed should remain and the rest should get washed away. Positive photoresist AZ7220 is used along with a biprism to fabricate grating structures and pillars in a square lattice. Figure 3.7(a) shows such a grating structures fabricated using a 5° BK7 biprism having a pitch of

$3.75 \pm 0.02 \mu\text{m}$. Feature size can be varied by changing the exposure dose, which is a product of irradiance and exposure time. An increase in exposure dose results in wider patterns on photoresist. This in turn results in a reduction of feature size when positive photoresist is used. This is illustrated in Figure 3.7(a) and Figure 3.7(b), where an increase in exposure dose from 80 mJ/cm^2 to 200 mJ/cm^2 results in a reduction of feature width. Furthermore, exposing the sample twice with a 90° sample rotation in between the exposures will provide square lattice features. This is illustrated in Figure 3.8 where discrete photoresist pillars are fabricated using a 5° BK7 biprism. The irradiance and duration of each exposure used is 20 mW/cm^2 and 5 s respectively. An increase in biprism angle results in a reduction of pitch as mentioned earlier. A biprism of 12.5° acute angle is employed to fabricate square lattice structures of reduced pitch. A double exposure technique, with a dose of 80 mJ/cm^2 during each exposure, produced square lattice structures of pitch size $1.6 \pm 0.01 \mu\text{m}$, as shown in Figure 3.9. Structures obtained here are interconnected, which can be made discrete by optimizing the exposure dose and development time. By using Hexamethyldisilazane (HDMS) primer, and setting the exposure dose to 40 mJ/cm^2 and the development time to 50 sec , discrete structures were obtained, as shown in Figure 3.10. The primer mostly used to improve adhesion between the silicon wafer and resist, also acts as an antireflection agent. To reduce the pitch further, a biprism of 30° acute angle is employed. The fabricated structures are shown in Figure 3.11 have an average pitch of $450 \pm 8 \text{ nm}$, but are

observed to be distorted. The possible reasons for distortion are assumed to be back reflection, interference of ghost beams, and the difference in polarization.

Figure 3.6 (a) Grating pattern (b) Square lattice pattern (c) Hexagonal lattice pattern.

Figure 3.7 Grating structures fabricated using 5° biprism (a) Structures with an exposure dose of 80 mJ/cm² (b) Structures when the exposure dose was increased to 200 mJ/cm².

Figure 3.8 SEM images of features fabricated using 5° BK7 biprism (a)Top view (b) Slanted view.

Figure 3.9 SEM images of features fabricated using 12.5° BK7 biprism (a)Top view (b) Slanted view.

Figure 3.10 SEM images of features fabricated using 12.5° BK7 biprism (a)Top view (b) Slanted view.

Figure 3.11 SEM image (top view) of feature fabricated using 30° BK7 biprism.

3.3.2 Triprism Based Three Beam Interference Lithography and 364 nm UV source

So far it has been illustrated that double exposure LIL using biprism yields square lattice patterns. To obtain hexagonal lattice patterns using two-beam interference, normally double exposure is employed with a 60° sample rotation between exposures [102, 103]. The patterns are made more circular using triple exposure by rotating the sample by 60° and 120° , in between exposures. In this work, a three facet prism or triprism is used to produce hexagonal lattice structures, employing only one exposure. The need for sample rotation between exposures is eliminated by the use of the triprism, which functions based on the principle of three wave interference. Fabricated features consist of a hexagonal array of holes and pillars as shown in Figure 3.12. Both positive and negative photoresist's are employed with three facet prism to produce holes and pillars in a hexagonal lattice respectively. Initially, a triprism is used along with AZ7220 coated wafer to obtain hexagonal lattice patterns as

shown in Figure 3.6(c). Here, a single exposure, using a triprism, produces hexagonal patterns on the resist.

Figure 3.12(a) shows the array of holes fabricated using positive photoresist AZ7220 at an exposure dose of 40 mJ/cm^2 . And employing negative photoresist SU8 2000.5, array of pillars at an exposure dose of 25 mJ/cm^2 are obtained, as given in Figure 3.12(b). In the former case where positive resist is employed, the developer dissolves only areas of maximum intensity in the hexagonal lattice as indicated in the Figure 3.6(c), which are formed by constructive interference of three wavefront's and produces holes on the photoresist. Meanwhile when negative photoresist is used, the areas of maximum intensity remains and inverted structures i.e., pillars are obtained. By controlling the exposure dose positive resist could be used to fabricate pillars too. This is shown in Figure 3.13, where the exposure dose was increased to 200 mJ/cm^2 , to obtain resist pillars. The major limitation of this technique is that one multi-facet prism gives only one particular pitch. Variation of pitch over a wide range could be realized by employing different angle prisms or by using different refractive index material for the prism. However, this particular interferometric method, employing a multi-facet prism, is best suited for applications where one particular pitch is desired over a large area.

Figure 3.12 Structures fabricated using tripism (a) SEM image of holes in a hexagonal lattice obtained using positive photoresist AZ7220 at an exposure dose of 40 mJ/cm²(b) Pillars in hexagonal lattice obtained using negative photoresist SU-8 2000.5 at an exposure dose of 25 mJ/cm².

Figure 3.13 Structures fabricated using tripism (a) SEM image of holes in a hexagonal lattice obtained using positive photoresist AZ7220 at an exposure dose of 40 mJ/cm² (b) Interconnected Pillars in hexagonal lattice obtained by increasing the exposure dose to 88 mJ/cm² (c) Pillars in hexagonal lattice obtained at a dose of 200 mJ/cm².

3.3.3 Prism based Deep UV Lithography (Biprism & Pyramidal prism)

The experimental setup is very similar to that discussed before in Figure 3.5 and is schematically illustrated in Figure 3.14. A Gaussian beam from a 266 nm pulsed light source is directed on to a custom fabricated prism made of fused

silica after expansion(10X) and collimation. The sample is mounted on a rotatable wafer stage which is placed in the region of interference to record the interference pattern. An electronic shutter is installed just after the light source to precisely control the exposure time and in turn the exposure dose. A positive tone photoresist, AZ7220 (AZ Electronics Material) is used to record the interference patterns. A thinning process is performed by mixing the resist with EBR solvent (PGMEA) in the ratio 1:1, after which it is spin coated at a speed of 1500 rpm on a cleaned silicon wafer. After the exposure, patterns are developed using AZ300MIF developer. Gratings were fabricated by a single exposure technique and square lattice features using a double exposure technique using a biprism. In the latter case the sample was rotated by 90° between the two exposures. The results obtained are discussed in the following section.

A grating line distribution, with a period of 418.3nm, obtained is expected to be produced by the interference of two coherent wavefront's by employing biprism of angle 30.4° along with 266 nm laser. The 2D periodic intensity distribution obtained by combining four coherent plane waves will have, three major intensity regions namely high (H), medium (M) and low (L). This is discussed in detail in chapter 5. With a positive tone resist and after optimization of process parameters such as exposure dose and developing time, holes in the H region and pillars in the L regions are expected to be fabricated. Prior to obtaining these distinct features, the M region comes into play and forms interconnection between the holes and pillars, which could be removed by increasing the exposure dose.

Figure 3.14 Experimental setup using (a) biprism and (b) pyramidal prism along with a DUV source.

Figure 3.15 SEM images of grating lines fabricated using 266 nm light source and biprism of angle 30.4° : (a) Magnified and (b) Demagnified view. Pitch size of around 410 ± 11 nm and feature width of 206 ± 5 nm is observed.

Initially, using a single exposure technique, gratings were fabricated as shown in Figure 3.15. The magnified and demagnified image of the patterns produced are shown in Figure 3.15(a) and Figure 3.15(b) respectively. Lines with pitch around 410 ± 11 nm and feature width of around 206 ± 5 nm were obtained employing a 30.4° biprism. The exposure dose and development time used are 54 mJ/cm^2 and 10s respectively. In subsequent experiments, a, double exposure technique employing the same prism was used to fabricate square lattice features (see the scanning electron microscopy (SEM) images shown in Figure 3.16 and Figure 3.17). Initially, a dose of 54 mJ/cm^2 was used during each exposure and the sample was developed for 10 s. Interconnected structures as shown in Figure 3.16 were obtained in a relatively large area of about 2 mm^2 . The exposure dose was later doubled to 108 mJ/cm^2 to distinguish the features and the obtained pillar patterns are shown in Figure 3.17. Demagnified and magnified image of the patterns produced are shown in Figure 3.17(a) and Figure 3.17(b) respectively. These features have a resolution around 200 nm, with a minimum of about 190 nm. In both cases, average pitch size was calculated to be around 410 ± 11 nm which is very close to the theoretical value of 418.3 nm obtained earlier using Eq. (3.5). Initially, a double exposure technique has been employed here to fabricate square lattice features using a biprism. If a pyramidal prism is employed instead of a biprism, square lattice features could be fabricated with just one exposure, thus removing errors that might arise during the sample rotation between the exposures. Holes with diameter around 270 ± 20 nm in a square lattice with pitch of 408 ± 6 nm were

obtained using a pyramidal prism with prism angle of 30.4° , using positive resist. The SEM images of the fabricated features are shown in Figure 3.18. Further reduction in pitch could be achieved by employing a prism of higher refractive index. But this is not feasible as most of the commonly available glass material has very low transparency at 266 nm. Another option is to employ a large angle prism for sub-350 nm pitch at the expense of interference area.

Figure 3.16 SEM images of interconnected resist (AZ 7220) structures in square lattice produced using 266 nm and biprism of 30.4° angle: (a) Top view, (b) Slanted view.

Figure 3.17 SEM images of distinguished resist features in square lattice with a pitch of around 410 ± 11 nm and smallest feature size of 190 nm fabricated by doubling the exposure dose: (a) demagnified and (b) magnified view. Note that the middle region of interconnection has been removed here.

Figure 3.18 SEM images of holes in square lattice fabricated using a pyramidal prism using single exposure technique. (a) demagnified and (b) magnified view. Features were found to have a pitch size of 408 ± 6 nm and hole diameter of 270 ± 20 nm.

3.4 Summary

In this chapter, investigations into the wavefront division approach for interference lithography has been performed. The use of simple UV and DUV lithography configurations using multi-facet prisms to fabricate two dimensional periodic structures in a large area was demonstrated. Initially, 5° , 2.5° and 30° biprism's were used to fabricate grating and square array of pillars using a 364 nm laser source in a relatively large area at a very high speed. Theoretically, it was shown that pitch size can go as small as 210 nm at a wavelength of 363.8 nm, using biprism interferometer lithography. With an i-line laser, feature width could be controlled by controlling exposure dose and development time. Hexagonal features were also fabricated using single exposure triprism based LIL, eliminating the need of multiple exposure and associated rotation of sample in between the exposures. Fabricated features include holes

and pillars in hexagonal lattice employing positive and negative photoresist respectively. Later, a deep UV light source with wavelength of 266 nm was employed in a biprism interferometer configuration to define periodic patterns with sub-500 nm pitch and sub-200 nm resolution. A 30.4° biprism made of fused silica material was employed to fabricate grating lines with pitch size of 410 ± 11 nm, which is in close proximity to the theoretically predicted value. 2D periodic features in square lattice were also fabricated using biprism based double exposure technique and using a pyramidal prism based single exposure technique. The features fabricated include some interesting interconnected structures.

The relatively simple setup and high speed of this technique could be of great use in fabricating features such as 2D photonic crystal with micron to sub-500 nm pitch size. However, one of the major limitations of this prism technique employing collimated beam is that a single prism cannot be used to fabricate features with varying periodicity. Pitch size can be varied by using different angle prisms or by using different refractive index material for the prism. As mentioned before, employing a 364 nm (λ) source and a large angle prism (provided TIR condition is not satisfied), the minimum pitch size which could be achieved is 210 nm. Moreover, the pattern quality and uniformity decline as we go towards this value. So to fabricate uniform high quality patterns with periodicity of the range of $\lambda/2$ and $< \lambda/2$, other techniques has to be explored. In this context, the next chapter focuses on investigating solid immersion lithography for the patterning of high resolution 2D periodic features.

Chapter 4: Periodic Feature Patterning by Prism based Solid Immersion Interference Lithography

In the previous chapter, fabrication of 2-D periodic patterns using a simple multi facet prism based technique was discussed. However, it is an air based technique where the minimum pitch size achievable is $\lambda/2$. In this chapter, focus was laid on achieving periodic features with much smaller pitch size and higher resolution using a prism based solid immersion lithographic technique. Two configurations for solid immersion lithography are mainly investigated, one using a prism-grating based lithographic setup and the other using a multiple lens based configuration

4.1 Introduction

Most of the classical optical interference techniques use air as the medium between the projection optics and the writing medium which limits the maximum achievable numerical aperture of the system to unity [64, 65]. However, the numerical aperture of the system can be increased by employing the concept of immersion lithography, where the refractive index of the surrounding medium is increased to scale down the pitch and feature size. Various configurations have been investigated to date to fabricate features using immersion interference lithography. Most of the reported work explores the implementation of the immersion concept using a deep ultraviolet (DUV) light source [60, 63, 67]. Even though this provides better resolution, it requires deep

UV optics for its implementation, which is very expensive. In this research, focus was laid on developing a lithographic system to achieve higher resolution using an i-line laser source and visible wavelength optics.

Two optical configurations based on a 364 nm laser source, a high index prism and solid immersion lithography (SIL) concepts are investigated to achieve periodic features with a sub-100 nm half pitch size. The first configuration involves the use of a conventional grating and mirror based arrangement and is discussed in the next section. And the second configuration uses lenses instead of mirrors to guide the laser beams and is discussed in detail in the later part of this chapter.

4.2 Prism-grating and mirror based SIL

A schematic of the grating and mirror based immersion system is shown in Figure 4.1. In this configuration, a spatially filtered 364 nm laser beam is directed on to a 2D transmission grating which splits the incoming beam. Multiple first order diffracted beams from the grating are used for interference. The transmission grating is designed to provide first order beams at an angle of 45° to the normal. The zero-order beam is blocked by a stopper. Vertically aligned mirrors are used to direct the beam on to the high index prism. Mirrors are provided with rotation capability to control the angle of incidence (α) of the beams. Beams are then made to intersect at the prism base at an angle θ to the normal. This is a conventional interference configuration where mirrors are used

to guide the beams. And as mentioned, beams are made to interfere inside a prism with very high refractive index and transmission at 364 nm, thus realizing the immersion concept.

Figure 4.1 Experimental setup for grating & mirror based solid immersion lithography.

4.2.1 Theory and experiment

Fringe pitch (p_{air}) of the patterns produced by interference of two coherent beams in air (assuming α as the half angle of intersection) is given by the following equation.

$$p_{air} = \frac{\lambda}{2 \sin \alpha} \quad 4.1$$

And pitch size (p_n), when the interference takes place in a medium of refractive index ' n ' and with half angle of intersection θ is given by,

$$p_n = \frac{\lambda}{2 n \sin \theta} \quad 4.2$$

Where, λ is the wavelength of the laser beam, n is the refractive index of the surrounding medium and θ is the half angle of intersection of the two beams inside the prism. The obvious way to reduce feature size is to decrease the wavelength. But as mentioned before, going into deep ultraviolet range requires expensive lasers with large coherence length and expensive DUV optics. Moreover, for the implementation of the solid immersion concept, the medium should have minimum absorbance as well. Increasing the second factor n is another effective way to reduce the period. Here, this is achieved by employing a high index prism and making the beams interfere at the bottom of the prism. This concept is called solid immersion lithography, where the patterns are later transferred to the resist using an index matching liquid. The pitch size could be reduced almost by a factor of 2 by employing suitable prism material. The third factor on which the pitch depends is the half angle of intersection θ . Here, since a prism is employed for immersion lithography, this refers to the half angle of intersection inside the prism at its bottom surface. The larger the angle of intersection, the smaller is the period. But it is limited by the critical angle of incidence at the prism-index matching liquid interface.

Figure.4.2 Schematic of high index prism used for solid immersion lithography.

Now consider Figure.4.2, which shows a schematic representation of beams interfering inside the prism (with refractive index n). The pitch size of the patterns produced by the interference of two coherent beams is given by,

$$p_n = p_{prism} = \frac{\lambda}{2 n \sin \theta} \quad 4.3$$

Where the half angle of intersection (θ) inside the prism (of angle β) can be derived by applying snell's law at the slanted surface. Let the angle the incident beam makes with respect to vertical axis be α . Since the prism angle is β , the angle of incidence with respect to the normal to the slanted prism surface is ' $\beta - \alpha$ ' as shown in Figure.4.2. Applying Snell's law at the slanted surface,

$$\begin{aligned} \sin(\beta - \alpha) &= n \sin(\theta_1) \\ \Rightarrow \theta_1 &= \sin^{-1} \left(\frac{\sin(\beta - \alpha)}{n} \right) \end{aligned} \quad 4.4$$

Again from Figure.4.2,

$$\theta = \theta_2 = \beta - \theta_1$$

Substituting θ_1 from Eq. (4.4), the half angle of intersection inside the prism (θ) is given by,

$$\theta = \beta - \sin^{-1} \left(\frac{\sin(\beta - \alpha)}{n} \right) \quad 4.5$$

So the pitch size (p_{prism}) can be written as

$$p_{prism} = \frac{\lambda}{2 n \sin \left(\beta - \sin^{-1} \left(\frac{\sin(\beta - \alpha)}{n} \right) \right)} \quad 4.6$$

If the angle of incidence inside the prism increases, the pitch size reduces. But for the patterns to be transferred to the resist, it should be below the critical angle at the prism-index matching liquid interface, which is given by,

$$\theta_{critical} = \sin^{-1} \left(\frac{n_{liquid}}{n} \right) \quad 4.7$$

In this work, the refractive index of the resist and the contact liquid employed is 1.7. This helps in enabling efficient pattern transfer. Now, the critical angle at the high index prism (1.939) and liquid (1.7) interface is calculated as 61.25° . Variation of the half angle of intersection (θ) inside the prism (of angle $\beta = 60^\circ$) and pitch size of the patterns (p_{prism}) with respect to the angle of incidence (α) are plotted in Figure 4.3 and Figure 4.4 respectively. It is evident from the plot in Figure 4.3 that the critical angle criteria for this configuration ($\theta_{critical} = 61.25^\circ$) is satisfied when the incidence angle $\alpha = 62.4^\circ$. From Figure 4.4, it can be noted that the pitch can go as small as 107 nm when the angle of incidence (α) takes the value of 62.4° . To scale down beyond this

value using the same configuration, a higher index matching liquid need to be employed.

Figure 4.3 Angle of incidence of the beam with respect to vertical axis in air (α) vs that inside the 60° prism (θ) based on Eq. (4.5).

Figure 4.4 Pitch size of the grating patterns vs. angle of incidence in air (α) based on Eq. (4.6).

In this work, the angle of incidence was set to 60° , and according to Figure 4.4, the fabricated patterns should have a pitch size of 108 nm . A positive tone photoresist, AZ7220 (AZ Electronics Material) was employed to record the interference patterns. The resist is spin coated at a speed of 4000 rpm on a cleaned silicon wafer. Index matching liquid ($n = 1.7$) from cagillere (series B) is then applied on the resist. This sample is then placed beneath the prism on the sample holder. After the exposure, patterns are developed using AZ300MIF developer. High resolution grating lines were fabricated by a single exposure of only one second. The results obtained are discussed in the next section.

4.2.2 Experimental results and discussion

SEM images of the features patterned on positive tone resist are shown in Figure 4.5. Large area, highly uniform grating patterns are clearly visible in Figure 4.5(a). The pitch size of the fabricated grating is around $110 \pm 6\text{ nm}$ and the half pitch is around $55 \pm 4\text{ nm}$, as shown in the magnified SEM image in Figure 4.5(b). This is quite close to the theoretically estimated value of 108 nm . The slight deviation can be attributed to manufacturing defect of the prism or error in controlling incidence angle. As evident from Eq. (4.2), pitch can be scaled down by either using a lower wavelength light source, a prism of higher refractive index or increasing θ . A lower wavelength source has already been successfully employed to fabricate higher resolution features, but this requires deep UV optical components and a prism with high DUV transmittance, which raises the cost of production and system complexity significantly. However, the

current setup mainly employs visible optics making the system simpler. The choice of prism material also becomes simpler when an i-line source is employed, which is not the case with a DUV system. Sapphire material is a good option at DUV wavelength as it has a higher refractive index and transmission, but it is very expensive compared to an i-line prism (for example LAK31A prism used in this work). That said, if the application requires features well below 90 nm period, focus has to be turned to these much more expensive options. The pitch size can also be reduced by increasing the half angle of intersection (θ). However, the critical angle bar needs to be raised to permit higher intersection angle at the prism-liquid interface. The maximum θ can go in this current setup is 61.25° with an index matching liquid of $n = 1.7$. A liquid with index even closer to the refractive index of prism (1.939) will raise the critical angle bar well above the value of 61.25° . This in turn necessitates the need of a resist with a higher matching index for efficient pattern transfer.

Figure 4.5 SEM images of the fabricated grating pattern :(a) demagnified and (b) magnified view. Patterns were found to have sub-60 nm half pitch resolution.

4.3 Lens and prism based laser interference SIL

Most of the solid immersion interference configurations require larger number of optical components like beam splitters, mirrors etc and this number increases with increase in number of beams involved as seen in the previous configuration. In this context, this section focuses on the lithographic system using the same i-line laser source and a multiple converging lens configuration. This is achieved by employing multiple lenses to collect and direct the beams on to the sample instead of multiple mirror assembly. A simple optical configuration using a 60° prism, 364 nm laser source and converging lenses shown in Figure 4.6 is proposed to implement the immersion lithography concept.

Figure 4.6 Proposed lens based configuration for SIL.

4.3.1 Theory

As mentioned in the previous section, lenses are employed instead of mirrors to direct beams on to the sample. And then the beams are guided to the sample through a high index prism to achieve better resolution. Converging lenses are used to achieve this objective. It could be achieved by either using a single converging lens or multiple lenses. In this section, the pros and cons of both cases are discussed in detail. The dependency of pitch size of the periodic patterns on various factors like the splitting angle, the distance of the lens from the sample and splitter, focal length etc are studied. The number of factors affecting θ increases with the number of lenses employed and in turn gives better experimental control. All these issues are discussed in the following sections, starting with single lens system.

4.3.1.1 Theory: Lens based SIL - Single Lens system

Figure 4.7 Schematic of single converging lens system used in the proposed solid immersion lithography configuration.

Let's first consider a thin biconvex lens with focal length f , as shown in Figure 4.7 as the converging lens used in the proposed lithographic system. If the distances from the object to a lens and from the lens to the image are u and v respectively, for a lens of negligible thickness, in air, the distances are related by the thin lens formula given by,

$$\frac{1}{f} = \frac{1}{u} + \frac{1}{v} \quad 4.8$$

$$v = \frac{u \cdot f}{u - f} \quad 4.9$$

From Figure 4.7,

$$\begin{aligned} \tan \alpha &= \frac{h}{v} \quad \& \quad \tan \varphi = \frac{h}{u} \\ \Rightarrow \tan \alpha &= \frac{u \cdot \tan \varphi}{v} \end{aligned} \quad 4.10$$

Substituting v from Eq. (4.9),

$$\Rightarrow \alpha = \tan^{-1} \left(\frac{(u - f) \cdot \tan \varphi}{f} \right) \quad 4.11$$

Substituting α from Eq. (4.11) in Eq. (4.5), the half angle of intersection in a single lens based system is obtained and it is given by,

$$\theta = \beta - \sin^{-1} \left(\frac{\sin \left(\beta - \tan^{-1} \left(\frac{(u - f) \cdot \tan \varphi}{f} \right) \right)}{n} \right) \quad 4.12$$

And so pitch size (p) given by Eq. (4.2) now becomes,

$$p = \frac{\lambda}{2 n \sin \beta - \sin^{-1} \left(\frac{\sin \left(\beta - \tan^{-1} \left(\frac{(u-f) \cdot \tan \varphi}{f} \right) \right)}{n} \right)} \quad 4.13$$

In this work, a LAK31A prism of angle $\beta = 60^\circ$ and index $n = 1.939$ is employed. An inverted pyramidal prism is used to split the beam at an angle $\varphi = 25^\circ$. Consider a biconvex lens of diameter and focal length 100 mm. The dependency of pitch on the distance of lens from the splitter (u) is given by the plot in Figure 4.8. It can be noted that an increase in the distance u results in a smaller pitch. But it also means an increase in working distance, which is not preferred. For a smaller pitch, a lens with a larger diameter should be used to collect and refract the beams. For example, using a biconvex lens of diameter 100 mm, and split angle $\varphi = 25^\circ$, the maximum u can go is up to 107 mm. And when the working distance is calculated ($\sim u + v$), it comes closer to 1528 mm, which is very large and so is not practical to implement in a compact optical system. For a particular pitch this can be reduced by increasing the split angle, but this in turn requires a lens with larger diameter (>100 mm). The working distance could be brought down considerably by employing another converging lens of approximately equal diameter, which is discussed in the following section.

