Optical Nano-antennas

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Abstract

Wavelength division multiplexing/demultiplexing in radio frequency communication networks and fibre-optic networks requires sophisticated hardware. One of the challenges in designing subwavelength optical networks is the simplification of the design. When considering the size reduction and feasibility, a design based on fewer constituent elements is highly desirable. Ultra-compact wavelength multiplexers/demultiplexers will play a vital role in all-optical communication networks of the future. The challenge is to use the simplest and fewest antenna elements to achieve a functionality as sophisticated as a wavelength demultiplexer. How a pair of metallic nano-rods with different lengths can achieve such complex functionality is investigated both numerically and analytically. The study is extended to a single asymmetric cross-shaped nanoparticle.

Holes perforated in metallic films are considered as slot antennas and are of particular interest in the design of miniaturised colour filters, refractive index sensors, biosensors, wave plates and other miniaturised optical elements. As part of this project, two different techniques of fabricating arrays of holes were employed. The quality and accuracy of fabricated arrays using electron ion beam lithography were compared to those fabricated with focused ion beam lithography. In both cases, periodicities of the arrays were fabricated accurately, whereas the aperture dimensions deviated from those intended. Some authors base their designs on resonant localized surface plasmons associated with the shape resonances of the apertures that are highly susceptible to geometrical defects arising during the fabrication. Detuning such shape resonances to achieve a certain phase requirements, therefore, are also prone to fabrication errors. Arrays of circular holes on the other hand are simpler to fabricate with accurate periodicities. A novel approach in designing miniaturised wave plates based purely on the surface plasmon polaritons that depend solely on the detuned periodicities of the array is proposed with simulation agreeing the experiment. A novel technique in converting a hole array (that is supported on a glass substrate) into a free standing array is also applied to the abovementioned wave plate. Experimental results of such array showed the device acts as a highly efficient refractive index sensor as well as a tuneable quarter wave plate. Prior to this, however, a simple analytical model that explains the
origin of the peaks and phase relations in the spectral line associated with hole arrays was developed. The optical responses of arrays based on surface plasmon polaritons are highly sensitive to the angle of incidence. Condition for which such arrays become desensitized to the incident angle was investigate and shown that despite claims regarding depolarization observed in hole arrays, it is possible to steadily control their polarization response.

Bullseye antennas with symmetric cross-shaped apertures at their centre are investigated and designed for shaping the radiation pattern of the transmitted light in the far-field. Designs were carried out using finite element methods and experimental data agree with those obtained numerically. A bullseye structure with an asymmetric cross-shaped aperture was also modelled, fabricated and characterised. Experimental data confirms that it is possible to control the radiation pattern as well as the polarization state of the transmitted light when tailoring the surface surrounding an asymmetric cross-shaped aperture with concentric circular corrugations. Another benefit of a resonant slot antenna is its ability to interact with a nearby quantum emitter. Strong and highly localized fields confined to the cavity can interact with those of a quantum emitter positioned inside it. In the strong coupling regime, this leads to an increase in the radiated power by the system as a whole. Although the enhancement to the radiative decay rate is not strictly associated with the antenna theory, one can draw an analogy between the antenna’s gain and the increase in the radiated power observed in a plasmonic antenna when integrated with a quantum emitter. Numerical solutions showed a high yield in the scattered power with a highly directional radiation pattern when a nan-diamond is positioned inside a cross-shaped aperture in a bullseye setting.

The integration process of such nanoparticles with subwavelength apertures is cumbersome at the present. A novel approach is therefore proposed in coupling the emission of a NV- colour centre to the plasmonic surface modes based on the utilization of diamond substrates.

The influence of the film thickness and the substrate’s refractive index on the surface modes at the superstrate is an important study as the interaction between the holes in a slot antenna array is influenced by these modes. The investigation of such effects, however, is not possible in a period array of holes due to the convolution of the surface plasmons with the Bloch waves. Studies of the surface plasmon polaritons launched by
an isolated sub-wavelength slit perforating a metallic thin film showed the existence of a non-travelling interference envelope when the thickness of the film becomes comparable to the skin depth.
Declaration

This is to certify that:

(i) The thesis comprises only my original work towards the PhD except where indicated in the Preface

(ii) Due acknowledgement has been made in the text to all other material used

(iii) The thesis is less than 100,000 words in length, exclusive of tables, maps, bibliographies and appendices

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Amir Djalalian-Assl
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To those who are no longer with us.
## Contents