Figure 4.8 Pitch (p) dependency on lens distance from the splitter (u) for single biconvex lens system.

4.3.1.2 Theory: Lens based SIL - Two Lens System

Now consider a two lens converging system for the proposed interference configuration. Let f_1 be the focal length of the first lens and f_2 be the focal length of the second lens. Applying the lens formula from Eq. (4.8) to the first lens in the system shown in Figure 4.9,

$$\frac{1}{f_1} = \frac{1}{u} + \frac{1}{v} \quad 4.14$$

It can be rewritten as,

$$v = \frac{u \cdot f_1}{u - f_1} \quad 4.15$$

Figure 4.9 Schematic representation of two converging lens system used in the proposed solid immersion lithography configuration.

Applying the formula from Eq. (4.8) to the second lens in the system shown in Figure 4.9,

$$\frac{1}{f_2} = \frac{1}{v'} - \frac{1}{v-d}$$

This can be rewritten as,

$$v' = \frac{(v-d) \cdot f_2}{v-d+f_2} \quad 4.16$$

Again from Figure 4.9,

$$h_1 = u \cdot \tan \phi \quad 4.17$$

Now,

$$\tan \gamma = \frac{h_1}{v} = \frac{h_2}{v-d} \quad 4.18$$

$$\Rightarrow h_2 = \frac{h_1 \cdot (v-d)}{v}$$

$$\Rightarrow h_2 = \frac{u \cdot \tan \varphi \cdot (v-d)}{v} \quad 4.19$$

Again from Figure 4.9,

$$\tan \alpha = \frac{h_2}{v'}$$

Substituting v' and h_2 from Eq. (4.16) and Eq. (4.19) in the above equation,

$$\Rightarrow \tan \alpha = \frac{u \cdot \tan \varphi \cdot (v-d)}{v} \cdot \frac{(v-d+f_2)}{[(v-d) \cdot f_2]}$$

$$\tan \alpha = \frac{u \cdot \tan \varphi}{v} \cdot \frac{(v-d+f_2)}{f_2}$$

Now substituting v from Eq. (4.15) in the above equation we get,

$$\Rightarrow \tan \alpha = \frac{u \cdot \tan \varphi}{\left(\frac{u \cdot f_1}{u-f_1}\right)} \cdot \frac{\left(\frac{u \cdot f_1}{u-f_1} - d + f_2\right)}{f_2}$$

$$\tan \alpha = \frac{\tan \varphi}{f_1 \cdot f_2} \cdot (u \cdot f_1 - u \cdot d + d \cdot f_1 + u \cdot f_2 - f_1 \cdot f_2)$$

$$\tan \alpha = \tan \varphi \cdot \left(\frac{u}{f_1} + \frac{u}{f_2} - \frac{u \cdot d}{f_1 \cdot f_2} + \frac{d}{f_2} - 1\right)$$

$$\Rightarrow \alpha = \tan^{-1} \left(\tan \varphi \cdot \left(\frac{u}{f_1} + \frac{u}{f_2} - \frac{u \cdot d}{f_1 \cdot f_2} + \frac{d}{f_2} - 1\right) \right) \quad 4.20$$

Now consider a laser beam incident on a high index prism (n), at an angle α to the normal. This is illustrated in Figure 4.9 Schematic representation of two

converging lens system used in the proposed solid immersion lithography configuration. If the prism angle is β and the half angle of intersection at the base of the prism is θ , according to Eq. (4.5), it can be related as,

$$\theta = \beta - \sin^{-1} \left(\frac{\sin \left(\beta - \tan^{-1} \left(\tan \varphi \cdot \left(\frac{u}{f_1} + \frac{u}{f_2} - \frac{u \cdot d}{f_1 \cdot f_2} + \frac{d}{f_2} - 1 \right) \right) \right)}{n} \right) \quad 4.21$$

Where, α is given by Eq. (4.20). Now, the pitch (p) of the resulting interference pattern is given by,

$$p = \frac{\lambda}{2 n \sin \left(\beta - \sin^{-1} \left(\frac{\sin \left(\beta - \tan^{-1} \left(\tan \varphi \cdot \left(\frac{u}{f_1} + \frac{u}{f_2} - \frac{u \cdot d}{f_1 \cdot f_2} + \frac{d}{f_2} - 1 \right) \right) \right)}{n} \right) \right)} \quad 4.22$$

Compared to Eq. (4.13) for a single lens system, the number of parameters on which the pitch depends is seen to be increased. It now depends on the focal lengths of both the lenses (f_1 & f_2), distance of the first lens from splitter (u), distance between the two lenses (d), refractive index (n), prism angle (β), and wavelength (λ). In this work, a prism of angle $\beta = 60^\circ$ and index $n = 1.939$ is employed and an inverted pyramidal prism is used to split the beam at an angle $\varphi = 25^\circ$. Let us first consider two biconvex lenses of focal lengths and diameters both having the value 100mm. The dependency of pitch on the distance of the first lens from the splitter (u) for these biconvex lenses (for a separation of $d = 10 \text{ mm}$) is studied and is plotted in Figure 4.10(a). It should be noted that much lower pitch could be achieved by employing two lenses instead

of one. Moreover, for $u = 107 \text{ mm}$, and $d = 10 \text{ mm}$ the maximum working distance ($\sim u + d + v'$) from Figure 4.9, is approximated as 94 mm using Eq. (4.15) and Eq. (4.16). This is much smaller when compared to single lens system and is experimentally practical to implement. Fixing u at 40 mm, the variation of pitch with respect to distance (d) between the lenses is plotted in Figure 4.10 (b). It almost follows a straight line and the slope is found to be around .25, which means for a 10 mm movement between the lenses, the pitch size could be changed by 2.5 nm. Thus it provides much accurate control over the achievable pitch size at much smaller working distance. With the same lenses, if smaller pitch is desired, the split angle ϕ and distance u can be changed accordingly to achieve the target. Although there are more factors determining the half angle of intersection, it helps in precisely controlling the pitch size.

Figure 4.10 Pitch (p) dependency on lens distance from the splitter (u) for a two lens (biconvex) system with focal lengths of $f_1 = 100 \text{ mm}$ and $f_2 = 100 \text{ mm}$ for $d = 10 \text{ mm}$. (b) Variation of pitch (p) with distance (d) between the two lenses for $u = 10 \text{ mm}$.

4.3.2 Experimental Methodology: Lens based SIL

Figure 4.11 Experimental setup.

The experimental setup used is schematically illustrated in Figure 4.11. A spatially filtered 364 nm laser beam is directed on to an inverted pyramidal prism to split it equally into four beams at an angle of 25° with respect to the normal. An inverted prism is used instead of a grating to obtain smaller splitting angle. Moreover, it is cheaper than a custom fabricated transmission grating. A set of converging lens assembly is used to collect and direct the beam on to a high index prism. This consists of biconvex lenses with focal lengths of $f_1 = 75 \text{ mm}$ and $f_2 = 100 \text{ mm}$ and diameters of 75 mm and 100 mm respectively. A positive tone photoresist, AZ7220 (AZ Electronics Material) was employed to record the interference patterns. The resist is spin coated at a speed of 4000 rpm on a cleaned silicon wafer. Index matching liquid ($n=1.7$) from Cagillere is then applied on to the resist. This sample is then placed beneath the prism on the

sample holder. After the exposure, patterns are developed using AZ300MIF developer. Square lattice features were fabricated by single exposure using four-beam interference. The results obtained are discussed in the next section.

4.3.3 Results and discussion: Lens based SIL

In the experimental work, two biconvex lens pairs were employed together with a high index prism and an inverted prism splitter to realize four-beam interference as shown in Figure 4.11. The two biconvex lenses has focal lengths of $f_1 = 75 \text{ mm}$ and $f_2 = 100 \text{ mm}$ and diameters of 75 mm and 100 mm respectively. Here, the beam incident at the top prism is linearly polarized. And the four beams incident at the base of the second prism is in a TE-TM polarized state. Four-beam interference with a TE-TM polarized condition will result in a square lattice pattern with a periodicity of $\sqrt{2}\lambda/2n\sin\theta$, the derivation of which is discussed in detail in chapter 5. The dependency of half angle of intersection inside the prism and the pitch ($\sqrt{2}p$) obtained using Eq. (4.21) and Eq. (4.22), on the distance u for a separation between lenses of $d = 25 \text{ mm}$ is plotted and is shown in Figure 4.12(a) and Figure 4.12(b) respectively. The values of other parameters are shown in the inset. The half angle of intersection could be varied from 30° to 45.3° by varying the distance u from 10 mm to 100 mm . The pitch size varies from 268 nm to 186 nm for a fixed distance between the two lens of $d = 25 \text{ mm}$. When $u = 75 \text{ mm}$, the pitch size does not depend on the distance between the lenses and remains at 205 nm because the beams incident on second lens is collimated at this condition.

Figure 4.12 (a) Dependency of half angle of intersection inside the prism and (b) pitch size ($\sqrt{2}p$) on the distance u for a separation of $d=25$ mm for two lens system with focal lengths 75 mm and 100 mm.

SEM images of the square lattice features patterned on positive tone resist are shown in Figure 4.13 and Figure 4.14. Figure 4.13 shows the square lattice patterns with pitch size of 210 ± 8 nm fabricated with parameters: $u = 70$ mm & $d = 25$ mm, with the lens set described in Figure 4.12. Here, four-beam interference, where one pair of beams is TE polarized and the other pair TM polarized was employed. Figure 4.13(a) shows interconnected square lattice patterns and in Figure 4.13(b) some of the middle regions are shown to be removed. For this condition of $u = 70$ mm & $d = 25$ mm as evident from Figure 4.12, half angle of intersection was expected to be 39.2° . Later the same lens set with focal lengths of 75 mm and 100 mm, was employed to fabricate square lattice features with higher period. The distance parameters were changed to: $u = 40$ mm & $d = 25$ mm, to produce features with slightly higher pitch size of 237 nm as obtained by theory. The half angle of intersection was expected to be

40° as evident from Figure 4.12(a). However, the SEM image shown in Figure 4.14(a) indicates that the obtained pitch size is 240 ± 6 nm. The fabricated pillars were found to have an average diameter of 160 ± 10 nm. The slight deviation can be attributed to manufacturing defect of the prism or error in accurately controlling distance parameters.

Figure 4.13 SEM images of square lattice features fabricated using two converging lens system with distance parameters set at: $u = 70$ mm & $d = 25$ mm (a) holes in square lattice and (b) interconnected pillars in square lattice with pitch size of around 210 ± 8 nm.

Figure 4.14 SEM images of square lattice features fabricated using two converging lens system with distance parameters changed to: $u = 40$ mm & $d = 25$ mm (a) resist pillars with average diameter of around 160 ± 10 nm in square lattice and (b) demagnified view showing pillars in large area.

As evident from Eq. (4.22), an increase in the distance u (distance between splitter and first lens) or an increase in split angle will result in lower pitch and higher resolution. To realize this, lenses with larger diameters to collect and converge the beams are required. As mentioned before, lower wavelength source can be employed to fabricate higher resolution features. But this requires deep UV lenses and a prism which can transmit DUV. This raises the cost of production. The current setup mainly uses optical elements that are generally used for visible wavelength and so is much cheaper.

4.4 Summary

In summary, two configurations for solid immersion lithography were investigated in this chapter. The first one using grating and mirrors is a technique, where the pitch is controlled by rotating the mirror assembly and thus controlling the angle at the base of the high index prism. High resolution features could be fabricated where feature size is limited by critical angle at the prism-index matching liquid interface. Sub-60 nm uniform periodic features with 110 ± 6 nm pitch were fabricated using this i-line laser based configuration. As the source used is of wavelength 364 nm, visible wavelength optical components and a high index prism (LAK31A) of angle 60° could be employed, thereby reducing the expenses compared to DUV source. The pitch size and resolution can be scaled down further using the same configuration by employing a suitable index matching liquid to increase the critical angle. The second configuration involves the use of multiple converging lenses for directing the beam on the

sample instead of mirrors. This is advantageous especially when numbers of beams involved are increased. For two and four-beam interference number of lenses used is the same (two lenses in this work). The number of optical components is half when compared to grating based four-beam configuration where four mirrors are required to guide the beams. Both single lens system and two lens system for interference lithography have been theoretically investigated. Square lattice features with pitch size of 210 ± 8 nm and 240 ± 6 nm were patterned using an i-line laser source and two lenses (biconvex lenses)-prism based system. The lack of quality of the features obtained here can be mainly attributed to the experimental errors that might have occurred during the sticking and removal of prism from the resist sample before and after exposure respectively. Even though prism based wavefront splitting and immersion techniques have been found to be very powerful techniques for two dimensional feature patterning with pitch size as small as 100 nm, the theoretical analysis was mainly focused on calculating the period of the periodic patterns fabricated. But, none of the interference study is complete without considering the influence of polarization. For example, in the experimental work involving two lens system, the four beams are TE-TM polarized which resulted in a pitch size of $\sqrt{2}p$ (where, $p = \lambda / 2n \sin \theta$). The polarization state of individual beams in multiple-beam interference has a major effect on the intensity of patterns produced. The range of structures that can be fabricated can be broadened by employing different polarization combinations for the beams employed for interference. In this context, next chapter discusses in detail the effect of

polarization in two-beam and four-beam interference. It also discusses the fabrication of 3D periodic photonic crystal structures employing five-beam interference.

Chapter 5: Effect of Polarization in Multiple-beam Interferometric Lithography

In the previous chapters, investigations have been done on two dimensional periodic feature patterning using prism based wavefront splitting and immersion interference lithographic techniques. An attractive method to broaden the range of structures that can be fabricated is to modify the intensity distribution by employing multiple beams with different polarization combinations. The polarization state of individual beams has a major effect on the interference patterns in multiple-beam interference lithography. In this chapter, emphasis is laid on analysing the effect of polarization, particularly linear and circular polarization in two-beam, four-beam and five-beam interferometric lithography, for producing 1D, 2D and 3D periodic structures.

5.1 Introduction

It has already been shown in the previous chapters that optical interference lithography could be successfully applied to fabricate periodic features in one and two dimensions. A one dimensional periodic fringe pattern could be formed by interfering two coherent waves in space. Such a 1D pattern has a sinusoidal intensity distribution. The same method could be extended to produce two dimensional periodic patterns by exposing the same material twice or thrice at different angles to a two-beam interference pattern. Double exposure after a 90° sample rotation yields 2D pattern with a square symmetry and triple exposure with a 120° sample rotation between exposures results in a hexagonal

lattice features [102, 103]. Another method for the fabrication of 2D and 3D periodic patterns is to employ a greater number of coherent beams for interference [98, 104]. Four coherent plane waves could be used to fabricate periodic patterns in a square symmetry where the polarization of individual beams has significant effect on the image contrast and period. Such multiple-beam interference techniques have been widely used in the fabrication of various 2D and 3D periodic micro structures for photonic crystal applications. Varying the number of beams, polarization and phase of individual beams result in various interesting 2D and 3D features [105-108]. In this context, this chapter initially focuses on analysing the effect of polarization on two-beam and four-beam interference in detail.

Various configurations involving multiple beams have been successfully employed to fabricate different types of three dimensional periodic structures with full photonic band gap [109-111]. A minimum of four beams is required for 3D periodic pattern generation by interference. Out of the various 3D periodic structures, the one with a diamond like lattice is of particular interest as it is reported to have a full band gap of around 25% of the centre frequency [112-116]. A common method used to fabricate a diamond like photonic crystal is to superpose two face centred cubic (FCC) or face centred tetragonal (FCT) interference patterns after shifting one pattern in space [113, 117]. However, it involves double exposures and displacement of either sample or exposing mask with respect to each other. The formation of body centred tetragonal, β -Tin type structure and diamond lattice structures by employing five-beam interferometric

methods with a circularly polarized central beam is investigated. Further, detailed theoretical analysis of five-beam interference and effect of polarization is analyzed to fabricate stand alone 3D periodic features.

5.2 Theoretical analysis

The electric field of a plane wave can be defined by [117, 118],

$$E_i(r, t) = E_i \cos [k_i \cdot r - \omega t + \varphi_i] \hat{e}_i \quad 5.1$$

Where, E_i is the amplitude, ω the frequency, k_i is the wave vector, φ_i is the phase, and $\hat{e}_i$ represents the unit polarization vector respectively. The unit polarization vector is real for linearly polarized light wave and is complex for circularly polarized wave.

The intensity distribution obtained by the interference of two or more coherent light waves can be expressed as [117],

$$I = \langle \sum_j E_i^2(r, t) \rangle \quad 5.2$$

$$+ \sum_{i < j} (E_i E_j) |\hat{e}_i \cdot \hat{e}_j^*| \cos [(k_i - k_j) \cdot r + (\varphi_i - \varphi_j) + \delta_{ij}]$$

Where, $\delta_{ij} = \arg\{\hat{e}_i \cdot \hat{e}_j^*\}$. Also note that $\hat{e}_j^*$ represent the conjugate of the unit polarization vector $\hat{e}_j$.

5.2.1 Two-beam interference

Two plane waves with different polarizations will interfere differently. For transverse electric (TE) polarization, the electric fields of the two vectors overlap completely whereas for other polarization combinations, the extent of overlap depends on the angle between the two waves. First let us consider an interference pattern generated by two waves originating from a common coherent source and converging in the x-z plane at equal angle ' θ ' with respect to the z axis as shown in Figure 5.1. The electric field vectors of the two waves can be defined, based on Eq. (5.1), as

$$E_1(r, t) = E_1 \hat{e}_1 \cos [(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_1] \quad 5.3$$

$$E_2(r, t) = E_2 \hat{e}_2 \cos [-(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_2]$$

Figure 5.1 Two-beam interference in x-z plane.

One dimensional periodic feature can be fabricated by recording the interference of two coherent waves. The total time-independent intensity distribution $I(r)$ at the intersection of the two waves is given by,

$$I(r) = \frac{(E_1^2 + E_2^2)}{2} + E_1 E_2 |\hat{e}_1 \cdot \hat{e}_2^*| \cos[(2k \sin \theta)x + (\varphi_1 - \varphi_2) + \delta_{12}] \quad 5.4$$

$$\text{Where,} \quad \delta_{12} = \arg\{\hat{e}_1 \cdot \hat{e}_2^*\}$$

Where, E_1 & E_2 are the amplitudes, φ_1 & φ_2 are the phase constants and $\hat{e}_1$ & $\hat{e}_2$ the unit polarization vectors of the two waves respectively. Here, k is the wave vector k and is given by: $k = 2\pi/\lambda$. The spatial-cosine term in intensity distribution in Eq. (5.4) describes a periodic pattern that varies along the x axis with a periodicity given by: $p = \lambda/2 \sin \theta$. The intensity contrast of the interference fringes varies depending on the polarization of the beams employed and is given by $V = |\hat{e}_1 \cdot \hat{e}_2^*|$ [119, 120]. For example, if the two beams, in this case are transverse electric (TE) polarized, the polarization vectors are given by,

$$\hat{e}_1 = \hat{e}_2 = \hat{y}$$

$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = 1$$

$$\Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = 1 \quad \& \quad \delta_{12} = \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

Now the intensity distribution defined by Eq. (5.4) becomes,

$$I(r)_{TE} = E_0^2 \left(\frac{1}{2} + \cos[k_0 2 \sin \theta x] \right); \quad \text{if } E_1 = E_2 = E_0 \quad 5.5$$

Thus the contrast in this case is 1, which means a TE-TE condition gives the maximum contrast for two-beam interference. The intensity distribution and contrast of fringe patterns are derived for different sets of polarization

combinations involving TE, TM, left circularly polarized (LC) and right circularly polarized (RC) beams and these are summarized in Table 5.1 for $E_0 = 1$ & $\varphi_1 - \varphi_2 = 0$. The derivation is given in detail in Appendix A.1.

Table 5.1 Contrast and intensity distribution for two-beam interference with various polarization combinations for $E_0 = 1$ & $\varphi_1 - \varphi_2 = 0$.

Polarization Combination	Intensity Distribution $I(r)$; $E_0 = 1$ & $\varphi_1 - \varphi_2 = 0$.	Contrast of the fringe pattern $V (\hat{e}_1 \cdot \hat{e}_2^*)$
TE-TE	$\left(\frac{1}{2} + \cos[k_0 2 \sin \theta x]\right)$	1
TM-TM	$\left(\frac{1}{2} + \cos 2\theta \cos[k_0 2 \sin \theta x]\right)$	$ \cos 2\theta $
TE-TM	1	0
TE-LC	$\left(\frac{1}{2} + \frac{1}{\sqrt{2}} \cos\left[k_0 2 \sin \theta x + \frac{\pi}{2}\right]\right)$	$\frac{1}{\sqrt{2}}$
TE-RC	$\left(\frac{1}{2} + \frac{1}{\sqrt{2}} \sin[k_0 2 \sin \theta x]\right)$	$\frac{1}{\sqrt{2}}$
TM-LC	$\left(\frac{1}{2} + \frac{ \cos 2\theta }{\sqrt{2}} \cos[k_0 2 \sin \theta x]\right)$	$\frac{ \cos 2\theta }{\sqrt{2}}$
TM-RC	$\left(\frac{1}{2} + \frac{ \cos 2\theta }{\sqrt{2}} \cos[k_0 2 \sin \theta x]\right)$	$\frac{ \cos 2\theta }{\sqrt{2}}$
LC-LC	$\left(\frac{1}{2} + \frac{(1 + \cos 2\theta) \cos[k_0 2 \sin \theta x]}{2}\right)$	$\frac{1 + \cos 2\theta}{2}$
RC-RC	$\left(\frac{1}{2} + \frac{(1 + \cos 2\theta) \cos[k_0 2 \sin \theta x]}{2}\right)$	$\frac{1 + \cos 2\theta}{2}$
LC-RC	$\left(\frac{1}{2} + \frac{(1 - \cos 2\theta) \cos[k_0 2 \sin \theta x]}{2}\right)$	$\frac{1 - \cos 2\theta}{2}$

The contrast of the fringe patterns varies for various polarization combinations. In case of TE polarized light, the contrast of interference

pattern(V) is unity because the electric fields of the two plane waves always overlap, regardless of angle between them. For the TM case however, the contrast depends on the angle between the waves and is given by $V = \cos 2\theta$. The pitch remains the same in both cases. From Table 5.1, it is clear that the TE-TM combination yields the minimum contrast of zero, which means that it doesn't interfere to produce fringe patterns. In all other cases, except for TE-TE, TE-LC and TE-RC, the contrast depends on the angle of incidence θ .

5.2.2 Four-beam interference

If the major effect of polarization in the case of interference of two beams is that it varies the image contrast, in four-beam interference it plays an even bigger role which will be analysed in detail here. Two dimensional periodic intensity distributions with square symmetry can be formed by interfering four coherent plane waves that converge at equal angles along two perpendicular planes. The electric field vectors of four perfectly aligned coherent beams, two incident at $\pm\theta$ to a plane (x-z plane) and the other two incident at $\pm\theta$ at an orthogonal plane (y-z plane), as shown in Figure 5.2 can be described by,

$$\begin{aligned}
 E_1(r, t) &= E_1 \hat{e}_1 \cos [(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_1] \\
 E_2(r, t) &= E_2 \hat{e}_2 \cos [-(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_2] \\
 E_3(r, t) &= E_3 \hat{e}_3 \cos [(k \sin \theta)y - (k \cos \theta)z - \omega t + \varphi_3] \\
 E_4(r, t) &= E_4 \hat{e}_4 \cos [-(k \sin \theta)y - (k \cos \theta)z - \omega t + \varphi_4]
 \end{aligned} \tag{5.6}$$

Where, k is the magnitude of propagation vector, ω is the angular frequency, $\hat{e}_i$ is the polarization vector and φ_i is the phase constant of each plane wave.

Figure 5.2 Four-beam interference configuration.

The time independent intensity distribution obtained by the interference of the four waves given in Eq. (5.6) is obtained by using Eq. (5.2) and is given by,

$$\begin{aligned}
 I(r) = & \frac{(E_1^2 + E_2^2 + E_3^2 + E_4^2)}{2} & 5.7 \\
 & + E_1 E_2 |\hat{e}_1 \cdot \hat{e}_2^*| \cos[k_0 2 \sin \theta x + (\varphi_1 - \varphi_2) + \delta_{12}] \\
 & + E_1 E_3 |\hat{e}_1 \cdot \hat{e}_3^*| \cos[k \sin \theta x - k \sin \theta y + (\varphi_1 - \varphi_3) + \delta_{13}] \\
 & + E_1 E_4 |\hat{e}_1 \cdot \hat{e}_4^*| \cos[k \sin \theta x + k \sin \theta y + (\varphi_1 - \varphi_4) + \delta_{14}] \\
 & + E_2 E_3 |\hat{e}_2 \cdot \hat{e}_3^*| \cos[-k \sin \theta x - k \sin \theta y + (\varphi_2 - \varphi_3) + \delta_{23}] \\
 & + E_2 E_4 |\hat{e}_2 \cdot \hat{e}_4^*| \cos[-k \sin \theta x + k \sin \theta y + (\varphi_2 - \varphi_4) + \delta_{24}] \\
 & + E_3 E_4 |\hat{e}_3 \cdot \hat{e}_4^*| \cos[k_0 2 \sin \theta y + (\varphi_3 - \varphi_4) + \delta_{34}]
 \end{aligned}$$

Now let us consider the cases where the wave pairs are either transverse electric (TE) or transverse magnetic (TM). If both pairs of plane waves in the configuration shown in Figure 5.2 are TE polarized, then the polarization vectors are defined by,

$$\hat{e}_1 = \hat{e}_2 = \hat{y} \quad \& \quad \hat{e}_3 = \hat{e}_4 = \hat{x} \quad 5.8$$

This scenario is equivalent to the case where two sets of two-beam interference patterns that intersect perpendicularly are added together, as the orthogonally polarized waves don't interfere. Here, it should be noted that the polarization vectors are real and not complex and so the argument value of function of $\hat{e}_i \cdot \hat{e}_j^*$ is zero.

$$\Rightarrow \delta_{12} = \delta_{13} = \delta_{14} = \delta_{23} = \delta_{24} = \delta_{34} = 0 \quad 5.9$$

Now calculating the magnitude function of $|\hat{e}_i \cdot \hat{e}_j^*|$,

$$\begin{aligned} |\hat{e}_1 \cdot \hat{e}_2^*| &= |\hat{y} \cdot \hat{y}| = 1 \\ |\hat{e}_1 \cdot \hat{e}_3^*| &= |\hat{e}_1 \cdot \hat{e}_4^*| = |\hat{e}_2 \cdot \hat{e}_3^*| = |\hat{e}_2 \cdot \hat{e}_4^*| = |\hat{y} \cdot \hat{x}| = 0 \\ |\hat{e}_3 \cdot \hat{e}_4^*| &= |\hat{x} \cdot \hat{x}| = 1 \end{aligned} \quad 5.10$$

And let, $E_1 = E_2 = E_3 = E_4 = E_0$.

The time independent intensity distribution in the case of four-beam interference, where both the beam pairs in Figure 5.2 are TE-TE polarized can be obtained by substituting the previously obtained values of the argument and magnitude of the function $\hat{e}_i \cdot \hat{e}_j^*$ in Eq. (5.7). After substitution and simplification, the intensity distribution can be expressed as,

$$I(r)_{TE-TE} = E_0^2 \{2 + \cos[k_0 2 \sin \theta x] + \cos[k_0 2 \sin \theta y]\} \quad 5.11$$

It is worth noting the z-independency of the intensity distribution. This means that the same intensity distribution is obtained on all planes parallel to

the x-y plane and that it can be imaged with infinite depth of focus in theory. But practically this is not the case and it is limited by power and coherence length of the source employed. Also in TE-TE case, any change in phase constant that might be caused by environmental disturbances only results in a shift of the patterns in the x-y plane.

Now in the second scenario, let us consider one pair of plane waves in Figure 5.2 to be TE polarized and the other pair to be TM polarized. The unit polarization vectors for the four beams in Eq. (5.6), in this case, are given by,

$$\hat{e}_1 = \hat{e}_2 = \hat{y} \quad , \quad 5.12$$

$$\hat{e}_3 = (\cos \theta \hat{y} + \sin \theta \hat{z}) \quad \& \quad \hat{e}_4 = (\cos \theta \hat{y} - \sin \theta \hat{z})$$

As in the previous case, the polarization vectors are real and not complex and so $\delta_{12} = \delta_{13} = \delta_{14} = \delta_{23} = \delta_{24} = \delta_{34} = 0$.

The magnitude value of the function $\hat{e}_i \cdot \hat{e}_j^*$ are given by,

$$|\hat{e}_1 \cdot \hat{e}_2^*| = |\hat{y} \cdot \hat{y}| = 1$$

$$|\hat{e}_1 \cdot \hat{e}_3^*| = |\hat{e}_2 \cdot \hat{e}_3^*| = |\hat{y} \cdot (\cos \theta \hat{y} + \sin \theta \hat{z})| = \cos \theta$$

$$|\hat{e}_1 \cdot \hat{e}_4^*| = |\hat{e}_2 \cdot \hat{e}_4^*| = |\hat{y} \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| = \cos \theta$$

$$|\hat{e}_3 \cdot \hat{e}_4^*| = |(\cos \theta \hat{y} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| = \cos 2\theta \quad 5.13$$

If all the waves has the same amplitude and are in phase or are out of phase by the same value, the intensity distribution is obtained by substituting the argument and magnitude values of the function $\hat{e}_i \cdot \hat{e}_j^*$ obtained previously in Eq. (5.7) and is given by,

$$I(r)_{TE-TM} = E_0^2 \{ 2 + \cos(2k \sin \theta x) + |\cos 2\theta| \cos(2k \sin \theta y) + 4|\cos \theta| \cos(k \sin \theta x) \cos(k \sin \theta y) \} \quad 5.14$$

In the third scenario, where all the plane waves shown in Figure 5.2 are TM polarized, the polarization vectors for the four beams mentioned in Eq. (5.6) are given by,

$$\begin{aligned} \hat{e}_1 &= (\cos \theta \hat{x} + \sin \theta \hat{z}); \quad \hat{e}_2 = (\cos \theta \hat{x} - \sin \theta \hat{z}); \\ \hat{e}_3 &= (\cos \theta \hat{y} + \sin \theta \hat{z}) \quad \& \quad \hat{e}_4 = (\cos \theta \hat{y} - \sin \theta \hat{z}) \end{aligned} \quad 5.15$$

Calculating the argument and magnitude of the function $(\hat{e}_i \cdot \hat{e}_j^*)$,

$$\begin{aligned} \delta_{12} &= \delta_{13} = \delta_{14} = \delta_{23} = \delta_{24} = \delta_{34} = 0 \\ |\hat{e}_1 \cdot \hat{e}_2^*| &= |(\cos \theta \hat{x} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{x} - \sin \theta \hat{z})| = \cos 2\theta \\ |\hat{e}_3 \cdot \hat{e}_4^*| &= |(\cos \theta \hat{y} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| = \cos 2\theta \\ |\hat{e}_1 \cdot \hat{e}_3^*| &= |(\cos \theta \hat{x} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} + \sin \theta \hat{z})| = \sin^2 \theta \\ |\hat{e}_2 \cdot \hat{e}_3^*| &= |(\cos \theta \hat{x} - \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} + \sin \theta \hat{z})| = \sin^2 \theta \\ |\hat{e}_1 \cdot \hat{e}_4^*| &= |(\cos \theta \hat{x} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| = \sin^2 \theta \\ |\hat{e}_2 \cdot \hat{e}_4^*| &= |(\cos \theta \hat{x} - \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| \\ &= \sin^2 \theta \end{aligned} \quad 5.16$$

And if all the waves have the same amplitude E_0 and are in phase or are out of phase by the same value, the intensity distribution is obtained by substituting the argument and magnitude values of the function $\hat{e}_i \cdot \hat{e}_j^*$ obtained previously in Eq. (5.7) and is given by Eq.(5.17),

$$\begin{aligned}
I(r)_{TM-TM} = E_0^2 \{ & 2 + |\cos 2\theta| \cos(2k \sin \theta x) \\
& + 4|\sin^2 \theta| \cos(k \sin \theta x) \cos(k \sin \theta y) \\
& + |\cos 2\theta| \cos(2k \sin \theta y) \}
\end{aligned} \tag{5.17}$$

In brief, Eq. (5.11), Eq. (5.14), and Eq. (5.17) gives the intensity distributions for four-beam interference for the cases TE-TE, TE-TM and TM-TM respectively when all the beams are in phase or out of phase by the same value and has the same amplitude. All the three cases can be used to produce two dimensional patterns with square symmetry. Following similar method as that involving TE-TE, TE-TM and TM-TM cases, the intensity distribution can be derived for different sets of polarization combinations involving TE, TM, left circularly polarized (LC) and right circularly polarized (RC) beams and these are given in Table 5.2. Derivation of intensity distribution for all these cases is shown in detail in Appendix A.2.

A 2D colour map of intensity for the combinations mentioned in Table 5.2, is plotted for comparison and is shown in Figure 5.3. The simulations are performed under the assumption that all the four interfering beams have unit amplitude and are in phase. All the combinations are seen to produce 2D patterns with square symmetry. However, it should be noted that only a TE-TE combination produces patterns with period of $p = \lambda/2 \sin \theta$, as shown in Figure 5.3 (a). Square lattice patterns obtained by all other combinations are seen to have a periodicity of $\sqrt{2}p$ and the lattice is oriented at 45° with respect to the two planes of incidence.

Table 5.2 Intensity distributions for four-beam interference with various polarization combinations.

Polarization combination	Intensity Distribution $I(r)$; when $E_0 = 1$ & $\varphi_1 - \varphi_2 = 0$.
TE-TE	$(2 + \cos[k_0 2 \sin \theta x] + \cos[k_0 2 \sin \theta y])$
TM-TM	$(2 + \cos 2\theta \cos(2k \sin \theta x) + 4 \sin^2 \theta \cos(k \sin \theta x) \cos(k \sin \theta y) + \cos 2\theta \cos(2k \sin \theta y))$
TE-TM	$(2 + \cos(2k \sin \theta x) + \cos 2\theta \cos(2k \sin \theta y) + 4 \cos \theta \cos(k \sin \theta x) \cos(k \sin \theta y))$
TE-LC	$\left(2 + 1 \cos[2k \sin \theta x] + \frac{ \cos \theta }{\sqrt{2}} \cos[k \sin \theta x - k \sin \theta y] + \frac{ \cos \theta }{\sqrt{2}} \cos[k \sin \theta x + k \sin \theta y] \right.$ $+ \frac{ \cos \theta }{\sqrt{2}} \cos[-k \sin \theta x - k \sin \theta y]$ $+ \frac{ \cos \theta }{\sqrt{2}} \cos[-k \sin \theta x + k \sin \theta y] + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y] \Bigg)$
TE-RC	$\left(2 + 1 \cos[2k \sin \theta x] + \frac{ \cos \theta }{\sqrt{2}} \cos[k \sin \theta x - k \sin \theta y] + \frac{ \cos \theta }{\sqrt{2}} \cos[k \sin \theta x + k \sin \theta y] \right.$ $+ \frac{ \cos \theta }{\sqrt{2}} \cos[-k \sin \theta x - k \sin \theta y]$ $+ \frac{ \cos \theta }{\sqrt{2}} \cos[-k \sin \theta x + k \sin \theta y] + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y] \Bigg)$
TM-LC	$\left(2 + \cos 2\theta \cos[2k \sin \theta x] + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[k \sin \theta x - k \sin \theta y + \frac{-\pi}{2} - \tan^{-1} \left(\frac{\sin^2 \theta}{\cos \theta} \right) \right] \right.$ $+ \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[k \sin \theta x + k \sin \theta y - \pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \right]$ $+ \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[-k \sin \theta x - k \sin \theta y - \pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \right]$ $+ \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[-k \sin \theta x + k \sin \theta y + \frac{-\pi}{2} - \tan^{-1} \left(\frac{\sin^2 \theta}{\cos \theta} \right) \right]$ $+ \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y] \Bigg)$

TM-RC	$ \begin{aligned} & \left(2 + \cos 2\theta \cos[2k \sin \theta x] + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[k \sin \theta x - k \sin \theta y + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \right] \right. \\ & \quad + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[k \sin \theta x + k \sin \theta y + \pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \right] \\ & \quad + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[-k \sin \theta x - k \sin \theta y + \pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \right] \\ & \quad + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[-k \sin \theta x + k \sin \theta y + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \right] \\ & \quad \left. + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y] \right) \end{aligned} $
LC-LC	$ \begin{aligned} & \left(2 + \left(\frac{1 + \cos 2\theta}{2} \right) \cos(2k \sin \theta x) + 4 \sin^2 \theta \cos(k \sin \theta x) \cos(k \sin \theta y) \right. \\ & \quad \left. + \left(\frac{1 + \cos 2\theta}{2} \right) \cos(2k \sin \theta y) \right) \end{aligned} $
RC-RC	$ \begin{aligned} & \left(2 + \left(\frac{1 + \cos 2\theta}{2} \right) \cos(2k \sin \theta x) + 4 \sin^2 \theta \cos(k \sin \theta x) \cos(k \sin \theta y) \right. \\ & \quad \left. + \left(\frac{1 + \cos 2\theta}{2} \right) \cos(2k \sin \theta y) \right) \end{aligned} $
LC-RC	$ \begin{aligned} & \left(2 + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta x] \right. \\ & \quad + \left(\frac{\sqrt{\sin^4 \theta + 4 \cos^2 \theta}}{2} \right) \cos \left[k \sin \theta x - k \sin \theta y + \tan^{-1} \left(\frac{2 \cos \theta}{\sin^2 \theta} \right) \right] \\ & \quad + \left(\frac{\sqrt{\sin^4 \theta + 4 \cos^2 \theta}}{2} \right) \cos \left[k \sin \theta x + k \sin \theta y + \pi + \tan^{-1} \left(\frac{2 \cos \theta}{\sin^2 \theta} \right) \right] \\ & \quad + \left(\frac{\sqrt{\sin^4 \theta + 4 \cos^2 \theta}}{2} \right) \cos \left[-k \sin \theta x - k \sin \theta y + \pi + \tan^{-1} \left(\frac{2 \cos \theta}{\sin^2 \theta} \right) \right] \\ & \quad + \left(\frac{\sqrt{\sin^4 \theta + 4 \cos^2 \theta}}{2} \right) \cos \left[-k \sin \theta x + k \sin \theta y + \tan^{-1} \left(\frac{2 \cos \theta}{\sin^2 \theta} \right) \right] \\ & \quad \left. + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y] \right) \end{aligned} $

Figure 5.3 2D colour maps of intensity distribution obtained by the interference of four beams with various combinations of polarizations given in Table 5.1. Here the incident angle is taken as $\theta = 45^\circ$.

However, it should be noted that in all the combinations, the intensity doesn't vary in the z-direction and thus it only yields a 2D periodic patterns. To obtain periodicity in the z-direction with k vectors of the four beams being the same as described in Eq. (5.6), another beam should be used. In this thesis, the intensity distribution has been analysed by employing a fifth beam which is circularly polarized propagating in z-direction and is discussed in the next section.

5.2.3 Five-beam interference with a circularly polarized central beam

Now let us consider the interference of five beams in an umbrella like configuration as shown in Figure 5.4, where the fifth beam is travelling in the z-direction. Note that the other four beams employed have the k vector as described by Eq. (5.6). The electric field vectors of the five beams, in this case, are given by,

$$\begin{aligned}
 E_1(r, t) &= E_1 \hat{e}_1 \cos [(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_1] \\
 E_2(r, t) &= E_2 \hat{e}_2 \cos [-(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_2] \\
 E_3(r, t) &= E_3 \hat{e}_3 \cos [(k \sin \theta)y - (k \cos \theta)z - \omega t + \varphi_3] \\
 E_4(r, t) &= E_4 \hat{e}_4 \cos [-(k \sin \theta)y - (k \cos \theta)z - \omega t + \varphi_4] \\
 E_5(r, t) &= E_5 \hat{e}_5 \cos [-kz - \omega t + \varphi_5]
 \end{aligned} \tag{5.18}$$

Figure 5.4 Five-beam umbrella like configuration.

And when the fifth beam is circularly polarized, the polarization vectors can be written as [118],

$$\hat{e}_{L5} = \frac{\hat{x} + i\hat{y}}{\sqrt{2}} \text{ (left circular beam); } \hat{e}_{R5} = \frac{\hat{x} - i\hat{y}}{\sqrt{2}} \text{ (right circular beam)} \quad 5.19$$

The total time independent intensity distribution, when the central beam is left circularly polarized and the four other beams are in TE-TM polarized state is given by,

$$\begin{aligned} I(r)_{5B \text{ TE-TM}} = & \frac{(E_1^2 + E_2^2 + E_3^2 + E_4^2 + E_5^2)}{2} \quad 5.20 \\ & + E_1 E_2 |\hat{e}_1 \cdot \hat{e}_2^*| \cos[2k \sin \theta x + (\varphi_1 - \varphi_2) + \delta_{12}] \\ & + E_1 E_3 |\hat{e}_1 \cdot \hat{e}_3^*| \cos[k \sin \theta x - k \sin \theta y + (\varphi_1 - \varphi_3) + \delta_{13}] \\ & + E_1 E_4 |\hat{e}_1 \cdot \hat{e}_4^*| \cos[k \sin \theta x + k \sin \theta y + (\varphi_1 - \varphi_4) + \delta_{14}] \\ & + E_1 E_5 |\hat{e}_1 \cdot \hat{e}_{L5}^*| \cos[k \sin \theta x - k \cos \theta z + kz + (\varphi_1 - \varphi_5) + \delta_{15}] \\ & + E_2 E_3 |\hat{e}_2 \cdot \hat{e}_3^*| \cos[-k \sin \theta x - k \sin \theta y + (\varphi_2 - \varphi_3) + \delta_{23}] \\ & + E_2 E_4 |\hat{e}_2 \cdot \hat{e}_4^*| \cos[-k \sin \theta x + k \sin \theta y + (\varphi_2 - \varphi_4) + \delta_{24}] \\ & + E_2 E_5 |\hat{e}_2 \cdot \hat{e}_{L5}^*| \cos[-k \sin \theta x - k \cos \theta z + kz + (\varphi_2 - \varphi_5) + \delta_{25}] \\ & + E_3 E_4 |\hat{e}_3 \cdot \hat{e}_4^*| \cos[2k \sin \theta y + (\varphi_3 - \varphi_4) + \delta_{34}] \\ & + E_3 E_5 |\hat{e}_3 \cdot \hat{e}_{L5}^*| \cos[k \sin \theta y - k \cos \theta z + kz + (\varphi_3 - \varphi_5) + \delta_{35}] \\ & + E_4 E_5 |\hat{e}_4 \cdot \hat{e}_{L5}^*| \cos[-k \sin \theta y - k \cos \theta z + kz + (\varphi_4 - \varphi_5) + \delta_{45}] \end{aligned}$$

From Eq. (5.12) and Eq. (5.19), $\hat{e}_1 = \hat{e}_2 = \hat{y}$, $\hat{e}_3 = (\cos \theta \hat{y} + \sin \theta \hat{z})$, $\hat{e}_4 = (\cos \theta \hat{y} - \sin \theta \hat{z})$ & $\hat{e}_{L5}^* = \frac{\hat{x} - i\hat{y}}{\sqrt{2}}$. Here, the argument, $\delta_{ij} = \arg\{\hat{e}_i \cdot \hat{e}_j^*\}$ is calculated as; $\delta_{12} = \delta_{13} = \delta_{14} = \delta_{23} = \delta_{24} = \delta_{34} = 0$.

$$\Rightarrow \delta_{15} = \delta_{25} = \arg\{\hat{e}_1 \cdot \hat{e}_{L5}^*\} = \arg\left\{\hat{y} \cdot \frac{\hat{x} - i\hat{y}}{\sqrt{2}}\right\} = \left(-\frac{\pi}{2}\right)$$

$$\Rightarrow \delta_{35} = \arg\{\hat{e}_3 \cdot \hat{e}_{L5}^*\} = \arg\left\{(\cos \theta \hat{y} + \sin \theta \hat{z}) \cdot \frac{\hat{x} - i\hat{y}}{\sqrt{2}}\right\} = \arg\left\{\frac{-i \cos \theta}{\sqrt{2}}\right\} = \left(-\frac{\pi}{2}\right)$$

$$\Rightarrow \delta_{45} = \arg\{\hat{e}_4 \cdot \hat{e}_{L5}^*\} = \arg\left\{(\cos \theta \hat{y} - \sin \theta \hat{z}) \cdot \frac{\hat{x} - i\hat{y}}{\sqrt{2}}\right\} = \arg\left\{\frac{-i \cos \theta}{\sqrt{2}}\right\} = \left(-\frac{\pi}{2}\right)$$

And in the similar way, if the central beam is right circularly polarized

$$\Rightarrow \delta_{15} = \delta_{25} = \delta_{35} = \delta_{45} = \left(\frac{\pi}{2}\right)$$

Now calculating the magnitude function of $|\hat{e}_i \cdot \hat{e}_j^*|$,

$$|\hat{e}_1 \cdot \hat{e}_2^*| = |\hat{y} \cdot \hat{y}| = 1$$

$$|\hat{e}_1 \cdot \hat{e}_3^*| = |\hat{e}_2 \cdot \hat{e}_3^*| = |\hat{y} \cdot (\cos \theta \hat{y} + \sin \theta \hat{z})| = \cos \theta$$

$$|\hat{e}_1 \cdot \hat{e}_4^*| = |\hat{e}_2 \cdot \hat{e}_4^*| = |\hat{y} \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| = \cos \theta$$

$$|\hat{e}_3 \cdot \hat{e}_4^*| = |(\cos \theta \hat{y} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| = \cos 2\theta$$

$$|\hat{e}_1 \cdot \hat{e}_{L5}^*| = |\hat{e}_2 \cdot \hat{e}_{L5}^*| = \left|\hat{y} \cdot \frac{(\hat{x} - i\hat{y})}{\sqrt{2}}\right| = \frac{1}{\sqrt{2}}$$

$$|\hat{e}_3 \cdot \hat{e}_{L5}^*| = \left|(\cos \theta \hat{y} + \sin \theta \hat{z}) \cdot \frac{(\hat{x} - i\hat{y})}{\sqrt{2}}\right| = \frac{\cos \theta}{\sqrt{2}}$$

$$|\hat{e}_4 \cdot \hat{e}_{L5}^*| = \left|(\cos \theta \hat{y} - \sin \theta \hat{z}) \cdot \frac{(\hat{x} - i\hat{y})}{\sqrt{2}}\right| = \frac{\cos \theta}{\sqrt{2}} \quad 5.21$$

Substituting the obtained argument and magnitude values of the function

$(\hat{e}_i \cdot \hat{e}_j^*)$ in Eq. (5.20), the time independent intensity distribution (where

the phase constant value is the same for all four plane waves) can be derived as,

$$\begin{aligned}
 I(r)_{5B\ TE-TM} = E_0^2 \left\{ \frac{5}{2} + \cos[2k \sin \theta x] \right. \\
 + 4 \cos \theta \cos[k \sin \theta x] \cos[k \sin \theta y] \\
 + \frac{2}{\sqrt{2}} \sin[-k \cos \theta z + kz] (\cos \theta \cos[k \sin \theta y] \\
 \left. + \cos[k \sin \theta x]) + \cos 2\theta \cos[2k \sin \theta y] \right\}
 \end{aligned} \tag{5.22}$$

This equation for intensity distribution holds true even if the central beam is right circularly polarized.

Now let us consider the case of five-beam interference where the two sets of orthogonal beams are TE-TE polarized and the central beam is left circularly polarized. The intensity distribution in this case, where all the beams has the same phase constant and amplitude, is expressed as,

$$\begin{aligned}
 I(r)_{5B\ TE-TE} = E_0^2 \left\{ \frac{5}{2} + \cos[2k \sin \theta x] + \cos[2k \sin \theta y] \right. \\
 + \frac{2}{\sqrt{2}} (\sin[-k \cos \theta z + kz]) (\cos[k \sin \theta x]) \\
 \left. + \frac{2}{\sqrt{2}} (\cos[-k \cos \theta z + kz]) (\cos[k \sin \theta y]) \right\}
 \end{aligned} \tag{5.23}$$

The derivation of the previous equation is given in Appendix A.3 and the equation is valid for the case where the central beam is right circularly polarized too.