1. Literature Review ............................................................................................................ 1  
   1.1. Surface Plasmon Polaritons .................................................................................... 3  
   1.2. Localized Surface Plasmon (Resonance) .............................................................. 3  
   1.3. Optical Antennas ..................................................................................................... 6  
      1.3.1. Metallic Nanorods and Nanoparticles ............................................................ 8  
      1.3.2. Subwavelength Metallic Apertures .................................................................. 13  
      1.3.3. Radiative Decay Rate ..................................................................................... 16  
      1.3.4. Surface Corrugations ..................................................................................... 17  
   1.4. Plasmonic Metamaterial ........................................................................................... 23  
   1.5. Objectives ............................................................................................................... 26  
2. Concepts .......................................................................................................................... 28  
   2.1. Time-Harmonic Maxwell’s Equations ..................................................................... 28  
   2.2. Poynting Theorem .................................................................................................. 32  
   2.3. Radiative Decay Rate Enhancement ....................................................................... 33  
   2.4. Surface Plasmon Polaritons .................................................................................... 35  
   2.5. Surface Plasmon Polaritons and Bloch Waves ...................................................... 39  
   2.6. Finite Element Method ........................................................................................... 40  
3. Experimental Setups ......................................................................................................... 43  
   3.1. Electron Beam Lithography ..................................................................................... 43  
      3.1.1. The Instrument ............................................................................................... 44  
      3.1.2. Process .......................................................................................................... 44  
   3.2. Focused Ion Beam .................................................................................................... 46  
      3.2.1. The Instrument ............................................................................................... 47
3.2.2. Bitmap File Preparation ................................................................. 49
3.3. Confocal Microscope (Lifetime and Depth of Field Measurements) ........ 50
3.4. Inverted Microscope (Spectroscopy and Polarization Measurements) ... 53
4. Wavelength Dependent Optical Steering ............................................. 55
  4.1. Theory ............................................................................................. 55
  4.2. T configuration: Detached Two Nanoantennas ................................. 59
  4.3. Cross configuration: Single Nanoantenna ........................................ 61
  4.4. Conclusion ..................................................................................... 63
5. Fabrication of Resonant Aperture Antenna Arrays ................................ 64
  5.1. Symmetric Cross-shaped Aperture Antennas (Simulation) ................. 64
  5.2. Fabrication - Electron Beam Lithography ........................................ 66
  5.3. Fabrication - Focused Ion Beam ..................................................... 71
    5.3.1. Black and White, 10 nm per Pixel ............................................. 71
    5.3.2. Lines Segments ........................................................................ 75
  5.4. Conclusion ..................................................................................... 81
6. Cylindrical Hole Arrays ....................................................................... 82
  6.1. SPP Bloch Modes and Wood Anomalies ......................................... 82
  6.2. Impact of Cavity’s Dielectric Constant on The SPP Bloch Modes ....... 89
  6.3. Incident Light and SPP coupling .................................................... 91
  6.4. Conclusion ..................................................................................... 95
7. Polarization Response of Bi-Periodic Hole Arrays ................................. 97
  7.1. Surface Plasmon Wave Plates ....................................................... 98
    7.1.1. Fabrication, Results and Discussion ........................................ 105
  7.2. Polarization Dependent Refractive Index Based Sensor with Tuneable Quarter Wave Plate and Colour Filter Functionalities ............................. 108
7.2.1. Results and Discussion ................................................................. 111
7.3. SPP-LSP Coupling ........................................................................ 114
7.4. Conclusion ...................................................................................... 116
8. Bullseye (Single Aperture Surrounded by Corrugations) ...................... 118
  8.1. Introduction ..................................................................................... 118
  8.2. Analysis and Design ....................................................................... 121
    8.2.1. Excited with a normally incident light ..................................... 129
    8.2.2. C6 with a nano-diamond inside the aperture ............................ 132
    8.2.3. Surface Plasmon-Coupled-Enhanced Transmission in diamond substrates with a NV near the surface ......................... 136
  8.3. Fabrication and Characterization ...................................................... 141
  8.4. Results and Discussion .................................................................. 143
  8.5. Conclusion ...................................................................................... 145
9. Polarizing Bullseye Structure ............................................................... 147
  9.1. Design, Fabrication and Characterization ...................................... 148
  9.2. Conclusion ...................................................................................... 150
10. Travelling SPPs with Non-travelling Interference Envelope .................. 152
  10.1. Simulations, Results and Discussion ........................................... 153
  10.2. Conclusion ...................................................................................... 161
Finale ....................................................................................................... 163
Appendix A - Other BE Devices ............................................................ 165
Appendix B – Nano-diamond: Preparation, characterization and integration steps.... 173
References .............................................................................................. 176
Citations to Previously Published Work