Similarly, the intensity distribution in the case where the four plane waves are TM-TM polarized is given by,

$$\begin{aligned}
I(r)_{5B\,TM-TM} = E_0^2 \bigg\{ & \frac{5}{2} + \cos 2\theta (\cos[2k \sin \theta x] + \cos[2k \sin \theta y]) \\
& + 4 \sin^2 \theta (\cos[2k \sin \theta x])(\cos[2k \sin \theta y]) \\
& + \frac{2 \cos \theta}{\sqrt{2}} (\cos[kz - k \cos \theta z]) (\cos [k \sin \theta x]) \\
& + \frac{2 \cos \theta}{\sqrt{2}} (\sin[kz - k \cos \theta z]) (\cos [k \sin \theta y]) \bigg\}
\end{aligned} \tag{5.24}$$

The derivation is given in detail in Appendix A.3. Eq. (5.22), Eq. (5.23) and Eq. (5.24) describe the time independent intensity distributions obtained by employing a fifth circularly polarized central beam in z-direction, along with the two sets of orthogonal plane waves (in x-z and y-z planes). Here, all the beams are assumed to have the same phase constant value. This addition of a fifth beam will result in a variation of intensity in the z-direction and produces some interesting 3D patterns like body centred tetragonal lattice patterns and will be discussed in detail in a later section.

5.3 Experimental Methodology

The experimental set up for four-beam and five-beam interference lithography is shown in the Figure 5.5 . A spatially filtered, 364 nm CW laser beam is directed on to an electronic shutter with a temporal resolution of 10 ms, which controls the exposure time. A quarter wave plate follows the shutter, which converts the laser beam to a circularly polarized beam. This circularly polarized beam is directed perpendicularly onto a custom-fabricated cross-grating (Ibsen Photonics Pte Ltd), which diffracts the input beam into multiple beams. The cross-phase grating is designed to generate four first order beams and a zero-

order beam. A linear polarizer, mounted on a custom fabricated rotating mount, is placed in the optical path of each diffracted beam to obtain either an s-polarized or p-polarized beam. An optional hard stop is employed to block the zero-order beam so as to realize four-beam interference. The diffracted beams are directed such that they are about 45° with respect to the z -axis and they are made to fall on the resist, each at an angle of 35° with respect to z axis. The substrate used is a silicon wafer coated with positive resist AZ 9260. A $1\ \mu\text{m}$ thick layer of resist was coated using a spin coater at a speed of 4000 rpm for 30 sec. After exposure, the features patterned were developed in AZ400K developer, followed by a DI water rinse. Field emission scanning electron microscopy (FESEM) was employed to characterize the developed patterns. The results obtained using four and five beams interference are discussed in the next section.

Figure 5.5 Experimental setup for grating based multiple-beam interference lithography.

5.4 Results and Discussion

The intensity distribution in the case of the interference of two, four and five beams has been formulated for different polarization combinations involving linear and circular polarized states in previous sections. In the case of two-beam interference, it was observed that changes in the polarization state of the beams changes the contrast of the fringe pattern, as evident from Table 5.1. This in turn determines the contrast of the photoresist line features when recorded. In the case of four beams interference, the polarization changes results in square lattice features with different periodicity and orientation. The intensity distributions for the various sets of polarization combinations where the two sets of beams are in two orthogonal planes, incident at equal angle with respect to z axis, were formulated and documented in Table 5.2.

For experimental verification, four-beam interference with three different polarization conditions are considered: TE-TE, TE-TM and TM-TM, whose intensity distributions are given by equations, Eq. (5.11), Eq. (5.14), and Eq. (5.17) respectively, where the phase constant is considered zero or same for all waves. The 2D and 3D colour map of intensity distribution for TE-TE pattern for $\theta = 30^\circ$ defined by Eq. (5.11) are shown in Figure 5.6. Figure 5.6(a) gives the top view of the TE-TE pattern, clearly showing the square symmetry of the patterns with period $p = \lambda/2 \sin \theta = 364nm$. Figure 5.6(b) and Figure 5.6(c) shows the slanted and sliced 3D view of the intensity distribution, clearly showing the independency of intensity on z direction. Three major intensity

regions are identified here: High intensity (H), low intensity (L) and medium intensity (M) regions, which becomes important when the patterns are transferred to a resist. As evident from Eq. (5.11), the maximum intensity obtained in TE-TE interference is $I_{Max(TE-TE)} = 4E_0^2$. Here, since the amplitude is taken as unity the maximum intensity equals 4 as shown in the colour bar in Figure 5.6(a). In the experimental work, positive photoresist (AZ9260) is employed along with a 364 nm laser source, as shown in Figure 5.5. The SEM images of the obtained resist patterns are shown in Figure 5.6(d) and Figure 5.6(e) and the obtained square lattice periodicity is around 359 ± 17 nm. At lower exposure doses, the region M remains along with region L, as evident from Figure 5.6(d), whereas region H, being a high intensity region is washed out. And when the exposure dose is further increased, only L region remains producing distinct pillar structures in a square lattice as shown in Figure 5.6(e).

Figure 5.6 Simulation and experimental results of four-beam interference with beams TE-TE polarized. (a) Top view, (b) slanted view and (c) sliced view of the intensity distribution given by Eq. (5.11). And SEM images of the fabricated pattern in resist: (d) Top view of interconnected patterns and (e) top view of distinct resist pillars in square lattice with periodicity 359 ± 17 nm.

A colour map of the intensity distribution, in the case of four-beam interference with TE-TM polarized beams for $\theta = 30^\circ$, as defined by Eq. (5.14), along with the experimentally obtained results is shown in Figure 5.7. As evident from the 2D colour map in Figure 5.7(a), a square lattice in the TE-TM pattern is oriented at 45° with respect to the two planes of incidence (x and y axis in this case). Also the maximum intensity of the TE-TM distribution is given by $I_{Max(TE-TM)} = 2E_0^2(1 + \cos \theta)^2$ which is higher ($\theta \approx 90^\circ$, $I_{Max(TE-TM)} \approx 8E_0^2$) when compared to the TE-TE case which is given by $I_{Max(TE-TE)} = 4E_0^2$. For example, the maximum intensity is $\cong 6.9$, corresponding to $\theta = 30^\circ$. This is much higher than that obtained for TE-TE pattern. Moreover the middle region of intensity, mentioned in the TE-TE case, can be ignored here as the intensity at these saddle points between maxima and minima is very weak and can be included in the minima region. Figure 5.7(b) and Figure 5.7(c) gives the slanted and sliced view of the intensity distribution, confirming that the TE-TM pattern too has the same distribution all along the z direction. Figure 5.7(d) and Figure 5.7(e) shows the SEM images of the resist patterns fabricated using TE-TM polarized beams via four-beam interference. The image shows holes in a square lattice with a period of 477 ± 10 nm, corresponding to $\sqrt{2}p$ (507.7 nm when $p=359$ nm). Holes have an average diameter of around 233 ± 21 nm and correspond to high (H) intensity region mentioned in Figure 5.7(a). So it can said that when a positive resist is employed, the TE-TE condition is better suited to fabricate pillars in a square lattice and TE-TM is better suited to produce holes in a square lattice.

Figure 5.7 Simulation and experimental results of four-beam interference with beams TE-TM polarized. (a) Top view, (b) slanted view and (c) sliced view of intensity distribution given by Eq. (5.14). And SEM images of the fabricated holes in square lattice of periodicity 477 ± 10 nm ($\sqrt{2}p$): (d) magnified top view and (e) de-magnified top view.

Figure 5.8 shows the colour map and experimental results of patterns obtained by four-beam interference with TM-TM polarized beams for $\theta = 30^\circ$ as defined by Eq. (5.17). The 2D and 3D simulation results, assuming unit amplitude and zero phase for all beams, are plotted in Figure 5.8(a), Figure 5.8(b) and Figure 5.8(c). The slanted and sliced view shows that the intensity doesn't vary in the z direction, as in the other two polarization cases discussed before. The 2D colour map in Figure 5.8(a) and SEM image of fabricated pattern in Figure 5.8(b) clearly shows the three different intensity regions namely: high (H), low (L) and medium (M). An increase in exposure dose is required to

completely wash out the resist in the medium region to obtain just pillars corresponding to low region in the square lattice. The periodicity of such a square lattice would be p , as clearly evident from the lines connecting low regions (L) in Figure 5.8(a). Also such pillars will not have a perfect shape and will resemble the shape in the L region in Figure 5.8(e). Employing a negative resist will yield pillars corresponding to high (H) region in Figure 5.8(a). Such pillars will have more or less a perfect circular shape and will be in a square lattice as marked by the dotted slanted square in Figure 5.8(a), and will have periodicity defined by $p = \sqrt{2}\lambda/2 \sin \theta$. Thus, using the same set up and varying polarizations, different patterns can be recorded on photoresist. However, interference of four beams which are incident at equal angles in two orthogonal planes results only in a 2D periodic pattern. The intensity doesn't vary in the third dimension. This could be remedied by carefully varying the plane and angle of incidence of individual beams. However, in this thesis, focus was laid on achieving periodicity in the third dimension by employing a fifth beam to the two sets of beams in orthogonal planes.

Figure 5.8 Simulation and experimental results of four-beam interference with beams TM-TM polarized. (a) Top view, (b) slanted view and (c) sliced view of intensity distribution given by Eq. (5.17). And (d) Top view of SEM image of the fabricated pattern in resist.

Five-beam interference - Simulation results:

The addition of a fifth beam which is circularly polarized to the four beams in orthogonal planes produces some interesting 3D structures. Three cases of five-beam interference are discussed in this section: central circular beam with the other two beam pairs TE-TM polarized, TE-TE polarized and TM-TM polarized. The 3D colour maps of intensity distribution given by Eq. (5.22), Eq. (5.23) and Eq. (5.24), corresponding to these three cases of five-beam interference is shown in Figure 5.9.

Figure 5.9 (a) Beam arrangement for realizing five-beam interference. (b) 3D colour map of five-beam interference distribution with circularly polarized central beam (LC/RC) and the other two beam pairs TE-TM polarized, (c) E_5 – LC/RC; E_1, E_2, E_3 & E_4 – all TE polarized and (d) E_5 – LC/RC; E_1, E_2, E_3 & E_4 – all TM polarized.

The combination of four coherent plane waves (with equal amplitudes), that converge at equal angle θ along two orthogonal planes, were observed to produce a 2D square lattice patterns as evident from Figure 5.3. When one set of plane waves is TE polarized and the other set is TM polarized, the interference

pattern obtained has a square symmetry and is shown in Figure 5.3(b). An addition of a fifth circularly polarized (either left circularly polarized or right circularly polarized) beam of equal amplitude that propagates in the z-direction to this combination produces patterns with an additional periodicity in the z-direction, as evident from Figure 5.9(a). This intensity pattern yield different structures when recorded, by applying a different intensity threshold value as evident from Figure 5.10. Here, for the intensity distribution plot in Figure 5.9(b), if the intensity cut off value is set to 2, dumbbell shaped patterns can be recorded, as shown in Figure 5.10(a) and Figure 5.10(b). And when this cut off value is increased to 6, body centred tetragonal (BCT) structures as shown in Figure 5.10(c) and Figure 5.10(d) can be recorded using a negative photoresist. Experiment performed using this five-beam interference condition using positive photoresist and the setup shown in Figure 5.5, was observed to produce features resembling that of the inverse of pattern shown in Figure 5.10(a). The SEM images obtained are shown in Figure 5.11. The top view shows that the period is almost close to $\sqrt{2} \lambda / 2 \sin \theta \cong 488 \text{ nm}$. The areas denoted by black circle shows curved resist features corresponding to the curved pattern of the dumbbell shaped pattern obtained in simulation. Reflection from the back surface is found to have a large effect, as evident from the periodic patterns on each pillar.

Figure 5.10 (a), (b): Dumbbell shaped 3D pattern obtained by five-beam interference defined by Eq. (5.22) with intensity cut off value of 2. (c) and (d): body centred tetragonal structure formed by increasing the intensity cut off value to 6.

Figure 5.11 SEM images of five-beam interference patterns recorded in a positive photoresist using a 364 nm laser source, where the central beam is circularly polarized and the four plane waves are in TE-TM polarized state (a) top view and (b), (c) and (d) slanted view. Some areas are marked using black circle to show the resemblance of the structure to that of inverse pattern of the feature shown in Figure 5.10(a).

Similarly, a 3D intensity distribution is plotted for five-beam interference where all the plane waves are TM polarized described by Eq. (5.24). The distribution shown in Figure 5.9(d) is similar to that of TE-TM polarized case which is shown in Figure 5.9(b), except for the large value of intensity at the saddle points. So with a smaller intensity cut off value, the structures obtained will resemble dumbbell shaped pattern, as in the case of TE-TM and with a

higher cut off value, it resembles a body centred tetragonal lattice and is shown in Figure 5.12.

Figure 5.12 (a) Intensity distribution obtained by five-beam interference, where the central beam is circularly polarized and the other four beams are TM polarized, (b) interconnected structure obtained by applying intensity cut off value of 4 and (c) body centred tetragonal structure obtained by increasing intensity cut off.

Finally, if the beams E_1, E_2, E_3 & E_4 described in Figure 5.9(a) are TE polarized and the central beam E_5 is either left circularly or right circularly polarized, structures resembling a diamond lattice can be fabricated. This is shown in Figure 5.13, where the intensity distribution described by Eq. (5.23) is plotted by applying intensity cut off value. The structure obtained is a β -Tin type structure. This structure is actually a body centred tetragonal structure with two atoms per lattice point. It has a periodicity of $a = \lambda / \sin \theta$ in the x and y directions and a periodicity of $c = \lambda / (1 - \cos \theta)$ in the z direction. Here, the ratio ‘c/a’ can be varied to convert the β -tin type structure to a diamond structure. A diamond structure is usually obtained by using five-beam interference by the addition of two face centred cubic pattern with a little

displacement (quarter of the period). This kind of diamond like structure is reported to have a full photonic band gap of around 25% [121]. Here, the ratio c/a can be varied to convert the β -tin type structure to a diamond structure. When the $c = \sqrt{2}a$, the β -tin type structure gets transformed to a diamond structure and the transformation is shown in Figure 5.14(a). An unit cell of β -tin type structure embodied in between two adjacent diamond unit cells is clearly shown in Figure 5.14(a). The corner atoms of the cubic diamond unit cells are connected by white solid lines and that of β -tin type structure are connected by red dashed lines. When $c \neq \sqrt{2}a$, the unit cell inside the white solid lines is no longer cubic in nature and so the structure cannot be termed a diamond cubic structure. In the case of five-beam interference with circularly polarized central beam and four TE polarized beams incident at two orthogonal planes, diamond type structure can be patterned only when all the plane waves are incident at equal angle of $\theta = 70.523^\circ$ with respect to the normal. Only at this angle of incidence, the criteria of $c = \sqrt{2}a$ is satisfied. The resulting diamond type pattern obtained by plotting Eq. (5.23) using an angle of incidence of $\theta = 70.523^\circ$ and by applying an intensity cut off value of 5.5 is shown in Figure 5.15. Figure 5.15(a) shows the side view of the obtained diamond type pattern similar to that shown in the unit cell in Figure 5.14(a). And Figure 5.15(b) shows the view of diamond type structure in $[111]$ plane which is similar to $[111]$ plane view of diamond unit cell as shown in Figure 5.14(b).

Figure 5.13 (a) Intensity distribution obtained by five-beam interference for an angle of incidence $\theta = 45^\circ$, where the central beam is circularly polarized and the other four beams are TE polarized, (b) interconnected tetragonal structure obtained by applying intensity cut off value of 3 and (c) β -tin type structure obtained by increasing intensity cut off value.

Figure 5.14 (a) β -tin type structures to diamond cubic structure transformation. Unit cell of β -tin type structure is shown by blue atoms inside tetragonal lattice marked by red dashed line and two adjacent unit cells of diamond lattice is shown inside cubic lattices connected by white solid line. (b) $[111]$ plane view of diamond unit cell.

Figure 5.15 Diamond like patterns produced by five-beam interference by satisfying the condition $c/a=\sqrt{2}$ for β -tin type structure: (a) slanted view and (b) view in $[111]$ plane for comparison with that in Figure 5.14(b).

5.5 Summary

In this chapter, the effect of polarization on two-beam, four-beam and five-beam interference pattern has been studied in detail. In two-beam interference, polarization of individual beams has a major effect only in contrast, whereas four-beam and five-beam interference pattern is found to be much more sensitive to polarization. A detailed analysis of the intensity distributions for various cases of four-beam interference, where the beams are converged at equal angles in two orthogonal planes has been performed for different polarization combinations. It was observed that all the cases produces pattern with a square symmetry with a period of $\sqrt{2}\lambda/2\sin\theta$, except when all beams are TE polarized where the period of square lattice was found to be $\lambda/2\sin\theta$. Experimental verification was done for three cases: TE-TE, TE-TM and TM-TM by recording the patterns in a positive photoresist. Later, circularly polarized beam was

employed along with four plane waves in orthogonal planes to produce 3D patterns. It was found that body centred tetragonal structures could be obtained by five-beam interference with a circularly polarized central beam. It was also found that if the four plane waves in orthogonal planes in five-beam interference are TE polarized, β -tin type and diamond like structures, which have interesting photonic band gap properties, can be realized.

Chapter 6: Enhanced SPP Propagation and Light Trapping Applications using Sub-wavelength Periodic Structures

In the previous chapters, various optical lithographic methods for periodic feature patterning have been discussed in detail. Two dimensional features were successfully fabricated using various simple techniques. The methodology to fabricate 3D complete band gap photonic crystal structures were also discussed in detail. These techniques can cover a large range of periodicity starting right from 100 nm to 1 μm . Controlling the periodicity of structures at the nanoscale in multiple dimensions enables the manipulation of the optical response of materials for plasmonic applications, such as enhanced field concentration and plasmonic light trapping. In this chapter, two major applications, namely enhanced transmission through 2D metallic grating and light trapping in silicon based solar cell, are investigated.

6.1 2-D Periodic Perforated Metal array for Dye Based Surface Plasmon Lithography

6.1.1 Introduction

Near field surface plasmon assisted optical lithography using periodic metallic masks can be used to fabricate high density features with resolution scaling well beyond the diffraction limit. But, transmission through the apertures is very small due to the damping of surface Plasmon (SP) because of scattering

from the rough metallic surfaces. This in turn causes propagation loss on top of the inherent loss resulting from the complex dielectric constant of the metal. This limits the effective penetration depth through the apertures, thereby creating an obvious obstacle in utilizing them in practical optical devices and circuits. Here an attempt is made to decrease the SPP propagation loss by employing a gain medium around the periodic mask structure. The concept and methodology for dye based plasmon enhancement in embedded mask based near field lithographic configuration for high aspect ratio feature fabrication is proposed. The effect of dye medium on the total field transmission through sub-50 nm features of the metal mask is explored numerically. Also, the design aspects such as variation in thickness, diameter and type of dye material are explored in order to maximize the transmission.

6.1.2 Methodology: Proposed configuration

The schematic diagram (Figure 6.1) shows the proposed configuration of the plasmonic mask used in this work. This embedded-amplitude mask consists of an aluminium layer perforated with 2D biperiodic circular arrays. The Al film is covered on either side by two dielectric layers with almost matching dielectric constant to reduce surface plasmon-polariton (SPP) propagation losses which could arise due to dielectric mismatch. Polymethylmethacrylate (PMMA) is selected as one of the dielectric layers as it has almost matching dielectric constant with that of the negative near-UV photoresist used in the present study ($n=1.7$). In addition PMMA can be easily doped by the dye molecules of interest

and it has good transmission in the UV regime. Two types of plasmonic samples: one with bare PMMA film and the other with dye embedded PMMA are employed to investigate the E-field transmission enhancement as shown in Figure 6.1. Finally, an index matching liquid is employed as a spacer layer between the metal film and the negative near-UV photoresist to eliminate the gap variation between the mask and the photoresist in the lithography process.

Figure 6.1 Schematic diagram of the proposed configuration.

6.1.3 Experimental Issues and the Importance of Optical Lithography

To realize the proposed configuration, a metallic mask needs to be fabricated on a dye doped PMMA substrate. This could then be used together with index matching liquid layer to realize SP lithography. Once the dye doped PMMA substrate is prepared, patterning needs to be carried out by suitable lithographic process to write periodic feature, after which a metal layer could be deposited. Since the required mask periodicity is in the range of 170 nm, one of the proposed optical lithographic configurations discussed in previous chapter

could be employed. It should be noted that alternatives like x-ray or electron beam lithography cannot be employed here as PMMA is sensitive to x-ray and electron beam radiation. Moreover, the exposure of Dye doped PMMA to such radiations will nullify the dye fluorescence property. Thus for sensitive applications like the one discussed here, optical lithographic techniques discussed in this thesis plays crucial role.

6.1.4 Numerical Simulation: Results and discussion

Surface plasmon enhancement in a perforated Al mask is numerically demonstrated using the finite-difference-time-domain (FDTD) method. The commercially available FDTD from Lumerical solutions is employed for all simulations. Different set of simulations were carried out in order to determine the transmission wavelength for aluminium, to optimize the aluminium thickness, to study transmission modes and finally to compare the E-field transmission through perforated masks with and without a dye medium. All the simulations used periodic boundary condition along the x and z-directions to emulate an infinite biperiodic structure. A perfectly matched layer (PML) is considered along the y-direction.

Initially, analysis was carried out to determine the period, thickness and wavelength for the aluminium mask to be used here. High transmission, at around 365 nm wavelength, was observed for aluminium structures with periodic holes of diameter 40 nm, at a pitch size of 170 nm and with thickness of 100 nm. Considering this, and the fact that an i-line laser is commonly available, 365 nm

was employed as an excitation wavelength. As well as 365 nm, transmissions at two other wavelengths, namely 435 nm and 485 nm, were also observed for this structure. The transmission spectrum exhibits a prominent peak at 485 nm. The transmission spectra of such arrays are characterized by resonances which are related to a (i, j) scattering order of the array. The wavelength of the transmission peak can be given approximately by the surface plasmon dispersion for a smooth film [76].

$$\lambda_{(i,j)} = \frac{p}{\sqrt{i^2 + j^2}} \sqrt{\frac{\epsilon_m \cdot \epsilon_d}{\epsilon_m + \epsilon_d}} \quad 6.1$$

Where ϵ_m and ϵ_d are respectively the dielectric constants of the metal and dielectric (air), P the period of the array and (i, j) represents the scattering order of the array. The peak at 365 nm corresponds to the second harmonic of TE (11) mode.

A perforated plasmonic sample will have a low transmission as it suffers from SP propagation losses associated with the metal. The damping of the SP is because of contributions due to dissipation in the metal and to radiative losses. This loss can overcome by gain medium such as a dye with optimum concentration [122-124]. A PMMA matrix doped with dye molecules is considered as the gain medium. The dye molecules are chosen to have large absorption and emission cross-sections at the excitation and emission wavelengths of interest. Besides this, the emission band width should have the wave component of the 485 resonance mode. Coumarin 102 is selected for the

present simulation as it has an emission band around 485 nm for an excitation at 365 nm wavelength [125-127]. The nature of the SP enhancement is studied by placing a radiating coumarin 102 dye near a metal nanoparticle. The simulation was done by considering an electric dipole to mimic a Coumarin 102 dye molecule, which has an emission at 485 nm. This emission wavelength is exactly equal to the wavelength of the dominant 485 nm resonance mode.

Figure 6.2 (a) Electric field distribution, and (b) its magnitude at the exit plane of the perforated metal mask of a type I configuration (Figure 6.1 (a)) which employs no gain medium. The thickness, size and period of the arrays employed are 100 nm, 40 nm and 170 nm respectively. (c) Electric field distribution, and (d) its magnitude at the exit plane of the perforated metal mask, type II configuration (Figure 6.1 (b)), in the presence of gain medium (PMMA doped with coumarin 102 dye).

Figure 6.2(a) and Figure 6.2(b) represents the electric field distribution and its magnitude at the exit plane of the perforated metal mask of type I shown in Figure 6.1(a), which employs no gain medium. It represents the normalized field with respect to an input excitation plane wave of wavelength 365 nm. The thickness, size and period of the arrays are 100 nm, 40 nm and 170 nm respectively. Figure 6.2(c) and Figure 6.2(d) represents the electric field distribution and its magnitude at the exit medium of the perforated metal mask, in the presence of again medium (dye) using the same excitation wavelength. An extraordinary transmission with about a 14.5 fold enhancement is observed due to the transfer of light from the 485 nm resonance mode excited inside the cavity.

Studies have also been carried out to compare the transmission through the perforated film due to excitation using a (1) dipole and a 365 nm source and (2) two 365 nm and 485 nm direct light source. The excitation at 365 nm, in the presence of the dipole, shows a much higher transmission compared to the latter, as evident from results plotted in Figure 6.3. This could be due to the strong fluorescence quenching as a consequence of the emission of plasmons directly to the metal film by the dye molecule [125-127]. The dye molecule, in the presence of the metal film, will be influenced by four decay channels [128, 129] that arise due to the coupling of the dipole to (i) long range surface plasmon polariton and (ii) short range surface plasmon polariton modes — in both cases, the dipole excites an SPP instead of a photon emission, (iii) coupling to electron-hole (E-H) pairs in the metal film by dipole-dipole interaction, and (iv) coupling to the

radiation modes of the structure. The enhancement of the cavity modes can be attributed to the strong interaction between the excited dye molecules and the electron vibration energy of the SP. In this case, the excited molecule more readily relaxes than the radiative relaxation processes. This could be due to large electromagnetic fields introduced by the density of states, which resulted in the emission of a SP instead of a photon. A detailed discussion of this work is given in Appendix B.

Figure 6.3 The transmission of the perforated film due to (1) a dipole and 365 nm source and (2) both 365 and 485 nm excitation.

6.2 Enhanced light trapping in Silicon based Solar cells: Periodic Structures for light trapping

Photovoltaic's based on thin absorbing layer have invoked tremendous interest over the past decade as a suitable solution for energy problems in future. Light trapping plays an important role in increasing the efficiencies of thin film solar cell by enhancing the optical absorption inside the thin active layer. In silicon based solar cells, proper light trapping techniques enables a reduction in the film thickness from the 150-200 μm range to $<2 \mu\text{m}$, without compromising the conversion efficiency. This helps in the reduction of material and hence a corresponding reduction in the production cost.

In this report, the usage of periodic structures for light trapping, both in the front and back surface of the active silicon layer have been investigated. The fabrication of the periodic structures can be carried out using the lithographic techniques discussed in previous chapters in a simple way.

6.2.1 Periodic Metal back reflectors

Initially, three thin film solar cell configurations, as shown in Figure 6.4, were considered for simulation. Figure 6.4(a) shows the bare silicon configuration with a silicon active layer of 250 nm uses just ITO as electrodes. The second configuration shown in Figure 6.4(b), uses a 50 nm planar aluminium layer as back reflector. Finally, the third type shown in Figure 6.4(c) uses an aluminium grating as the back reflector. The Lumerical FDTD package was used

to carry out simulations to calculate the absorption inside the active silicon layer, the basic principle of which is based on YEE lattice [130]. A perfectly matched layer on the horizontal axis and periodic layer on the vertical axis was applied as boundary conditions during simulation. To calculate the power absorbed in the bare silicon substrate, two power monitors were used: one at the top of the silicon and the other at the bottom surface. The amount of power transmitted through each surface normalized to the source power was then calculated, where transmission is given by the following formula,

$$T(f) = \frac{\frac{1}{2} \int \text{real}(P(f)) \cdot dS}{\text{source power}} \quad 6.2$$

where, $T(f)$ is the normalized transmission, $P(f)$ is the Poynting vector and dS is the surface normal. The absorbed power $P_{\text{abs}}(\lambda)$ is obtained by subtracting the transmitted power through each monitors as given by,

$$P_{\text{abs}}(\lambda) = -\text{transmission}(\text{monitor}_{\text{top}}) - -(\text{transmission}(\text{monitor}_{\text{bottom}})) \quad 6.3$$

In the case of a periodic back grating, the power monitors were employed on top and bottom of each section of the silicon active layer and the total absorption was later calculated by adding up the obtained values for each section. Note that for the third case using a grating back reflector, a 100 nm Al grating with half pitch of 50 nm and thickness of 50 nm was used in simulations.

Figure 6.4 Three thin film solar cell configurations considered for simulation
 (a) Bare silicon cell with ITO electrodes, (b) Silicon cell with planar aluminium back reflector layer and (c) Silicon thin film cell with metal (Al) grating back reflector.

The measured absorbed power through FDTD is normalized with respect to the input power and is plotted against the incident wavelength for all three configurations shown in Figure 6.4 and is shown in Figure 6.5. As expected, the absorption for bare silicon with ITO is very low above 800 nm wavelength. This is improved by using Al back reflectors, as is evident from the other two curves in Figure 6.5. When planar Al layer is added to the configuration, extra peaks appear above 800 nm corresponding to bulk plasmon excitation and this improves the absorption efficiency. The efficiency is improved further by using an Al grating along with a planar layer at the back surface, as shown by dotted brown curve in Figure 6.5. Broader and higher peaks are obtained in the IR range because of the surface plasmon polariton and the introduction of a Fabry Perot resonance. Here, the grating back surface couples light into the SPP modes that are supported at the metal-semiconductor interface as well as the guided modes in semiconductor slab. It was observed that for the wavelength band of 400 nm-1200 nm absorption was improved by a factor of 28% by

employing a planar back reflector instead of ITO. For the same wavelength range, an improvement of 12% in absorption was observed for the thin film solar cell based on a grating back reflector when compared to that based on a planar Al back layer. Thus, using a periodic metal back reflector layer was seen to improve the absorption efficiency greatly even for a simple grating based configuration. This can be further improved by using 2-D and 3-D metal back structures where additional controlled absorption due to the photonic band gap of these structures can be introduced.

Figure 6.5 Variation of normalized absorption with incident wavelength for the three configurations shown in Figure 6.4.

6.2.2 Periodic Metal back reflectors and front side grating

The simulations in the previous section have clearly demonstrated that metal grating back structures clearly improves the absorption efficiency of silicon thin film solar cell by a sizable margin. In this section, simulations are made to investigate, the effect that grating the front surface has on absorption. Here, two new configurations, as shown in Figure 6.6 are introduced, both with silicon front surface grating and metal back surface grating. In the first structure the front surface grating period was set as 100 nm, with 50 nm half pitch size and in the second the period was set at 500 nm (250 nm half pitch). Silicon grating height was set to 50 nm in both the above mentioned structures.

Figure 6.6 Silicon thin film solar cell configuration with metal (Al) grating (Period:100 nm) back reflector & silicon grating front surface:(a) Front side grating with period:100 nm;(b) Front surface grating with period:500 nm.

The calculated normal absorption curves for the wavelength range of 400 nm to 1200 nm is plotted in Figure 6.7, along with that for just the metal grating back reflector. For all three cases, an aluminium grating back reflector with a period of 100 nm and half pitch size of 50 nm was used. The introduction of a grating at the front surface increases the path length of light inside the active silicon layer by means of scattering and hence increases the absorption. It is clear from the plot in Figure 6.7 that in both the cases, using a grating on the front surface, the bulk plasmonic peak observed in the infra-red range is reduced. This is because scattering reduces the amount of normal incidence on the back metal grating. However, a comparison of the absorption efficiency shows that a front surface grating improves the absorption. An improvement of 7.21% in absorption, compared to that with Al grating back reflector, was observed when a silicon grating of 100 nm period was used at the front surface. Comparing with the configuration using just a planar metal back layer, the improvement was calculated to be 20%. However, for a front grating with periodicity of 500 nm, the absorption decreases slightly by 3% when compared to the 100 nm period case. The decrease can be attributed to a smaller amount of scattering due to the larger periodicity. Thus, it can be concluded that a thin film solar cell with a grating at the front and back layer, of sub-wavelength periodicity, improves the light trapping and hence absorption considerably by introducing plasmonic modes, guided modes, and scattering effects.

Figure 6.7 Variation of normalized absorption with incident wavelength for the three configurations: (1) silicon with an Al back reflector, (2) silicon with an Al back grating and Si front grating of period 100 nm & (3) silicon with an Al back grating and silicon front grating of period 500 nm.

6.2.3 Periodic Metal grating–Sphere system in silicon for enhanced light trapping by the Gap mode effect

As mentioned earlier, plasmonic concepts have been widely used in thin film solar cells to achieve enhanced light localization. In this thesis, focus was laid on investigating further enhancement employing the gap mode concept and a periodic back reflector.

6.2.3.1 Gap modes in solar cell

Gap modes are the localized electromagnetic normal modes that exist in the space between the metal nanoparticle and surface. The excitation of gap

modes strongly depends on the distance of the nanoparticle from the surface. When the particle-surface distance is sufficiently small, the system can support a series of gap modes. For a metal nanosphere-metal surface system, it is observed when the distance between the metal nanosphere from the surface (D) is sufficiently less than the radius of the metal nanosphere(R) i.e., $D/R < 1$. When this condition is satisfied, the electric field becomes more and more localized at the gap between the particle and the surface.

Initially, to understand the gap mode effect, a structure where silver (Ag) nanoparticles of diameter 20 nm are periodically embedded inside the silicon (Si) absorbing layer of thickness 200 nm is considered, as shown in Figure 6.8. A thin aluminium(Al) layer(10 nm) is then coated at a distance ' D ' from the Ag nanoparticles and an Indium Tin Oxide (ITO) layer of thickness 50 nm is taken as the transparent electrode. Numerical analysis is performed based on the finite difference time domain (FDTD) method. Perfectly matched layers on the horizontal axis and periodic on the vertical axis were employed as the boundary conditions during simulation. The simulation for bare solar cell and nanosphere assisted thin film solar cell was also carried out to compare the results.

Figure 6.8 Basic gap mode based solar cell configuration.

The variation of normalized absorption with the incident wavelength of the gap mode based geometry is shown in Figure 6.9(a). Along with this, the normalized absorption of bare silicon thin film solar cell (with Al reflector) and LSP enhanced thin film silicon solar cells (which uses Ag nanospheres in silicon placed far away from Al back reflector) are also plotted. In the case of particle plasmon enhanced solar cell, the Al reflector of thickness 10 nm is placed at a distance 100 nm from the Ag nanoparticles. In all three cases, the silicon thickness is kept constant. It is clear from the figure that enhanced absorption is possible in the case of the proposed configuration (particle-surface distance is taken as 1 nm) compared to the other two cases. About a 10% increment in absorption, compared to the bare thin film silicon solar cell and a 4% increment in absorption, compared to LSP enhanced thin film silicon solar cell is obtained using this configuration for the wavelength band of 400 nm to 1200 nm. Also, enhanced light localization and absorption is observed at longer wavelength range, which is not possible with bare Si thin film solar cells because of the indirect band gap of Si. It shows that broadband light localization and hence improved absorption is possible using this technique.

The metal particle-surface distance (D) is an important factor for the gap mode excitation. Figure 6.9(b) shows the variation of normalized absorption with respect to wavelength for $D = 1$ nm, $D = 5$ nm to $D = 10$ nm. When the distance decreases ($D/R < 1$); the electric field becomes more and more localized at the gap between the particle and the surface. The light localization and resonant

excitation of electromagnetic normal modes in the particle-surface system, results in absorption enhancement inside the active silicon layer.

Figure 6.9 Normalized absorption variation with incident wavelength: for gap mode assisted, LSP (nanoparticle) assisted and bare silicon cell. (b) Normalized intensity variation with particle-surface distance (D). Magnified image of the peaks from 600-625 nm are shown in the inset.

6.2.3.2 Gap mode in solar cell with periodic metal grating structure

It has been established in the previous section that the introduction of the gap mode effect improves absorption by a factor of about 10% when compared to bare silicon with a planar metal reflector. In this work, a configuration to take advantage of the periodic metal grating back reflector and this gap mode effect is proposed as shown in Figure 6.10 and is investigated for absorption enhancement. An Al grating of period 100 nm and line width of 50 nm is used at the back surface of the 250 nm thick silicon active layer. In addition, silver (Ag) nanoparticles of diameter 20 nm are periodically embedded inside the silicon (Si) absorbing layer, as shown in Figure 6.10. The distance between the nanosphere and the metal surface is denoted as ‘D’. Perfectly matched layers on the horizontal axis and periodic on the vertical axis were employed as the boundary conditions during simulation. To calculate the power absorbed in the active silicon layer, power monitors were used at the top and bottom surface of each rectangular silicon slab. The amount of power transmitted through each surface, normalized to source power, was then calculated for each silicon slab. The power absorbed is obtained by subtracting the transmitted power through each monitor and later adding up the absorbed power in each silicon slab. It should be noted that in order to estimate the obtain power absorbed by the thin silicon film more accurately; the power absorbed by the nanoparticle is subtracted from the total power absorbed.

Figure 6.10 Proposed solar cell configuration based on gap mode and periodic metallic grating.

The absorbed power is plotted against the incident wavelength for two different thin film configurations in the analysis: (a) silicon with Ag nanoparticle with a planar Al back layer for gap mode effect with $D=1$ nm and (b) silicon with an Al periodic grating back layer with Ag nanospheres with $D=1$ nm. Configuration (a) reported here is illustrated in Figure 6.8, and the latter one is the proposed configuration shown in Figure 6.10. The absorbed power calculated by the FDTD is normalized with respect to the input power and is shown in Figure 6.11. With a planar Al layer and Ag spheres, extra peaks are observed above 800 nm because of the reflection and gap mode effect which improves the absorption efficiency at longer wavelengths. The efficiency is improved further by using Ag nanoparticle and an Al grating back reflector and is shown by the curve with solid black line in Figure 6.11. This improved absorption is not only due to the light localization due to excitation of gap modes, but it also attributed to the grating back surface acting as an enabler to couple light into the SPP modes which are supported at the metal-semiconductor interface.

Figure 6.11. Absorption inside silicon active layer plotted against incident wavelength for two different gap mode based thin film solar cell configurations. Red line represents absorption in the case of planar Al layer based gap mode configuration and black line represents absorption in the case of grating back layer based gap mode configuration.

An increment of 9% compared to the planar Al-Ag system based configuration was observed due to the combined effect of gap mode excitation and grating back structure.

In order to analyse the effect of the particle-surface distance, the normalized absorption, calculated using methods described previously, was plotted against the wavelength for three different particle-surface distances ($D=1$ nm, $D=5$ nm, and $D=10$ nm), as shown in Figure 6.12. As evident from the plots, when D is reduced from 10 nm to 1 nm, the peak is enhanced and

broadened due to increased localization due to the gap mode effect. Out of all these three cases, the one employing a grating layer and $D=1$ nm has the best silicon absorption value and the absorption improvement over the planar Al-Ag configuration with $D=1$ nm was found to be 9% as previously mentioned. And the improvement factor for a grating with $D=5$ nm case over a planar layer with $D=1$ nm was found to be 8.5%, a little less compared to the grating with $D=1$ nm case owing to reduced localization due to the larger gap. This reduces even further to about 8% for $D=10$ nm, when compared to planar Al-Ag with $D=1$ nm configuration. This is plotted in Figure 6.13 and it is found that beyond $D/R=1$ (here the case where $D=10$ nm) enhancement factor is almost constant and the enhancement can be attributed to the LSP excitation and grating effect alone. It has already been reported that the gap modes resonance wavelength shifts to longer wavelengths when D decreases [128]. From Figure 6.12, it is found that there is a red shift in the resonance wavelength as D decreases from 10 nm to 1 nm. The obtained gap mode resonance wavelengths at the particle-surface distances of 1 nm, 5 nm, and 10 nm are found to be 640 nm, 636 nm and 633 nm respectively. This also proves the existence of gap modes in the space between the nanoparticle and surface.

The same configuration could be extended to include a periodic 3D back reflector instead of a metal grating reflector. A diamond like structure could be employed at the back surface to make use of its photonic band gap properties. The formation of such a structure using five-beam interference lithography has already been reported in chapter 5. Including a 3D photonic crystal structure at

the back surface, the proposed configuration has the potential to improve the absorption inside the silicon layer further. Thus absorption enhancement in the silicon active layer can be realized in the proposed solar cell configuration shown in Figure 6.10, with a 3D periodic back reflector, by utilizing various optical concepts such as; (a) excitation of localized surface plasmon resonances in Ag nanospheres, (b) scattering of light by Ag nanospheres and metallic grating into waveguide modes inside silicon, (c) excitation of surface plasmon polariton (SPP) modes at metal grating-dielectric interface, (d) excitation of gap modes between metallic structures and (e) reflection due to complete band gap 3D periodic back structure.

Figure 6.12. Variation of normalized absorption in silicon layer with respect to incident wavelength for different particle-surface distance (D). Magnified view of peaks from 625-650 nm and from 775-800 nm are shown in the inset.

Figure 6.13 Variation of percentage absorption enhancement with particle-surface distance. It can be seen that highest absorption is obtained for $D=1$ nm.

6.3 Summary

In conclusion, two major applications of periodic structures, namely enhanced transmission through a 2D metallic grating and light trapping in silicon based solar cell were numerically investigated. In the case of enhanced SP based lithography, the numerical simulation results show that field transmission through the sub-50 nm featured metal mask can be enhanced by a factor of 14.5, with the help of an amplifying medium. The gain medium assists the enhanced SP propagation by compensating intrinsic loss associated with the metal, thereby exciting the 485 nm resonance modes inside the cavity. This excited mode will further improve the total transmission of the sub-wavelength apertures. The enhancement of surface plasmons on the aluminium film is due to the fact that the plasmon resonance frequency is so closely matched to the emission

wavelength of the dye. This study reveals that the field enhancement with gain medium offers higher penetration depth in photoresist during lithography, which in turn would improve the aspect ratio of the fabricated features.

Also, absorption enhancement using periodic structures in thin film silicon based solar cell was also studied for various configurations. Initially, it was established that the metal grating back surface of sub-wavelength periodicity improves absorption considerably. It was found to improve absorption by a factor of about 12% when compared to a planar metal back layer configuration. Subsequently, a periodic silicon front surface was introduced and an improvement of 7% over an Al grating back reflector for a period of 100 nm, owing to scattering and guided modes, was observed. The gap mode effect in thin film solar cells was also studied and a new configuration which takes advantage of a periodic metal back reflector and gap mode effect was proposed. The configuration employing Ag spheres and Al back grating layer was found to improve absorption further by a factor of 9% over the planar metal layer-Ag sphere configuration, when the gap distance was set to 1 nm. Overall, the various configurations taking advantages of different principles like SPP, gap mode effect, scattering etc. was introduced and studied. And it was found that the periodic back structures and front structures improves light trapping and hence absorption efficiency considerably and will form integral parts of thin film solar cell structures in the future.

Chapter 7: Conclusion and Future Work

This chapter concludes the research carried out as part of the thesis and the major contributions made are stated in sequence. Some of the areas for further research and development are also identified and are proposed briefly in this chapter.

7.1 Conclusion

In this thesis, investigations were carried out on various mask less optical lithographic techniques for the fabrication of 1D, 2D and 3D periodic features with periodicity varying from few micrometres to 100 nm. Initially, a wavefront division approach using multi-facet prisms along with UV and DUV laser source was investigated for the fabrication of uniform large area two dimensional periodic features. This approach was identified as a powerful technique for the fabrication of uniform features with micron and sub-micron periodicity because it uses a zero path interferometer. Apart from uniformity, the system uses a minimum number of optical components and so can be categorised as a simple experimental setup. Initially, theoretical investigations of pitch variation in a biprism interferometer were carried out followed by experimental validation using various prisms. For example, both 5° and 2.5° biprism's were used to fabricate grating and square array of pillars with periodicity of $3.75 \pm 0.02 \mu\text{m}$, and $1.66 \pm 0.01 \mu\text{m}$ respectively using a 364 nm laser source. Theoretically, it was

shown that pitch size can go as small as 210 nm at a wavelength of 363.8 nm, using a biprism interferometer lithography. The feature width was controlled by controlling the exposure dose and development time. Hexagonal features were also fabricated using a single exposure triprism based LIL, eliminating the need of multiple exposure and the associated rotation of the sample in between the exposures. Resolution improvement could be directly achieved in such a system by replacing the UV source with a deep UV light source. In this thesis, a DUV source with wavelength of 266 nm was also employed in a biprism interferometer configuration to define periodic patterns with much improved resolution. Features with sub-500 nm pitch and sub-200 nm resolution were fabricated using a 30.4° fused silica biprism. 2D periodic features in a square lattice were also fabricated using a double exposure technique and using a pyramidal prism. Sources with shorter wavelengths could be used in such a system to improve resolution further but the higher cost of the laser source and the non-availability of transmission optics at shorter range make this a non-viable option. In this context, prism based solid immersion lithography was investigated for patterning, with improved resolution and pitch size which scales beyond $\lambda/2$ value.

Two configurations for the prism based solid immersion lithography were investigated in the later part of this thesis, in Chapter 4. The first configuration uses a grating to split the beam, mirrors to guide it and a high index prism to make it interfere. This is a conventional immersion interferometric technique where the pitch size is controlled by changing the angle of intersection by

rotating the mirrors. High resolution features could be fabricated where feature size is limited by the critical angle at the prism-index matching liquid interface. For example, uniform periodic features with a 110 ± 6 nm pitch and a 55 ± 4 nm resolution were fabricated using an i-line laser based configuration. As the source used is of wavelength 364 nm, visible wavelength optical components and a high index prism (LAK31A) of angle 60° could be used, thereby reducing the expenses compared to a configuration using a DUV source. The pitch size and resolution can be scaled down further using the same configuration by employing a suitable index matching liquid to increase the critical angle.

The second configuration uses an inverted prism to split the beam, and multiple converging lenses to direct the beam on to the high index prism. Here, the use of a mirror assembly is avoided and it is particularly advantageous when numbers of beams involved are increased to more than two. For instance, both two and four-beam interference requires only the same number of lenses (two lenses in this work). The number of optical components is half when compared to a mirror based four beam-configuration where four mirrors are required to guide the beams. Both the single lens and two lens systems for interference lithography has been theoretically investigated. It was found that the pitch can be controlled by adjusting the distance between lenses, changing the split angle or by changing the distance between the splitter and first lens. Square lattice features with a pitch size of 210 ± 8 nm and 240 ± 6 nm were patterned using an i-line laser source and two lenses (biconvex)-prism based system. Various lithographic techniques

for periodic feature patterning investigated in this thesis is compared to the standard mask based photolithographic technique and is shown in the Table 7.1.

Table 7.1 Biprism based LIL and solid immersion based LIL compared with the mask based photolithography.

Lithography Technique Properties	Mask based Photolithography (i-line)	Biprism LIL (30° angle, BK7 prism, i- line)	Biprism LIL (30.4°, Fused Silica prism, 266nm)	SIL – Amplitude division (i-line)
Period →	~200nm	450±8nm	410±11 nm	110±6nm
Resolution →	~100nm	200±4nm	206±5 nm	55±4nm
Speed →	Very Fast	Very fast	Very fast	Very fast
Writing nature →	Parallel	Parallel	Parallel	Parallel
Mask requirement →	Yes	No	No	No
3D patterning →	No	No	No	Yes

No interference study is complete without analysing the effect of polarization on the resultant intensity patterns. So in chapter 5, the effect of polarization on two-beam, four-beam and five-beam interference pattern has been reported. In two-beam interference, polarization of individual beams was found to have a major influence only in the fringe contrast whereas four-beam and five-beam interference pattern is found to be much more sensitive to polarization. The formation of periodic 2D square lattice patterns using four-beam interference, has been studied in detail for different polarization combinations involving TE, TM, LC and RC polarized light beams. It was observed that all polarization combinations produces patterns with square symmetry and a period

of $\sqrt{2}\lambda/2\sin\theta$, except when all beams are TE polarized, where the period of the square lattice was found to be $\lambda/2\sin\theta$. Experimental verification was done for three cases: TE-TE, TE-TM and TM-TM by recording the patterns in a positive photoresist. Later, formation of interesting 3D periodic patterns investigated using five-beam interference with circularly polarized central beam. It was found that body centred tetragonal structures could be obtained with a circularly polarized central beam and four other beams in the TE-TM polarized state. It was also found that if the four plane waves in orthogonal planes are TE polarized, a β -tin type structure could be realized, which can be converted into diamond cubic like structure by increasing the angle of incidence. This structure has interesting photonic band gap properties.

Having investigated and developed some of the prism based normal and immersion based optical interference lithographic systems, the focus was made on investigating relevant application areas of periodic features in photonics. Two major applications of periodic structures in the current hot field of plasmonics, namely dye based enhanced transmission through 2D metallic grating and light trapping in silicon based solar cell, were identified and investigated numerically in the later part of this thesis. Initially, a configuration for surface plasmon based lithography, employing a periodic metal array as mask and dye as a gain medium was proposed. The transmission through the sub-50 nm featured metal mask, which is due to excitation of a surface plasmon polariton was shown to be enhanced by a factor of 14.5 with the help of an amplifying dye medium. The

gain medium assists the enhanced SP propagation by compensating intrinsic loss associated with metal. The enhancement of surface plasmons on the aluminium film is due to fact that the SPP resonance frequency is closely matched with the emission wavelength of the dye. This study reveals that the field enhancement with gain medium offers higher penetration depth in photoresist during lithography, which in turn would improve the aspect ratio of the fabricated features. It should be noted that the fabrication of periodic metal mask on dye embedded polymer is challenging, and to the best of our knowledge the only viable option which can be used for patterning is optical lithographic technique. This is because of the sensitivity of dye and polymer to electron beam and x-ray radiation (and so both e beam and x-ray lithography cannot be used for patterning).