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List of Figures

Figure 1: Surface polarization at metal/dielectric interface [10].......................... 3
Figure 2: Excitation of SP on metallic nano-sphere by the electric field of incident light, which displaces the free electrons collectively from their equilibrium position. [10].... 4
Figure 3: Size effect on the resonances of a nanoparticle follows a distinct trend. Size reduction results in a blue shift and dampening of the spectra [20].......................... 4
Figure 4: Schematic illustration of nanostructures with various shapes [10]............. 5
Figure 5: Shape effect in nanoparticles follow a complex relation with no apparent trend [20]. .................................................................................................................. 5
Figure 6: RF dipole antenna and its radiation pattern showing the electric and magnetic field lines and the direction of their propagation [36]................................. 8
Figure 7: Toroidal radiation pattern of a dipole antenna [37]............................... 9
Figure 8: The angular directivity for a vertical gold nano-antenna having diameter D = 30nm with various lengths, coupled to a horizontal dipole emitter (red arrows) [38]... 9
Figure 9: RF antenna theory with only a surface current (left) and plasmonic antenna theory with a volume current (right) that leads to a much shorter (plasmon) wavelength as it can be seen in the dispersion relation (centre). Transferring a fixed length antenna from the RF regime to the plasmonic regime leads to fundamentally different radiation patterns (bottom) [42].................................................................................. 10
Figure 10: Analogy between loading of a regular RF dipole and an optical nanodipole antenna. a) A regular RF dipole antenna made of highly conductive metal is loaded with lumped circuit elements. b) An optical nano-antenna loaded with combinations of nanoparticles acting as nano-circuit elements [46]........................................ 11
Figure 11: Application of bow-tie antenna in lasers [58]...................................... 12
Figure 12: Gap enhancement in (a) bow-tie antenna, (b) first-, (c) second- (d) and third-iteration Sierpiński fractal bow-tie antenna [55]................................. 12
Figure 13: A plasmonic Wheatstone bridge circuit utilizing three optical nano-antennas for detecting a single molecule [60].

Figure 14: An example of a RF directional antenna consisting of a parabolic reflector and an antenna feed position at the focal point of the parabola [63].

Figure 15: a) surface current b) electric and magnetic dipoles and c) electromagnetic field lines formed inside and in the surrounding of an elliptical slit when the electric field vector of the excitation wave is parallel to the ellipse’s major axis [64].

Figure 16: (a) surface current (b) electric and magnetic dipoles and (c)-(d) electromagnetic field lines formed inside and in the surrounding of an elliptical slit when the electric field vector of the excitation wave is parallel to the ellipse’s minor axis [64].

Figure 17: spectrogram of a diffraction grating produced using various incident angle [78].

Figure 18: Schematic demonstration of a metallic diffraction grating [83].

Figure 19: Transmission spectrum of a slit with w = 0.150 μm and t = 1 μm. Vertical solid lines: Wavelengths satisfying Fabry-Pérot resonances. Vertical dashed line: Actual cavity resonances [84].

Figure 20: Transmission spectrum of a periodic slit-array with w = 0.150 μm, t = 1 μm and pitch of the grating d = 0.9 μm has the same number of Fabry-Pérot resonances, as seen in Figure 19, are clearly visible [84].

Figure 21: schematic of a single slit in metallic thin film surrounded by surface grooves [86].

Figure 22: Periodic texturing of the output surface surrounding a single aperture collimates the output beam with angular divergence of ±3° at resonance wavelength [92].

Figure 23: application of the periodic grating structure on the output surface of a Quantum Cascade Laser to reduce the beam’s divergence angle [93, 94].

Figure 24: Intensity spectra as a function of beaming angle: a) at λ = 560 nm maximum intensity is at 0° b) 800 nm has two maxima at ±30° [95].
Figure 25: Variation of slit width along the $x$-axis brings about the required phase retardation for constructive interference at a desired focal point in the $z$ direction [126].

Figure 26: A group of 5 annular ring apertures arranged in a cross formation, a meta-atom, forms the basis of a 2D square lattice.