The second major plasmonic application of sub-wavelength periodic features identified was that of thin film solar cell. Absorption enhancement using periodic structures in thin film silicon based solar cell was studied numerically for various configurations. Initially, it was established that a metal grating back surface of sub-wavelength periodicity improves absorption considerably. It was found to improve absorption by about 12% when compared to a planar metal back layer configuration. Later, a periodic silicon front surface was introduced and an improvement of about 7% over an Al grating back reflector for a period of 100 nm, owing to scattering and guided modes was observed. The gap mode effect in thin film solar cell was also studied and a new configuration which takes advantage of a periodic metal back reflector and the gap mode effect was

proposed. The configuration used Ag spheres and an Al back grating layer was found to improve absorption further by a factor of 9% over a planar metal layer-Ag sphere configuration, when the gap distance was set to 1 nm. Overall, various configurations taking advantages of different principles like SPP, gap mode effect, scattering etc. was introduced and studied. It was found that periodic back structures and front structures improves light trapping and hence absorption efficiency considerably and will form integral parts of thin film solar cell structures in the future. Fabrication and testing of such thin film cells are challenging and provides scope for future studies.

7.2 Major contributions

This thesis has presented investigations on various maskless optical interferometric systems for micron to sub-100 nm periodic feature patterning. Apart from these, numerical investigations on two important plasmonic applications namely, dye based enhanced transmission through 2D metallic grating and light trapping in silicon based solar cell were carried out successfully. The main contribution of this thesis are summarized as follows,

1. Multi facet prism based wavefront interferometric system using collimated beam for lithography was proposed, theoretically investigated and experimentally validated.

2. Grating and mirror based conventional lithographic system for solid immersion was theoretically and experimentally investigated and features with periodicity as low as $\lambda/3$ and resolution as high as $\lambda/6$ were fabricated.

3. Multiple converging lenses based solid immersion lithographic configuration was proposed, theoretically investigated and experimentally validated.

4. Detailed theoretical analysis of two and four-beam interference for various polarization combinations involving TE, TM and circularly polarized beams has been carried out.

5. Formation of 3D structures such as body centred tetragonal, β -tin type structure and diamond like structure employing five-beam interference with circularly polarized central beam has been investigated numerically.

6. A configuration for surface plasmon based lithography, employing periodic metal array as mask and dye as a gain medium was proposed for enhancing light propagation through sub-50 nm apertures and was numerically investigated.

7. Various methodologies for enhanced light trapping in thin film solar cells employing periodic structures and gap mode effect were numerically investigated.

Based on some of the research issues identified in this work, a few possible areas for future investigations are proposed and are discussed in detail in the following section.

7.3 Future Work

7.3.1 Fabrication and testing of thin film solar cell with periodic metal grating–sphere system

It was demonstrated using simulations in Chapter 6 that employing a gap mode concept using Ag-Al system improves light trapping and hence absorption efficiency considerably by exciting extra modes in the gap between them. The configuration employing Ag spheres and an Al planar back layer was found to improve absorption by 10% when compared to a configuration with just the planar back layer. Also the configuration employing Ag spheres and Al back grating layer was found to improve absorption further by a factor of 9% over a planar metal layer-Ag sphere configuration, when the gap distance was set to 1 nm. Overall, these configurations were found to take advantage of different principles like SPP, gap mode effect, scattering etc. to enhance light trapping inside active silicon layer and the fabrication of thin film inorganic solar cell incorporating this gap mode concept poses a major challenge and is expected to be explored in the future. Embedding Ag nanospheres inside silicon layer and maintaining the gap between them and back metal layer (either planar or grating) such that $D/R < 1$ is a major fabrication challenge.

7.3.2 Fabrication of 3D diamond like photonic crystals using five-beam interference lithography

A 3D diamond like photonic crystal are widely reported to have a high full photonic band gap. Realization of 3D periodic features has been demonstrated in Chapter 5 of this thesis. However, the interconnection of high intensity points in a diamond lattice is something to be explored in the future. Also, efficient pattern transfer should be carried out, if silicon-air photonic crystal is desired. One way is to use resist patterns obtained by interference lithography as a mould and then deposit silicon to create silicon-air photonic diamond like crystals. One major problem with this method is that photoresist will get washed away during high temperature silicon deposition technique. However, deposition of a thin layer of alumina on resist-air photonic crystal can solve this problem by enabling them to withstand temperatures, as high as 330°C. Process flow for this method is shown in the block diagram in Figure 7.1

Method to obtain silicon-air 3D photonic crystal from resist-air crystal obtained by interference lithography. The alumina coated photonic crystals can be filled with amorphous silicon using low temperature static chemical vapour deposition technique. Once such crystals are fabricated, it could serve as excellent back reflectors for thin film solar cells. This diamond like 3D back reflector can be employed instead of Al grating reflector which has been discussed in Chapter 6, to improve absorption further inside the silicon layer, by exploiting the complete band gap property. Numerical and experimental verification of this concept would be an excellent topic for future work.

Figure 7.1 Method to obtain silicon-air 3D photonic crystal from resist-air crystal obtained by interference lithography.

Appendices

Appendix A: Derivation of intensity distribution for different polarization combinations in multiple-beam interference lithography

The time intensity distribution obtained by the interference of two or more than two coherent light waves can be expressed as [117],

$$I = \langle \sum_j E_i^2(r, t) \rangle \quad \text{A.1}$$

$$+ \sum_{i < j} (E_i E_j) |\hat{e}_i \cdot \hat{e}_j^*| \cos [(k_i - k_j) \cdot r + (\varphi_i - \varphi_j) + \delta_{ij}]$$

With , $\delta_{ij} = \arg\{\hat{e}_i \cdot \hat{e}_j^*\}$

Where, E_i is the amplitude, ω the frequency, k_i is the wave vector, φ_i is the phase and $\hat{e}_i$ is the unit polarization vector respectively. Also note that $\hat{e}_j^*$ represent the conjugate of the unit polarization vector.

A.1 Two-Beam Interference

One dimensional periodic feature can be fabricated by interfering two coherent plane waves. If the two interfering beams with wavelength ' λ ' are linearly polarized waves and are incident at ' $\pm \theta$ ' with respect to the z axis, the intensity distribution is given by,

$$I(r) = \frac{(E_1^2 + E_2^2)}{2} + E_1 E_2 |\hat{e}_1 \cdot \hat{e}_2^*| \cos[k_0 2 \sin \theta x + (\varphi_1 - \varphi_2) + \delta_{12}] \quad \text{A.2}$$

Where, $k_0 = 2\pi/\lambda$

And the spatial-cosine term of intensity distribution describes a periodic pattern that varies along the x axis with a periodicity given by ' $p = \lambda/2 \sin \theta$ '. And the fringe contrast is defined by $|\hat{e}_1 \cdot \hat{e}_2^*|$ [119, 120].

The effect of polarization in the two beam interference pattern is that it changes the contrast of the obtained fringe pattern. The intensity distribution for different cases of two beam interference involving linearly polarized and circularly polarized beams are given in Table 5.1 and their derivation is given in detail below.

Figure A.1 Two beam interference.

1. Transverse electric (TE)- Transverse electric (TE):

The unit polarization vectors $\hat{e}_1$ and $\hat{e}_2$ for the two beams shown in Figure A.1, when they are both TE polarized are given by: $\hat{e}_1 = \hat{e}_2 = \hat{y}$

$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = 1$$

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The magnitude and argument of the function $\hat{e}_1 \cdot \hat{e}_2^*$ is given by,

$$\Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = 1 \quad \& \quad \delta_{12} = \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

Substituting the above in Eq. (A.2) and assuming that both the beams has equal amplitude E_0 and zero phase difference, the intensity distribution is given by,

$$I(r)_{TE} = E_0^2 \left(\frac{1}{2} + \frac{1}{\sqrt{2}} \cos[k_0 2 \sin \theta x] \right)$$

The fringe contrast is defined by $|\hat{e}_1 \cdot \hat{e}_2^*|$ and in this case it equals 1.

2. Transverse Magnetic (TM)- Transverse Magnetic (TM):

If both the beams in Figure A.1 are TM polarized, the unit polarization vectors are given by: $\hat{e}_1 = (\cos \theta \hat{x} + \sin \theta \hat{z})$ and $\hat{e}_2 = (\cos \theta \hat{x} - \sin \theta \hat{z})$.

$$\hat{e}_1 \cdot \hat{e}_2^* = \cos 2\theta ; \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = |\cos 2\theta| \quad \& \quad \delta_{12} = \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

Substituting the above in Eq. (A.2) and assuming that both the beams has equal amplitude E_0 and zero phase difference, the intensity distribution is given by,

$$I(r)_{TM} = E_0^2 \left(\frac{1}{2} + |\cos 2\theta| \cos[k_0 2 \sin \theta x] \right)$$

$$\text{Fringe contrast: } |\hat{e}_1 \cdot \hat{e}_2^*| = |\cos 2\theta|$$

3. Transverse electric (TE)- Transverse Magnetic (TM):

If beam 1 and beam 2 in Figure A.1 are TE and TM polarized respectively, the unit polarization vectors are given by: $\hat{e}_1 = \hat{y}$ and $\hat{e}_2 = (\cos \theta \hat{x} + \sin \theta \hat{z})$

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$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = 0$$

Substituting the above in Eq. (A.2) and assuming that both the beams has equal amplitude E_0 and zero phase difference, the intensity distribution is given by,

$$I(r)_{TE-TM} = E_0^2$$

And the fringe contrast, $|\hat{e}_1 \cdot \hat{e}_2^*| = 0$

4. Transverse electric (TE)-Left circularly polarized (LC):

If beam 1 and beam 2 in Figure A.1 are TE and LC polarized respectively, the unit polarization vectors are given by [118]: $\hat{e}_1 = \hat{y}$ and $\hat{e}_2 = \frac{\cos \theta \hat{x} + i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}$

$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = \frac{i}{\sqrt{2}} ; \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1}{\sqrt{2}} \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = \left(\frac{\pi}{2}\right)$$

The intensity distribution in this case is given by,

$$I(r)_{TE-LC} = E_0^2 \left(\frac{1}{2} + \frac{1}{\sqrt{2}} \cos \left[k_0 2 \sin \theta x + \frac{\pi}{2} \right] \right)$$

And the fringe contrast is given by, $|\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1}{\sqrt{2}}$

5. Transverse electric (TE)-Right circularly polarized (RC):

If beam 1 and beam 2 in Figure A.1 are TE and RC polarized respectively, the unit polarization vectors are given by: $\hat{e}_1 = \hat{y}$ and $\hat{e}_2 = \frac{\cos \theta \hat{x} - i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}$

$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = \frac{-i}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1}{\sqrt{2}} \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = \left(\frac{-\pi}{2}\right)$$

The intensity distribution is given by,

$$I(r)_{\text{TE-RC}} = E_0^2 \left(\frac{1}{2} + \frac{1}{\sqrt{2}} \cos \left[k_0 2 \sin \theta x - \frac{\pi}{2} \right] \right) = E_0^2 \left(\frac{1}{2} + \frac{1}{\sqrt{2}} \sin[k_0 2 \sin \theta x] \right)$$

And the fringe contrast is given by, $|\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1}{\sqrt{2}}$

6. Transverse Magnetic (TM)-Left circularly polarized (LC):

If beam 1 and beam 2 in Figure A.1 are TM and LC polarized respectively, the unit polarization vectors are given by: $\hat{e}_1 = (\cos \theta \hat{x} + \sin \theta \hat{z})$ and $\hat{e}_2 = \frac{\cos \theta \hat{x} + i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}$

$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = \frac{|\cos 2\theta|}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = \frac{|\cos 2\theta|}{\sqrt{2}} \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

Substituting the above in Eq. (A.2) and assuming that both the beams has equal amplitude E_0 and zero phase difference, the intensity distribution is given by,

$$I(r)_{\text{TM-LC}} = E_0^2 \left(\frac{1}{2} + \frac{\cos 2\theta}{\sqrt{2}} \cos[k_0 2 \sin \theta x] \right)$$

And the fringe contrast is given by, $|\hat{e}_1 \cdot \hat{e}_2^*| = \frac{|\cos 2\theta|}{\sqrt{2}}$

7. Transverse Magnetic (TM)--Right circularly polarized (RC):

If beam 1 and beam 2 in Figure A.1 are TM and RC polarized respectively, the unit polarization vectors are given by: $\hat{e}_1 = (\cos \theta \hat{x} + \sin \theta \hat{z})$ and $\hat{e}_2 = \frac{\cos \theta \hat{x} - i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}$

$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = \frac{|\cos 2\theta|}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = \frac{|\cos 2\theta|}{\sqrt{2}} \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

Substituting the above in Eq. (A.2) and assuming that both the beams has equal amplitude E_0 and zero phase difference, the intensity distribution is given by,

$$I(r)_{\text{TM-RC}} = E_0^2 \left(\frac{1}{2} + \frac{\cos 2\theta}{\sqrt{2}} \cos[k_0 2 \sin \theta x] \right)$$

And the fringe contrast is given by, $|\hat{e}_1 \cdot \hat{e}_2^*| = \frac{|\cos 2\theta|}{\sqrt{2}}$

8. Left circularly polarized (LC)- Left circularly polarized (LC):

If beam 1 and beam 2 in Figure A.1 are LC and LC polarized respectively, the unit polarization vectors are given by: $\hat{e}_1 = \frac{\cos \theta \hat{x} + i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}$ and $\hat{e}_2 = \frac{\cos \theta \hat{x} + i\hat{y} + \sin \theta \hat{z}}{\sqrt{2}}$

$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = \frac{1 + \cos 2\theta}{2} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1 + \cos 2\theta}{2} \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

Substituting the above in Eq. (A.2) and assuming that both the beams has equal amplitude E_0 and zero phase difference, the intensity distribution is given by,

$$I(r)_{\text{LC-LC}} = E_0^2 \left(\frac{1}{2} + \frac{(1 + \cos 2\theta) \cos[k_0 2 \sin \theta x]}{2} \right)$$

And the fringe contrast is given by, $|\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1 + \cos 2\theta}{2}$

9. Right circularly polarized (RC)- Right circularly polarized (RC):

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If beam 1 and beam 2 in Figure A.1 are RC and RC polarized respectively, the unit polarization vectors are given by $\hat{e}_1 = \frac{\cos \theta \hat{x} - i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}$ and $\hat{e}_2 = \frac{\cos \theta \hat{x} - i\hat{y} + \sin \theta \hat{z}}{\sqrt{2}}$

$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = \frac{1 + \cos 2\theta}{2} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1 + \cos 2\theta}{2} \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

Substituting the above in Eq. (A.2) and assuming that both the beams has equal amplitude E_0 and zero phase difference, the intensity distribution is given by,

$$I(r)_{LC-LC} = E_0^2 \left(\frac{1}{2} + \frac{(1 + \cos 2\theta) \cos[k_0 2 \sin \theta x]}{2} \right)$$

And the fringe contrast is given by, $|\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1 + \cos 2\theta}{2}$

10. Left circularly polarized (LC))- Right circularly polarized (RC):

If beam 1 and beam 2 in Figure A.1 are LC and RC polarized respectively, the unit polarization vectors are given by: $\hat{e}_1 = \frac{\cos \theta \hat{x} + i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}$ and $\hat{e}_2 = \frac{\cos \theta \hat{x} - i\hat{y} + \sin \theta \hat{z}}{\sqrt{2}}$

$$\Rightarrow \hat{e}_1 \cdot \hat{e}_2^* = \frac{-1 + \cos 2\theta}{2} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1 - \cos 2\theta}{2} \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

Substituting the above in Eq. (A.2) and assuming that both the beams has equal amplitude E_0 and zero phase difference, the intensity distribution is given by,

$$I(r)_{RC-RC} = E_0^2 \left(\frac{1}{2} + \frac{(1 - \cos 2\theta) \cos[k_0 2 \sin \theta x]}{2} \right)$$

And the fringe contrast is given by, $|\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1 - \cos 2\theta}{2}$

The intensity distribution and contrast of the fringe pattern formed by two beam interference for $E_0 = 1$ and $\varphi_1 - \varphi_2 = 0$ is tabularized and is shown in the Table 5.1.

A.2 Four-beam Interference

Let us consider the interference of four coherent waves that converge at equal angles along two perpendicular planes. The electric field vectors of four perfectly aligned coherent beams, two incident at $\pm\theta$ one plane and the other two incident at $\pm\theta$ on an orthogonal plane as show in Figure A.2 can be described by,

$$\begin{aligned} E_1(r, t) &= E_1 \hat{e}_1 \cos [(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_1] \\ E_2(r, t) &= E_2 \hat{e}_2 \cos [-(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_2] \\ E_3(r, t) &= E_3 \hat{e}_3 \cos [(k \sin \theta)y - (k \cos \theta)z - \omega t + \varphi_3] \\ E_4(r, t) &= E_4 \hat{e}_4 \cos [-(k \sin \theta)y - (k \cos \theta)z - \omega t + \varphi_4] \end{aligned} \quad \text{A.3}$$

Where, k is the magnitude of propagation vector, ω is the angular frequency, $\hat{e}_i$ is the polarization vector and φ_i is the phase constant of each wave. Time independent intensity distribution for four-beam interference with various polarization combinations for the two beam pairs shown in the Figure A.2 are given in the Table 5.2. The derivation of the same is discussed in detail in the following.

Figure A.2 Four-beam interference configuration.

The time independent intensity distribution obtained by the interference of the four waves shown in Figure A.2 is obtained by using Eq. (A.1) and Eq. (A.3) and is given by,

$$\begin{aligned}
 I(r) = & \frac{(E_1^2 + E_2^2 + E_3^2 + E_4^2)}{2} & \text{A.4} \\
 & + E_1 E_2 |\hat{e}_1 \cdot \hat{e}_2^*| \cos[k_0 2 \sin \theta x + (\varphi_1 - \varphi_2) + \delta_{12}] \\
 & + E_1 E_3 |\hat{e}_1 \cdot \hat{e}_3^*| \cos[k \sin \theta x - k \sin \theta y + (\varphi_1 - \varphi_3) + \delta_{13}] \\
 & + E_1 E_4 |\hat{e}_1 \cdot \hat{e}_4^*| \cos[k \sin \theta x + k \sin \theta y + (\varphi_1 - \varphi_4) + \delta_{14}] \\
 & + E_2 E_3 |\hat{e}_2 \cdot \hat{e}_3^*| \cos[-k \sin \theta x - k \sin \theta y + (\varphi_2 - \varphi_3) + \delta_{23}] \\
 & + E_2 E_4 |\hat{e}_2 \cdot \hat{e}_4^*| \cos[-k \sin \theta x + k \sin \theta y + (\varphi_2 - \varphi_4) + \delta_{24}] \\
 & + E_3 E_4 |\hat{e}_3 \cdot \hat{e}_4^*| \cos[k_0 2 \sin \theta y + (\varphi_3 - \varphi_4) + \delta_{34}]
 \end{aligned}$$

1) Transverse Electric (TE)- Transverse Electric (TE):

Now let us consider the cases where all the interfering beams are transverse electric polarized (TE). The four polarization vectors are defined by,

$$\hat{e}_1 = \hat{e}_2 = \hat{y} \quad \& \quad \hat{e}_3 = \hat{e}_4 = \hat{x}$$

This scenario is equivalent to the case where two sets of two beam interference patterns that intersect perpendicularly are added together, as the

orthogonal polarized waves doesn't interfere. The four-beam interference pattern produced by this set of polarization vectors i.e., TE-TE pattern, is given by time independent intensity distribution expressed as,

$$I(r)_{TE-TE} = \frac{(E_1^2 + E_2^2 + E_3^2 + E_4^2)}{2} \\ + E_1 E_2 |\hat{e}_1 \cdot \hat{e}_2^*| \cos[k_0 2 \sin \theta x + (\varphi_1 - \varphi_2)] \\ + E_3 E_4 |\hat{e}_3 \cdot \hat{e}_4^*| \cos[k_0 2 \sin \theta y + (\varphi_3 - \varphi_4)]$$

And when all waves has equal amplitude $E_1 = E_2 = E_3 = E_4 = E_0$ and zero phase difference, it can be simplified as,

$$I(r)_{TE-TE} = E_0^2 \{2 + \cos[k_0 2 \sin \theta x] + \cos[k_0 2 \sin \theta y]\} \quad \text{A.5}$$

2) Transverse Electric (TE)- Transverse Magnetic (TM):

Now let's consider the case where one pair of plane waves (beam 1 and beam 2) shown in Figure A.2 is TE polarized and the other pair (beam 3 and beam 4) is TM polarized. The unit polarization vectors for the four beams in this case are given by,

$$\hat{e}_1 = \hat{e}_2 = \hat{y} \quad , \quad \text{A.6}$$

$$\hat{e}_3 = (\cos \theta \hat{y} + \sin \theta \hat{z}) \quad \& \quad \hat{e}_4 = (\cos \theta \hat{y} - \sin \theta \hat{z})$$

And the time independent intensity distribution in Eq. (A.4) in the case of TE-TM case is expressed as,

$$\begin{aligned}
 I(r)_{TE-TM} = & \frac{(E_1^2 + E_2^2 + E_3^2 + E_4^2)}{2} + E_1 E_2 \cos[2k \sin \theta x + (\varphi_1 - \varphi_2)] \\
 & + E_1 E_3 |\cos \theta| \cos[k \sin \theta x - k \sin \theta y + (\varphi_1 - \varphi_3)] \\
 & + E_1 E_4 |\cos \theta| \cos[k \sin \theta x + k \sin \theta y + (\varphi_1 - \varphi_4)] \\
 & + E_2 E_3 |\cos \theta| \cos[-k \sin \theta x - k \sin \theta y + (\varphi_2 - \varphi_3)] \\
 & + E_2 E_4 |\cos \theta| \cos[-k \sin \theta x + k \sin \theta y + (\varphi_2 - \varphi_4)] \\
 & + E_3 E_4 |\cos 2\theta| \cos[2k \sin \theta y + (\varphi_3 - \varphi_4)]
 \end{aligned}$$

If all the waves has the same amplitude and are in phase or are out of phase by the same value, the intensity distribution is given by,

$$\begin{aligned}
 I(r)_{TE-TM} = E_0^2 \{ & 2 + \cos(2k \sin \theta x) + |\cos 2\theta| \cos(2k \sin \theta y) \\
 & + 4|\cos \theta| \cos(k \sin \theta x) \cos(k \sin \theta y) \}
 \end{aligned} \tag{A.7}$$

3) Transverse Magnetic (TM) - Transverse Magnetic (TM):

When all the beams are TM polarized, the polarization vectors for the four beams mentioned in Eq. (A.3) are given by,

$$\begin{aligned}
 \hat{e}_1 = (\cos \theta \hat{x} + \sin \theta \hat{z}); \quad \hat{e}_2 = (\cos \theta \hat{x} - \sin \theta \hat{z}); \\
 \hat{e}_3 = (\cos \theta \hat{y} + \sin \theta \hat{z}) \quad \& \quad \hat{e}_4 = (\cos \theta \hat{y} - \sin \theta \hat{z})
 \end{aligned} \tag{A.8}$$

The time independent intensity distribution in this case can be obtained from Eq. (A.1), Eq. (A.4) and Eq. (A.8) and can be expressed as,

$$\begin{aligned}
 I(r)_{TM-TM} = & \frac{(E_1^2 + E_2^2 + E_3^2 + E_4^2)}{2} \\
 & + E_1 E_2 |\cos 2\theta| \cos[2k \sin \theta x + (\varphi_1 - \varphi_2)] \\
 & + E_1 E_3 |\sin^2 \theta| \cos[k \sin \theta x - k \sin \theta y + (\varphi_1 - \varphi_3)] \\
 & + E_1 E_4 |\sin^2 \theta| \cos[k \sin \theta x + k \sin \theta y + (\varphi_1 - \varphi_4)] \\
 & + E_2 E_3 |\sin^2 \theta| \cos[-k \sin \theta x - k \sin \theta y + (\varphi_2 - \varphi_3)] \\
 & + E_2 E_4 |\sin^2 \theta| \cos[-k \sin \theta x + k \sin \theta y + (\varphi_2 - \varphi_4)] + \\
 & + E_3 E_4 |\cos 2\theta| \cos[2k \sin \theta y + (\varphi_3 - \varphi_4)]
 \end{aligned}$$

If all the waves has the same amplitude E_0 and are in phase or are out of phase by the same value, the intensity distribution is given by,

$$\begin{aligned}
 I(r)_{TM-TM} = E_0^2 \{ & 2 + |\cos 2\theta| \cos(2k \sin \theta x) \\
 & + 4|\sin^2 \theta| \cos(k \sin \theta x) \cos(k \sin \theta y) \\
 & + |\cos 2\theta| \cos(2k \sin \theta y) \}
 \end{aligned} \tag{A.9}$$

4) Left Circularly polarized (LC)- Left Circularly polarized (LC):

When both the beam pairs shown in Figure A.2 are LC polarized, the unit polarization vectors are given by [118],

$$\begin{aligned}
 \hat{e}_1 = \frac{\cos \theta \hat{x} + i\hat{y} + \sin \theta \hat{z}}{\sqrt{2}}; \quad \hat{e}_2 = \frac{\cos \theta \hat{x} + i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}; \\
 \hat{e}_3 = \frac{\cos \theta \hat{y} + i\hat{x} + \sin \theta \hat{z}}{\sqrt{2}} \quad \& \quad \hat{e}_4 = \frac{\cos \theta \hat{y} + i\hat{x} - \sin \theta \hat{z}}{\sqrt{2}}
 \end{aligned} \tag{A.10}$$

The time independent intensity distribution in this case can be obtained from Eq. (A.10), Eq. (A.1) and Eq. (A.4) and can be expressed as,

$$\begin{aligned}
 I(r)_{LC-LC} = & \frac{(E_1^2 + E_2^2 + E_3^2 + E_4^2)}{2} + E_1 E_2 \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta x + (\varphi_1 - \varphi_2)] \\
 & + E_1 E_3 |\sin^2 \theta| \cos[k \sin \theta x - k \sin \theta y + (\varphi_1 - \varphi_3)] \\
 & + E_1 E_4 |\sin^2 \theta| \cos[k \sin \theta x + k \sin \theta y + (\varphi_1 - \varphi_4) + \pi] \\
 & + E_2 E_3 |\sin^2 \theta| \cos[-k \sin \theta x - k \sin \theta y + (\varphi_2 - \varphi_3) + \pi] \\
 & + E_2 E_4 |\sin^2 \theta| \cos[-k \sin \theta x + k \sin \theta y + (\varphi_2 - \varphi_4)] \\
 & + E_3 E_4 \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y + (\varphi_3 - \varphi_4)]
 \end{aligned}$$

If all the waves has the same amplitude E_0 and are in phase or are out of phase by the same value, the intensity distribution is given by,

$$I(r)_{LC-LC} = E_0^2 \{2 + |\cos^2 \theta| \cos(2k \sin \theta x) + |\cos^2 \theta| \cos(2k \sin \theta y)\} \quad \text{A.11}$$

5) Right Circularly polarized (RC)- Right Circularly polarized (RC):

When both the beam pairs shown in Figure A.2 are RC polarized, the unit polarization vectors are given by,

$$\begin{aligned}
 \hat{e}_1 &= \frac{\cos \theta \hat{x} - i\hat{y} + \sin \theta \hat{z}}{\sqrt{2}}; \quad \hat{e}_2 = \frac{\cos \theta \hat{x} - i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}; \\
 \hat{e}_3 &= \frac{\cos \theta \hat{y} - i\hat{x} + \sin \theta \hat{z}}{\sqrt{2}} \& \hat{e}_4 = \frac{\cos \theta \hat{y} - i\hat{x} - \sin \theta \hat{z}}{\sqrt{2}}
 \end{aligned} \quad \text{A.12}$$

The time independent intensity distribution in this case can be obtained from Eq. (A.12), Eq. (A.1) and Eq. (A.4) and can be expressed as,

$$\begin{aligned}
 I(r)_{RC-RC} = & \frac{(E_1^2 + E_2^2 + E_3^2 + E_4^2)}{2} + E_1 E_2 \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta x + (\varphi_1 - \varphi_2)] \\
 & + E_1 E_3 |\sin^2 \theta| \cos[k \sin \theta x - k \sin \theta y + (\varphi_1 - \varphi_3)] \\
 & + E_1 E_4 |\sin^2 \theta| \cos[k \sin \theta x + k \sin \theta y + (\varphi_1 - \varphi_4) + \pi] \\
 & + E_2 E_3 |\sin^2 \theta| \cos[-k \sin \theta x - k \sin \theta y + (\varphi_2 - \varphi_3) + \pi] \\
 & + E_2 E_4 |\sin^2 \theta| \cos[-k \sin \theta x + k \sin \theta y + (\varphi_2 - \varphi_4)] \\
 & + E_3 E_4 \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y + (\varphi_3 - \varphi_4)]
 \end{aligned}$$

If all the waves has the same amplitude E_0 and are in phase or are out of phase by the same value, the intensity distribution is given by,

$$\begin{aligned}
 I(r)_{RC-RC} = & E_0^2 \{ 2 + |\cos^2 \theta| \cos(2k \sin \theta x) \\
 & + |\cos^2 \theta| \cos(2k \sin \theta y) \}
 \end{aligned} \tag{A.13}$$

6) Left Circularly polarized (LC)- Right Circularly polarized (RC):

When one pair of plane waves (beam 1 and beam 2) shown in Figure A.2 is LC polarized and the other pair (beam 3 and beam 4) is RC polarized, the unit polarization vectors are given by,

$$\hat{e}_1 = \frac{\cos \theta \hat{x} + i\hat{y} + \sin \theta \hat{z}}{\sqrt{2}}; \quad \hat{e}_2 = \frac{\cos \theta \hat{x} + i\hat{y} - \sin \theta \hat{z}}{\sqrt{2}}; \tag{A.14}$$

$$\hat{e}_3 = \frac{\cos \theta \hat{y} - i\hat{x} + \sin \theta \hat{z}}{\sqrt{2}} \quad \& \quad \hat{e}_4 = \frac{\cos \theta \hat{y} - i\hat{x} - \sin \theta \hat{z}}{\sqrt{2}}$$

$$\hat{e}_1 \cdot \hat{e}_2^* = \frac{1 + \cos 2\theta}{2} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = \frac{1 + \cos 2\theta}{2} \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

$$\hat{e}_1 \cdot \hat{e}_3^* = \frac{\sin^2 \theta + i2\cos \theta}{2} \Rightarrow |\hat{e}_1 \cdot \hat{e}_3^*| = \frac{\sqrt{\sin^4 \theta + 4\cos^2 \theta}}{2}, \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_3^*\} = \tan^{-1} \left(\frac{2\cos \theta}{\sin^2 \theta} \right)$$

$$\hat{e}_1 \cdot \hat{e}_4^* = \frac{-\sin^2 \theta + i2\cos \theta}{2} \Rightarrow |\hat{e}_1 \cdot \hat{e}_4^*| = \frac{\sqrt{\sin^4 \theta + 4\cos^2 \theta}}{2}, \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_4^*\} = \pi + \tan^{-1} \left(\frac{2\cos \theta}{\sin^2 \theta} \right)$$

$$\hat{e}_2 \cdot \hat{e}_3^* = \frac{-\sin^2 \theta + i2\cos \theta}{2} \Rightarrow |\hat{e}_2 \cdot \hat{e}_3^*| = \frac{\sqrt{\sin^4 \theta + 4\cos^2 \theta}}{2}, \quad \& \quad \arg\{\hat{e}_2 \cdot \hat{e}_3^*\} = \pi + \tan^{-1} \left(\frac{2\cos \theta}{\sin^2 \theta} \right)$$

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$$\hat{e}_2 \cdot \hat{e}_4^* = \frac{\sin^2 \theta + i2\cos \theta}{2} \Rightarrow |\hat{e}_2 \cdot \hat{e}_4^*| = \frac{\sqrt{\sin^4 \theta + 4\cos^2 \theta}}{2}, \& \arg\{\hat{e}_2 \cdot \hat{e}_4^*\} = \tan^{-1} \left(\frac{2\cos \theta}{\sin^2 \theta} \right)$$

$$\hat{e}_3 \cdot \hat{e}_4^* = \frac{1+\cos 2\theta}{2} \Rightarrow |\hat{e}_3 \cdot \hat{e}_4^*| = \frac{1+\cos 2\theta}{2} \quad \& \quad \arg\{\hat{e}_3 \cdot \hat{e}_4^*\} = 0$$

If all the waves has the same amplitude E_0 and are in phase or are out of phase by the same value, the intensity distribution is given by,

$$\begin{aligned} I(r)_{LC-RC} = E_0^2 \bigg\{ & 2 + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta x] \\ & + \left(\frac{\sqrt{\sin^4 \theta + 4\cos^2 \theta}}{2} \right) \cos \left[k \sin \theta x - k \sin \theta y + \tan^{-1} \left(\frac{2\cos \theta}{\sin^2 \theta} \right) \right] \\ & + \left(\frac{\sqrt{\sin^4 \theta + 4\cos^2 \theta}}{2} \right) \cos \left[k \sin \theta x + k \sin \theta y \right. \\ & \left. + \pi + \tan^{-1} \left(\frac{2\cos \theta}{\sin^2 \theta} \right) \right] \\ & + \left(\frac{\sqrt{\sin^4 \theta + 4\cos^2 \theta}}{2} \right) \cos \left[-k \sin \theta x - k \sin \theta y \right. \\ & \left. + \pi + \tan^{-1} \left(\frac{2\cos \theta}{\sin^2 \theta} \right) \right] \\ & + \left(\frac{\sqrt{\sin^4 \theta + 4\cos^2 \theta}}{2} \right) \cos \left[-k \sin \theta x + k \sin \theta y + \tan^{-1} \left(\frac{2\cos \theta}{\sin^2 \theta} \right) \right] \\ & \left. + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y] \right\} \end{aligned} \tag{A.15}$$

7) Transverse electric (TE)- Left Circularly polarized (LC):

When one pair of plane waves (beam 1 and beam 2) shown in Figure A.2 is TE polarized and the other pair (beam 3 and beam 4) is LC polarized, the unit polarization vectors are given by,

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$$\hat{e}_1 = \hat{e}_2 = \hat{y} ; \quad \hat{e}_3 = \frac{\cos \theta \hat{y} + i \hat{x} + \sin \theta \hat{z}}{\sqrt{2}},$$

$$\& \hat{e}_4 = \frac{\cos \theta \hat{y} + i \hat{x} - \sin \theta \hat{z}}{\sqrt{2}}$$

$$\hat{e}_1 \cdot \hat{e}_2^* = 1 \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = 1 \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

$$\hat{e}_1 \cdot \hat{e}_3^* = \frac{\cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_3^*| = \frac{|\cos \theta|}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_3^*\} = 0$$

$$\hat{e}_1 \cdot \hat{e}_4^* = \frac{\cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_4^*| = \frac{|\cos \theta|}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_4^*\} = 0$$

$$\hat{e}_2 \cdot \hat{e}_3^* = \frac{\cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_2 \cdot \hat{e}_3^*| = \frac{|\cos \theta|}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_2 \cdot \hat{e}_3^*\} = 0$$

$$\hat{e}_2 \cdot \hat{e}_4^* = \frac{\cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_2 \cdot \hat{e}_4^*| = \frac{|\cos \theta|}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_2 \cdot \hat{e}_4^*\} = 0$$

$$\hat{e}_3 \cdot \hat{e}_4^* = \frac{1+\cos 2\theta}{2} \quad \Rightarrow |\hat{e}_3 \cdot \hat{e}_4^*| = \frac{1+\cos 2\theta}{2} \quad \& \quad \arg\{\hat{e}_3 \cdot \hat{e}_4^*\} = 0$$

If all the waves has the same amplitude E_0 and are in phase or are out of phase by the same value, the intensity distribution is given by,

$$\begin{aligned} I(r)_{TE-LC} = E_0^2 \bigg\{ & 2 + 1 \cos[2k \sin \theta x] \\ & + \frac{|\cos \theta|}{\sqrt{2}} \cos[k \sin \theta x - k \sin \theta y] \\ & + \frac{|\cos \theta|}{\sqrt{2}} \cos[k \sin \theta x + k \sin \theta y] \\ & + \frac{|\cos \theta|}{\sqrt{2}} \cos[-k \sin \theta x - k \sin \theta y] \\ & + \frac{|\cos \theta|}{\sqrt{2}} \cos[-k \sin \theta x + k \sin \theta y] \\ & + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y] \bigg\} \end{aligned} \tag{A.16}$$

8) Transverse electric (TE)-Right Circularly polarized (RC):

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When one pair of plane waves (beam 1 and beam 2) shown in Figure A.2 is TE polarized and the other pair (beam 3 and beam 4) is RC polarized, the unit polarization vectors are given by,

$$\hat{e}_1 = \hat{e}_2 = \hat{y}; \quad \hat{e}_3 = \frac{\cos \theta \hat{y} - i\hat{x} + \sin \theta \hat{z}}{\sqrt{2}},$$

$$\& \hat{e}_4 = \frac{\cos \theta \hat{y} - i\hat{x} - \sin \theta \hat{z}}{\sqrt{2}}$$

$$\hat{e}_1 \cdot \hat{e}_2^* = 1 \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = 1 \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

$$\hat{e}_1 \cdot \hat{e}_3^* = \frac{\cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_3^*| = \frac{|\cos \theta|}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_3^*\} = 0$$

$$\hat{e}_1 \cdot \hat{e}_4^* = \frac{\cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_4^*| = \frac{|\cos \theta|}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_4^*\} = 0$$

$$\hat{e}_2 \cdot \hat{e}_3^* = \frac{\cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_2 \cdot \hat{e}_3^*| = \frac{|\cos \theta|}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_2 \cdot \hat{e}_3^*\} = 0$$

$$\hat{e}_2 \cdot \hat{e}_4^* = \frac{\cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_2 \cdot \hat{e}_4^*| = \frac{|\cos \theta|}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_2 \cdot \hat{e}_4^*\} = 0$$

$$\hat{e}_3 \cdot \hat{e}_4^* = \frac{1+\cos 2\theta}{2} \quad \Rightarrow |\hat{e}_3 \cdot \hat{e}_4^*| = \frac{1+\cos 2\theta}{2} \quad \& \quad \arg\{\hat{e}_3 \cdot \hat{e}_4^*\} = 0$$

If all the waves has the same amplitude E_0 and are in phase or are out of phase by the same value, the intensity distribution is given by,

$$\begin{aligned}
 I(r)_{TE-RC} = E_0^2 \Big\{ & 2 + 1 \cos[2k \sin \theta x] \\
 & + \frac{|\cos \theta|}{\sqrt{2}} \cos[k \sin \theta x - k \sin \theta y] \\
 & + \frac{|\cos \theta|}{\sqrt{2}} \cos[k \sin \theta x + k \sin \theta y] \\
 & + \frac{|\cos \theta|}{\sqrt{2}} \cos[-k \sin \theta x - k \sin \theta y] \\
 & + \frac{|\cos \theta|}{\sqrt{2}} \cos[-k \sin \theta x + k \sin \theta y] \\
 & + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y] \Big\}
 \end{aligned} \tag{A.17}$$

9) Transverse Magnetic (TM)-Left Circularly polarized (LC):

When one pair of plane waves (beam 1 and beam 2) shown in Figure A.2 is TM polarized and the other pair (beam 3 and beam 4) is LC polarized, the unit polarization vectors are given by,

$$\hat{e}_1 = (\cos \theta \hat{x} + \sin \theta \hat{z}); \hat{e}_2 = (\cos \theta \hat{x} - \sin \theta \hat{z});$$

$$\hat{e}_3 = \frac{\cos \theta \hat{y} + i\hat{x} + \sin \theta \hat{z}}{\sqrt{2}}, \& \hat{e}_4 = \frac{\cos \theta \hat{y} + i\hat{x} - \sin \theta \hat{z}}{\sqrt{2}}$$

$$\hat{e}_1 \cdot \hat{e}_2^* = \cos 2\theta \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = |\cos 2\theta| \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0$$

$$\hat{e}_1 \cdot \hat{e}_3^* = \frac{\sin^2 \theta - i \cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_3^*| = \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_3^*\} = \frac{-\pi}{2} - \tan^{-1} \left(\frac{\sin^2 \theta}{\cos \theta} \right)$$

$$\hat{e}_1 \cdot \hat{e}_4^* = \frac{-\sin^2 \theta - i \cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_4^*| = \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_1 \cdot \hat{e}_4^*\} = -\pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right)$$

$$\hat{e}_2 \cdot \hat{e}_3^* = \frac{-\sin^2 \theta - i \cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_2 \cdot \hat{e}_3^*| = \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_2 \cdot \hat{e}_3^*\} = -\pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right)$$

$$\hat{e}_2 \cdot \hat{e}_4^* = \frac{\sin^2 \theta - i \cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_2 \cdot \hat{e}_4^*| = \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}}, \quad \& \quad \arg\{\hat{e}_2 \cdot \hat{e}_4^*\} = \frac{-\pi}{2} - \tan^{-1} \left(\frac{\sin^2 \theta}{\cos \theta} \right)$$

$$\hat{e}_3 \cdot \hat{e}_4^* = \frac{1 + \cos 2\theta}{2} \quad \Rightarrow |\hat{e}_3 \cdot \hat{e}_4^*| = \frac{1 + \cos 2\theta}{2} \quad \& \quad \arg\{\hat{e}_3 \cdot \hat{e}_4^*\} = 0$$

If all the waves has the same amplitude E_0 and are in phase or are out of phase by the same value, the intensity distribution is given by,

$$\begin{aligned}
 I(r)_{TM-LC} = E_0^2 \Big\{ & 2 + |\cos 2\theta| \cos[2k \sin \theta x] \\
 & + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[k \sin \theta x - k \sin \theta y + \frac{-\pi}{2} - \tan^{-1} \left(\frac{\sin^2 \theta}{\cos \theta} \right) \right] \\
 & + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[k \sin \theta x + k \sin \theta y - \pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \right] \\
 & + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[-k \sin \theta x - k \sin \theta y - \pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \right] \\
 & + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[-k \sin \theta x + k \sin \theta y + \frac{-\pi}{2} - \tan^{-1} \left(\frac{\sin^2 \theta}{\cos \theta} \right) \right] \\
 & + \left(\frac{1 + \cos 2\theta}{2} \right) \cos[2k \sin \theta y] \Big\}
 \end{aligned} \tag{A.18}$$

10) Transverse Magnetic (TM)-Right Circularly polarized (RC):

When one pair of plane waves (beam 1 and beam 2) shown in Figure A.2 is TM polarized and the other pair (beam 3 and beam 4) is RC polarized, the unit polarization vectors are given by,

$$\begin{aligned}
 \hat{e}_1 &= (\cos \theta \hat{x} + \sin \theta \hat{z}); \hat{e}_2 = (\cos \theta \hat{x} - \sin \theta \hat{z}); \\
 \hat{e}_3 &= \frac{\cos \theta \hat{y} - i\hat{x} + \sin \theta \hat{z}}{\sqrt{2}}, \& \hat{e}_4 = \frac{\cos \theta \hat{y} - i\hat{x} - \sin \theta \hat{z}}{\sqrt{2}} \\
 \hat{e}_1 \cdot \hat{e}_2^* &= \cos 2\theta \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_2^*| = |\cos 2\theta| \quad \& \quad arg\{\hat{e}_1 \cdot \hat{e}_2^*\} = 0 \\
 \hat{e}_1 \cdot \hat{e}_3^* &= \frac{\sin^2 \theta + i\cos \theta}{\sqrt{2}} \quad \Rightarrow |\hat{e}_1 \cdot \hat{e}_3^*| = \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}}, \quad \& \quad arg\{\hat{e}_1 \cdot \hat{e}_3^*\} = \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \\
 \hat{e}_1 \cdot \hat{e}_4^* &= \frac{-\sin^2 \theta + i\cos \theta}{\sqrt{2}} \Rightarrow |\hat{e}_1 \cdot \hat{e}_4^*| = \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}}, \quad \& \quad arg\{\hat{e}_1 \cdot \hat{e}_4^*\} = \pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right) \\
 \hat{e}_2 \cdot \hat{e}_3^* &= \frac{-\sin^2 \theta + i\cos \theta}{\sqrt{2}} \Rightarrow |\hat{e}_2 \cdot \hat{e}_3^*| = \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}}, \quad \& \quad arg\{\hat{e}_2 \cdot \hat{e}_3^*\} = \pi + \tan^{-1} \left(\frac{\cos \theta}{\sin^2 \theta} \right)
 \end{aligned}$$

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$$\begin{aligned}\hat{e}_2 \cdot \hat{e}_4^* &= \frac{\sin^2 \theta + i \cos \theta}{\sqrt{2}} \Rightarrow |\hat{e}_2 \cdot \hat{e}_4^*| = \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}}, \quad \& \arg\{\hat{e}_2 \cdot \hat{e}_4^*\} = \tan^{-1}\left(\frac{\cos \theta}{\sin^2 \theta}\right) \\ \hat{e}_3 \cdot \hat{e}_4^* &= \frac{1 + \cos 2\theta}{2} \Rightarrow |\hat{e}_3 \cdot \hat{e}_4^*| = \frac{1 + \cos 2\theta}{2} \quad \& \arg\{\hat{e}_3 \cdot \hat{e}_4^*\} = 0\end{aligned}$$

If all the waves has the same amplitude E_0 and are in phase or are out of phase by the same value, the intensity distribution is given by,

$$\begin{aligned}I(r)_{TM-RC} &= E_0^2 \left\{ 2 + |\cos 2\theta| \cos[2k \sin \theta x] \right. \\ &\quad + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[k \sin \theta x - k \sin \theta y + \tan^{-1}\left(\frac{\cos \theta}{\sin^2 \theta}\right) \right] \\ &\quad + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[k \sin \theta x + k \sin \theta y + \pi + \tan^{-1}\left(\frac{\cos \theta}{\sin^2 \theta}\right) \right] \\ &\quad + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[-k \sin \theta x - k \sin \theta y + \pi + \tan^{-1}\left(\frac{\cos \theta}{\sin^2 \theta}\right) \right] \\ &\quad + \frac{\sqrt{\sin^4 \theta + \cos^2 \theta}}{\sqrt{2}} \cos \left[-k \sin \theta x + k \sin \theta y + \tan^{-1}\left(\frac{\cos \theta}{\sin^2 \theta}\right) \right] \\ &\quad \left. + \left(\frac{1 + \cos 2\theta}{2}\right) \cos[2k \sin \theta y] \right\} \tag{A.19}\end{aligned}$$

A.3 Five-beam interference

In this section the derivation of intensity distribution for five-beam interference where the central beam is left circularly polarized and the two sets of orthogonal beams shown in Figure A.3 are TE-TE polarized and TM-TM polarized are discussed in detail.

Figure A.3 Five-beam interference configuration.

The electric field vectors of the five beams in this case are given by,

$$E_1(r, t) = E_1 \hat{e}_1 \cos [(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_1]$$

$$E_2(r, t) = E_2 \hat{e}_2 \cos [-(k \sin \theta)x - (k \cos \theta)z - \omega t + \varphi_2]$$

$$E_3(r, t) = E_3 \hat{e}_3 \cos [(k \sin \theta)y - (k \cos \theta)z - \omega t + \varphi_3]$$

$$E_4(r, t) = E_4 \hat{e}_4 \cos [-(k \sin \theta)y - (k \cos \theta)z - \omega t + \varphi_4]$$

$$E_5(r, t) = E_5 \hat{e}_5 \cos [-kz - \omega t + \varphi_5]$$

The time independent intensity distribution for five-beam interference for the beam configuration shown in Figure A.3 is obtained by using above equations and Eq. (5.2). It is given by,

$$\begin{aligned}
 I(r)_{5B} = & \frac{(E_1^2 + E_2^2 + E_3^2 + E_4^2 + E_5^2)}{2} + E_1 E_2 |\hat{e}_1 \cdot \hat{e}_2^*| \cos[2k \sin \theta x + (\varphi_1 - \varphi_2) + \delta_{12}] \\
 & + E_1 E_3 |\hat{e}_1 \cdot \hat{e}_3^*| \cos[k \sin \theta x - k \sin \theta y + (\varphi_1 - \varphi_3) + \delta_{13}] \\
 & + E_1 E_4 |\hat{e}_1 \cdot \hat{e}_4^*| \cos[k \sin \theta x + k \sin \theta y + (\varphi_1 - \varphi_4) + \delta_{14}] \\
 & + E_1 E_5 |\hat{e}_1 \cdot \hat{e}_5^*| \cos[k \sin \theta x - k \cos \theta z + kz + (\varphi_1 - \varphi_5) + \delta_{15}] \\
 & + E_2 E_3 |\hat{e}_2 \cdot \hat{e}_3^*| \cos[-k \sin \theta x - k \sin \theta y + (\varphi_2 - \varphi_3) + \delta_{23}] \\
 & + E_2 E_4 |\hat{e}_2 \cdot \hat{e}_4^*| \cos[-k \sin \theta x + k \sin \theta y + (\varphi_2 - \varphi_4) + \delta_{24}] \\
 & + E_2 E_5 |\hat{e}_2 \cdot \hat{e}_5^*| \cos[-k \sin \theta x - k \cos \theta z + kz + (\varphi_2 - \varphi_5) + \delta_{25}] \\
 & + E_3 E_4 |\hat{e}_3 \cdot \hat{e}_4^*| \cos[2k \sin \theta y + (\varphi_3 - \varphi_4) + \delta_{34}] \\
 & + E_3 E_5 |\hat{e}_3 \cdot \hat{e}_5^*| \cos[k \sin \theta y - k \cos \theta z + kz + (\varphi_3 - \varphi_5) + \delta_{35}] \\
 & + E_4 E_5 |\hat{e}_4 \cdot \hat{e}_5^*| \cos[-k \sin \theta y - k \cos \theta z + kz + (\varphi_4 - \varphi_5) + \delta_{45}]
 \end{aligned} \tag{A.20}$$

If the fifth beam is circularly polarized, the polarization vectors can be written as,

$$\hat{e}_{L5} = \frac{\hat{x} + i\hat{y}}{\sqrt{2}} \text{ (left circular central beam)} \& \hat{e}_{R5} = \frac{\hat{x} - i\hat{y}}{\sqrt{2}} \text{ (right circular central beam)}.$$

The unit polarization vectors when the central beam is left circularly polarized and the four other beams are in TE-TE polarized state are given by,

$$\hat{e}_1 = \hat{e}_2 = \hat{y} \quad , \quad \hat{e}_3 = \hat{e}_4 = \hat{x} \quad \& \quad \hat{e}_{L5} = \frac{\hat{x} - i\hat{y}}{\sqrt{2}}$$

Calculating the argument and magnitude of the function $(\hat{e}_i \cdot \hat{e}_j^*)$,

$$|\hat{e}_1 \cdot \hat{e}_2^*| = |\hat{y} \cdot \hat{y}| = 1$$

$$|\hat{e}_3 \cdot \hat{e}_4^*| = |\hat{y} \cdot \hat{y}| = 1$$

$$|\hat{e}_1 \cdot \hat{e}_3^*| = |\hat{e}_2 \cdot \hat{e}_3^*| = |\hat{e}_1 \cdot \hat{e}_4^*| = |\hat{e}_2 \cdot \hat{e}_4^*| = 0$$

$$|\hat{e}_1 \cdot \hat{e}_{L5}^*| = |\hat{e}_2 \cdot \hat{e}_{L5}^*| = \left| \hat{y} \cdot \frac{(\hat{x} - i\hat{y})}{\sqrt{2}} \right| = \frac{1}{\sqrt{2}}$$

$$|\hat{e}_3 \cdot \hat{e}_{L5}^*| = |\hat{e}_4 \cdot \hat{e}_{L5}^*| = \left| \hat{x} \cdot \frac{(\hat{x} - i\hat{y})}{\sqrt{2}} \right| = \frac{1}{\sqrt{2}}$$

And argument $\delta_{ij} = \arg\{\hat{e}_i \cdot \hat{e}_j^*\}$

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$$\delta_{12} = \delta_{13} = \delta_{14} = \delta_{23} = \delta_{24} = \delta_{34} = 0$$

$$\Rightarrow \delta_{15} = \delta_{25} = \arg\{\hat{e}_1 \cdot \hat{e}_{L5}^*\} = \arg\left\{\hat{y} \cdot \frac{\hat{x}-i\hat{y}}{\sqrt{2}}\right\} = \left(-\frac{\pi}{2}\right)$$

$$\Rightarrow \delta_{35} = \delta_{45} = \arg\left\{\hat{x} \cdot \frac{\hat{x}-i\hat{y}}{\sqrt{2}}\right\} = \arg\left\{\frac{1}{\sqrt{2}}\right\} = 0$$

The intensity distribution in this case where the central beam is left circularly polarized and the four orthogonal plane waves are TE-TE polarized and has the same phase constant value, is obtained by substituting the above values of argument and magnitude of $(\hat{e}_i \cdot \hat{e}_j^*)$, in Eq. (5.20)

$$\begin{aligned} I(r)_{5B \text{ TE-TE}} = E_0^2 \bigg\{ & \frac{5}{2} + \cos[2k \sin \theta x] + \cos[2k \sin \theta y] \\ & + \frac{2}{\sqrt{2}} (\sin[-k \cos \theta z + kz]) (\cos[k \sin \theta x]) \\ & + \frac{2}{\sqrt{2}} (\cos[-k \cos \theta z + kz]) (\cos[k \sin \theta y]) \bigg\} \end{aligned} \quad \text{A.21}$$

The above equation is valid for the case where the central beam is right circularly polarized too.

Now consider the case where the four plane waves (beam 1 to 4) shown in Figure A.3 are TM-TM polarized. The polarization vectors are given by

$$\begin{aligned} \hat{e}_1 &= (\cos \theta \hat{x} + \sin \theta \hat{z}), \quad \hat{e}_2 = (\cos \theta \hat{x} - \sin \theta \hat{z}), \quad \hat{e}_3 = (\cos \theta \hat{y} + \sin \theta \hat{z}), \quad \hat{e}_4 = \\ &(\cos \theta \hat{y} - \sin \theta \hat{z}) \quad \& \quad \hat{e}_{L5} = \frac{\hat{x}-i\hat{y}}{\sqrt{2}} \end{aligned}$$

Calculating the argument and magnitude of the function $(\hat{e}_i \cdot \hat{e}_j^*)$,

$$|\hat{e}_1 \cdot \hat{e}_2^*| = |(\cos \theta \hat{x} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{x} - \sin \theta \hat{z})| = \cos 2\theta$$

$$|\hat{e}_3 \cdot \hat{e}_4^*| = |(\cos \theta \hat{y} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| = \cos 2\theta$$

$$|\hat{e}_1 \cdot \hat{e}_3^*| = |(\cos \theta \hat{x} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} + \sin \theta \hat{z})| = \sin^2 \theta$$

$$|\hat{e}_2 \cdot \hat{e}_3^*| = |(\cos \theta \hat{x} - \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} + \sin \theta \hat{z})| = \sin^2 \theta$$

Appendix

$$|\hat{e}_1 \cdot \hat{e}_4^*| = |(\cos \theta \hat{x} + \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| = \sin^2 \theta$$

$$|\hat{e}_2 \cdot \hat{e}_4^*| = |(\cos \theta \hat{x} - \sin \theta \hat{z}) \cdot (\cos \theta \hat{y} - \sin \theta \hat{z})| = \sin^2 \theta$$

$$|\hat{e}_1 \cdot \hat{e}_{L5}^*| = \left| (\cos \theta \hat{x} + \sin \theta \hat{z}) \cdot \frac{(\hat{x} - i\hat{y})}{\sqrt{2}} \right| = \frac{\cos \theta}{\sqrt{2}}$$

$$|\hat{e}_2 \cdot \hat{e}_{L5}^*| = \left| (\cos \theta \hat{x} - \sin \theta \hat{z}) \cdot \frac{(\hat{x} - i\hat{y})}{\sqrt{2}} \right| = \frac{\cos \theta}{\sqrt{2}}$$

$$|\hat{e}_3 \cdot \hat{e}_{L5}^*| = \left| (\cos \theta \hat{y} + \sin \theta \hat{z}) \cdot \frac{(\hat{x} - i\hat{y})}{\sqrt{2}} \right| = \frac{\cos \theta}{\sqrt{2}}$$

$$|\hat{e}_4 \cdot \hat{e}_{L5}^*| = \left| (\cos \theta \hat{y} - \sin \theta \hat{z}) \cdot \frac{(\hat{x} - i\hat{y})}{\sqrt{2}} \right| = \frac{\cos \theta}{\sqrt{2}}$$

And argument $\delta_{ij} = \arg\{\hat{e}_i \cdot \hat{e}_j^*\}$

$$\delta_{12} = \delta_{13} = \delta_{14} = \delta_{23} = \delta_{24} = \delta_{34} = 0$$

$$\Rightarrow \delta_{15} = \delta_{25} = 0$$

$$\Rightarrow \delta_{35} = \delta_{45} = \left(-\frac{\pi}{2}\right)$$

The intensity distribution in this case where the central beam is left circularly polarized and the four orthogonal plane waves are TE-TE polarized and has the same phase constant value, is obtained by substituting the above values of argument and magnitude of $(\hat{e}_i \cdot \hat{e}_j^*)$ in Eq. (5.20).

$$\begin{aligned} I(r)_{5B\,TM-TM} = E_0^2 \left\{ \frac{5}{2} + \cos 2\theta (\cos[2k \sin \theta x] + \cos[2k \sin \theta y]) \right. \\ + 4 \sin^2 \theta (\cos[2k \sin \theta x])(\cos[2k \sin \theta y]) \\ + \frac{2 \cos \theta}{\sqrt{2}} (\cos[kz - k \cos \theta z]) (\cos[k \sin \theta x]) \\ \left. + \frac{2 \cos \theta}{\sqrt{2}} (\sin[kz - k \cos \theta z]) (\cos[k \sin \theta y]) \right\} \end{aligned} \quad A.22$$

Appendix B: Dye assisted enhanced transmission in metallic masks for lithography

A detail investigation report of the concept and methodology for dye based plasmon enhancement in embedded mask based near field lithographic configuration, proposed in Chapter 6 is presented here. The design aspects such as thickness, diameter and type of dye material to maximize the transmission are analyzed and are discussed in detail.

The schematic diagram in Figure 6.1 shows the proposed configuration of the plasmonic mask used in this research. As mentioned in Chapter 6, two types of plasmonic samples as shown in Figure 6.1: one with bare PMMA film and the other with dye embedded PMMA are employed to investigate the E-field transmission enhancement. The details of numerical simulation and results are discussed here.

B.1 Numerical Simulation and Discussion

Surface plasmon enhancement in a perforated Al mask is numerically demonstrated using finite-difference-time-domain (FDTD) method. The commercially available FDTD from Lumerical solutions is employed for all simulations. Different set of simulations were carried out in order to determine transmission wavelength for aluminium, to optimize aluminium thickness, to

study transmission modes and finally to compare the E field transmission through perforated masks with and without dye medium. All simulations employ periodic boundary condition along x and z directions to have the effects of infinite biperiodic structure. And perfectly matched layer (PML) is considered along the y-direction.

Initially, to determine the wavelength at which maximum transmission is achievable, aluminium structure with periodic holes of diameter 40 nm at a pitch size of 170 nm and with thickness of 100 nm is employed. Figure B.1 represents the transmission intensity as the function of excitation wavelength ($T(\lambda)$). It shows high transmission at around 365 nm wavelength. Considering this and the fact that i-line laser is commonly available for future experimental work, 365 nm is employed as excitation wavelength.

Figure B.1 Normalized transmission spectrum of Al film with 2D biperiodic circular apertures. The thickness, size and period of the arrays are 100 nm, 40 nm and 170 nm respectively.

Appendix

Analysis of the wavelength dependent transmission function of the Al film shows that besides the 365 nm mode, two other modes at 435 nm and 485 nm respectively will be excited. Figure B.2 represents the wavelength dependent transmission $T(\lambda)$ function of 100 nm thick Aluminum film. The transmission spectrum exhibits a prominent peak at 485 nm. Contribution of this mode to total transmission through the sub-wavelength aperture is 10 times compared to that of the 365 mode. Two types of SPs will be supported by the perforated metal film: localized surface Plasmon polaritons (SPs) and long range SP (LSP). Localized SP can be presumed due to a defect state (a punctured hole in a thin film). On the other hand the frequency of long range SP is determined by the array period ($2\pi/L$). The transmission spectra of such arrays are characterized by resonances which are related to a (i,j) scattering order of the array. The wavelength of the transmission peak can be given approximately by the surface Plasmon dispersion for a smooth film.

$$\lambda_{(i,j)} = \frac{p}{\sqrt{i^2 + j^2}} \sqrt{\frac{\epsilon_m \cdot \epsilon_d}{\epsilon_m + \epsilon_d}} \quad \text{B.1}$$

Where ϵ_m and ϵ_d are respectively the dielectric constants of the metal and dielectric (air), P the period of the array and (i,j) represents the scattering order of the array. The peak at 365 nm corresponds to second harmonic of TE (11) mode. This plasmon mode does not change with the thickness. Besides this, standing electromagnetic fields will also be excited inside the nanohole [131]. These cavities resonance modes are entirely localize in nature and the no of resonance mode excited will depend on thickness of the

metal film. Hence each air nanohole can be considered as truncated circular waveguide with both ends open to free space. Totally two modes are excited for the film of thickness 100 nm. Al film of thickness 100 nm is considered for all the following numerical simulation as it posses enhanced transmission.

Figure B.2 Normalized transmission spectral intensity for the perforated Al film of thickness 100 nm.

Figure B.3 (a) Electric field distribution, and (b) its magnitude at the exit plane of the perforated metal mask of type I configuration (Figure 6.1 (a)) which employs no gain medium. The thickness, size and period of the arrays are 100 nm, 40 nm and 170 nm respectively.

Appendix

To study transmission enhancement, numerical analysis employing perforated masks without dye medium is carried out first. Figure B.3 represents the electric field distribution and its magnitude at the exit plane of the perforated metal mask of type I (Figure 6.1(a)) which employs no gain medium. It represents the normalized field with respect to input excitation beam. The thickness, size and period of the arrays are 100 nm, 40 nm and 170 nm respectively.

Generally perforated plasmonic sample will have low transmission as it suffer from SP propagation losses associated with the metal. Moreover, both sub-wavelength resolution (*RES*) and large depth of focus (*DOF*) cannot be achieved simultaneously due to inverse dependent nature of these two parameters.

$$DOF = k_2 \frac{\lambda}{NA^2} = (k_2/k_1) \frac{RES^2}{\lambda} \quad \text{B.2}$$

Where the quantities k_2 and k_1 are process-dependent constants. *DOF* can be enhanced by increasing the total transmission through sub-wavelength apertures by exciting two plasmonic modes 365 nm and 485 nm simultaneously. This can be realized by employing dye molecules with excitation and emission band widths lies around 365 nm and 485 nm respectively.

In addition total transmission can be further improved by reducing propagation losses of the SP. The damping of SP is the characterized by an exponential decay $e^{-\gamma x}$ of its intensity away from the metal surface. The damping constant γ contains contributions due to dissipation in the metal γ_m and to radiative loss γ_r . The radiative loss is due to fact that plasmon emits as light.

$$\gamma_m = \frac{2\pi}{\lambda_e} \frac{\varepsilon'_m}{(\varepsilon'_m)^2} \left(\frac{\varepsilon'_m \varepsilon'_d}{\varepsilon'_m + \varepsilon'_d} \right)^{3/2} \quad \text{B.3}$$

where ε'_m and ε''_m denote the real and imaginary part of the dielectric constant of the metal, where ε'_d is the real part of polymer matrix (PMMA). The refractive index of PMMA lies in the range 1.51-1.53. The dielectric constants of aluminium at the wavelengths 365 nm and 485 nm are $-19.42 + 3.606i$ and $-32.954 + 9.197i$ respectively. Using these values, the obtained damping loss γ_m is about $3.335 \times 10^5 \text{m}^{-1}$ at 485 nm. This loss can overcome by gain medium such as dye with optimum concentration [122-124]. PMMA matrix doped with dye molecules is considered as the gain medium. The dye molecules are chosen to have large absorption and emission cross-sections at excitation and emission wavelengths of interest. Besides this, the emission band width should have the wave component of the 485 resonance mode. Coumarin 102 is selected for the present simulation as it has emission band around 485 nm for excitation at 365 nm wavelength [125-127]. The gain γ_g needed to compensate for the SP loss is given by

$$\gamma_g = \frac{2\pi}{\lambda_e} \frac{\varepsilon'_d}{(\varepsilon'_d)^2} \left(\frac{\varepsilon'_m \varepsilon'_d}{\varepsilon'_m + \varepsilon'_d} \right)^{3/2} \quad \text{B.4}$$

Here ε''_d is the imaginary part of the dielectric constant of dye-PMMA matrix which is the function of dye concentration (N) and emission cross section (σ_e). It is assumed that the dye has a four level energy system and the fluorescence transition occurs between the energy levels 3 and 2. For the system

attained population inversion, dye medium acquires an imaginary part of the dielectric constant at the emission wavelength (λ_e) which is given by

$$\varepsilon_d'' = \frac{n\lambda_e}{2\pi} (N_3 - N_2)\sigma_e = \frac{n\lambda_e}{2\pi} N\sigma_e \quad \text{B.5}$$

Where, n is the refractive index of gain medium (PMMA), and N_2 & N_3 represent the dye concentrations at the energy level 2 and 3 respectively. For maximum population inversion: $N_3 \gg N_2$.

Assuming that the concentrations at all other energy levels are negligible, $(N_3 - N_2) \approx N$ (total concentration of the system). The calculated emission cross section of the coumarin 102 is $\sigma_e \approx 7 \times 10^{-17} \text{cm}^2$. Using this value of emission cross section, the concentration of dye molecules is obtained which is approximately $3.5 \times 10^{17} \text{cm}^3$ to get the gain coefficient. If the gain coefficient is less than unity, SP will still be lossy but the penetration depth will increase accordingly. In such higher concentration, each dye molecule in the PMMA matrix can be modelled as an isolated classical dipole [128, 129]. The lifetime of the dipole is influenced by the presence of the metal film and is significantly affected by the dipole's position and dipole-moment orientation respectively. The position-dependent decay rate normalized to the decay rate far from the metal film can be written as,

$$\tau(z) = \tau_0 \left[\frac{2}{3}\gamma^h(y) + \frac{1}{3}\gamma^v(y) \right]^{-1} \quad \text{B.6}$$

Where γ^h and γ^v represent normalized decay rate of horizontally and vertically oriented dipole molecules respectively.

The nature of SP enhancement is studied by placing a radiating coumarin 102 dye near a metal nanoparticle. The simulation has been done by considering an electric dipole to mimics a Coumarin 102 dye molecule which has an emission at 485 nm. This emission wavelength just matches with the wavelength of the dominant 485 nm resonance mode. Two crossed polarized dipoles, 66% ($2/3$) contributions from the horizontal dipole and 33% ($1/3$) contributions from the vertical dipole, were considered in the present simulation. The crossed dipole is positioned above the metal surface and centred between the holes. Figure B.4 envisages the delineation of transmission intensity as the function of the dipole distance with respect to metal surface. The increase in transmission is observed only for those molecules residing within 40 nm from the metal surface. So the crossed dipole is positioned at about 65 nm from the hole centre and 40 nm away from the metal surface in simulations.

The Figure B.5 represents electric field distribution and its magnitude at the exit medium of the perforated metal mask in the presence of gain medium (dye). It represents the output E-field normalised with respect to excitation field. Extraordinary transmission of about 14.5 fold enhancements is observed due to the transfer of light from main 485 nm resonance mode excited inside the cavity.

Figure B.4 Normalized transmission through the perforation as the function of position of dye molecule with respect to the metal plane.

Figure B.5 (a) Electric field distribution, and (b) its magnitude at the exit plane of the perforated metal mask, type II configuration (Figure 6.1(b)), in the presence of gain medium. The thickness, size and period of the arrays are 100 nm, 40 nm and 170 nm respectively.

Figure B.6 gives the transmissions of the perforated film due to (1) only dipole excitation, (2) direct 485 nm light excitation, (3) both dipole and 365 nm excitation and (4) both 365 and 485 nm excitation. The excitation at 365 nm in

the presence of dipole shows maximum transmission than all other excitations. Bare dipole excitation shows about 10 times transmission than excitation by direct 485 nm light beam. This could be due to strong fluorescence quenching as the consequence of emission of plasmon directly to the metal film by the dye molecule [128]. The dye molecule in the presence of the metal film will be influenced by four decay channels [132] that arise due to coupling of the dipole to (i) Long range surface plasmon polariton and (ii) Short range surface plasmon polariton modes- in both cases, the dipole excites a SPP instead of a photon emission, (iii) coupling to electron-hole (E-H) pairs in the metal film by dipole-dipole interaction, and (iv) coupling to the radiation modes of the structure. The graph in Figure B.6 represents the output E-field normalized with respect to input excitation beam. The excitation at 365 nm in the presence of dipole shows much higher transmission compared to others. As discussed in Chapter 6, this could be due to the strong fluorescence quenching as the consequence of emission of plasmon directly to the metal film by the dye.

Figure B.6 The transmissions of the perforated film due to (1) only dipole excitation, (2) direct 485 nm light excitation, (3) both dipole and 365 nm excitation and (4) both 365 nm and 485 nm excitation.

B.3 Summary

The proposed concept of enhancement of the plasmon propagation in mask based surface plasmon lithography using dye medium has been successfully studied using numerical simulations. The gain medium assists the enhanced SP propagation by compensating intrinsic loss associated with metal, thereby exciting 485 nm resonance mode inside the cavity. A 14.5 fold of field enhancement in the presence of PMMA/dye (Coumarin 102) medium compared to bare PMMA is observed. This excited mode will further improve the total transmission of the sub-wavelength apertures. The enhancement of surface plasmons on the Aluminium film is due to the plasmon resonance frequency is

closely matched with the emission wavelength of the dye. The study reveals that the field enhancement with gain medium would offer the opportunity to increase the penetration depth in photoresist during lithography, which in turn would improve the aspect ratio of the fabricated features.

Publications

Journal Publications

1. R. Sidharthan, F. Chollet, and V. M. Murukeshan, "Periodic Patterning Using Multi-Facet Prism Based Laser Interference Lithography", *Laser Physics*, 2009, Vol. 19, No. 3, pp. 505–510.
2. R. Sidharthan and V. M. Murukeshan, "Pattern definition employing prism based Deep UV lithography", *Micro & Nano letters*, 2011, Vol. 6, Iss. 3, pp. 109-112.
3. K. Sathiyamoorthy, R. Sidharthan, K. V Sreekanth and V.M Murukeshan, "Dye assisted enhanced transmission in near field optical lithography". *Optics Communications* 2010, 283(24), 5245-9.
4. K. V Sreekanth, R. Sidharthan and V.M Murukeshan, 2011 "Gap modes assisted enhanced broadband light absorption in plasmonic thin film solar cell" *Journal of Applied Physics* 110 033107-5.
5. R. Sidharthan, V. M. Murukeshan, and K. Sathiyamoorthy, "Sub-60 nm Periodic Grating Feature Patterning by Immersion Based 364 nm Laser Interference," *Journal of Nanoscience and Nanotechnology* 12, 6428-6431 (2012).
6. R. Sidharthan, and V. M. Murukeshan, "Periodic feature patterning by lens based solid immersion multiple beam laser interference lithography," *Laser Physics Letters* 9, 691 (2012).
7. R. Sidharthan, and V. M. Murukeshan, "Improved light Absorption in thin film solar cell using combination of gap modes and grating back reflector", (Under Review).
8. R. Sidharthan, V. M. Murukeshan, M. Kumar, J. Joseph," β -tin type structure formation and its transformation to diamond cubic type structure using a five-beam interference configuration", (Under Review).

Conference Publications

1. R. Sidharthan, Franck Chollet and V.M.Murukeshan, “Nanofabrication of periodic features using Biprism based interference lithography”, Cochin nano2009 (Won 3rd prize for best paper presentation).
2. R. Sidharthan, K. Sathiyamoorthy and V. M. Murukeshan, “Large area lithography based on template assisted microspheres”, COLA 2009, Singapore.
3. K. Sathiyamoorthy, R. Sidharthan, Sreekanth K. V and V. M. Murukeshan, “Enhancement of Surface Plasmon by Gain Assisted Medium”, ICMAT 2009, Singapore.
4. R. Sidharthan and V. M. Murukeshan, “Nano-scale patterning using pyramidal prism based wavefront interference lithography”, ICOPEN2011, Singapore.
5. R. Sidharthan, K. Sathiyamoorthy, and V. M. Murukeshan, “Nanoring patterning using surface plasmon assisted photolithography”, M. F. Costa, ed. (SPIE, Braga, Portugal, 2011), pp. 80011P-80018.

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