Figure 27: (a) Dispersion curves for SPP (the curved line), light in vacuum ($\omega = c k_0$) and the evanescent light produced by the prism ($\omega = c k_0/n$). (b) Otto configuration for generating SPPs. (c) Formation of evanescent wave at base of a prism when incident angle is larger than the critical angle. (d) Kretschmann configuration is an alternative way in generating SPP in metallic film having thicknesses in the order of skin depth [8].

Figure 28: Reflectivity vs. various incident angles in metallic films with various thicknesses (nm) [8].

Figure 29: Surface charge density (red = 1, blue = -1 and green = 0) at glass/silver interface when illuminated by a normally incident light, linearly polarized at $-45^\circ$, indicates that the strength of travelling SPP waves launched from the rims of the cavity at the centre, follows the complex phasor relation (2.24). $K_{SPP}$ is parallel to the in-plane component of the incident electric field. This direction is defined as $\varphi = 0$. The strength of the SPPs decreases as $\rho$ increases and/or as $|\varphi|$ approaches $90^\circ$.

Table 1: SPP Bloch modes in reciprocal space. Possible modes supported by a square lattice. Each wavevector is related to a different combination of $(i,j)$ in $G = iG_x + jG_y$. e.g. $G_1$ is related to the degenerate modes $G_1 = iG_x$ or $jG_y$. $G_2 = iG_x + jG_y$, $G_3 = i2G_x$ or $j2G_y$ and so on. Degenerate modes resulting from $-i$ and $-j$ are omitted.

Figure 30: (a) Unlike the finite difference method, FEM mesh elements can be a mixture of different types of polyhedrons with various sizes. (b) Single triangular mesh element within the domain.

Figure 31: Vistec EBPG5000plus

Figure 32: Steps in preparation and fabrication of nano-cavities using EBL

Figure 33: A Helios NanoLab FEI FIB instrument.
Figure 34: A Helios NanoLab FEI FIB instrument. Components inside the chamber... 48

Figure 35: (a)-(b) A typical electron-beam and ion-beam gun (or column). (c) close up of the Ion source. [147] ................................................................. 49

Figure 36: Schematics for the confocal setup ......................................................... 52

Figure 37: An inverted microscope customized for polarization measurements ......... 54

Figure 38: (Color online) a) Two nanorods with distinct orthogonal resonant modes that radiate light perpendicular to their dipole moments (shown by the dashed arrows). The angle $\psi$ defines the direction of polarization of the incident light. b) The radiated power (solid line) and the range of angles over which radiation is dispersed (dashed line) as functions of the normalized frequency difference between the nanorods' resonances. The radiated power is expressed as a ratio of $dP/d\Omega$ at the frequency of one of the resonances to $dP/d\Omega$ at a frequency midway between the two resonances. ............... 56

Figure 39: Schematic representation of the two nanorods in T configuration. Surface charge density (red = 1, blue = -1 and green = 0) clearly indicates the dipole formation along each nanorod .................................................. 60

Figure 40: Based on a numerical solution of Maxwell's equations: (a) The scattering power at $\psi = 0^\circ$ and $\psi = 90^\circ$ polarizations. At $\psi = 0^\circ$, excitation is limited to nanorod-1 (57 nm long), whereas at $\psi = 90^\circ$, excitation is limited to nanorod-2 (61 nm long). Simulated scattering spectra have peaks at $\lambda_{res1} = 697$ nm and $\lambda_{res2} = 712$ nm. Both spectra have FWHM of 25 nm. (b) The scattering power maxima and the relative angles at which they occur. The two sets of curves in (b) relate to the lower and upper scattering maxima shown in (a). ................................................................. 60

Figure 41: The far-field radiation pattern in x-y plane ................................................. 61

Figure 42: Radiation cross section spectra vs. the incident polarization for the asymmetric cross-shaped nanoparticles with arm-lengths $L_1 = 80$ nm, $L_2 = 95$ nm and arm-width $W = 24$ nm. $\lambda_{res1} = 815$ nm and $\lambda_{res2} = 885$ nm and FWHM is $\sim 95$ nm. At $\psi = 32^\circ$, (solid green line), scattered power is almost constant between $\lambda_{res1}$ and $\lambda_{res2}$... 62

Figure 43: Far-field radiation pattern in the x-y plane. Total beam steering angle of 55° is achieved for $\lambda_{res1} < \lambda < \lambda_{res2}$ ................................................................. 62
Figure 44: Angular positions of the maxima of the far-field vs. the wavelength for the asymmetric copper cross nanoparticle.

Figure 45: (a) schematic of a unit cell with periodicity $P$, used for modelling the cross-shaped aperture array having arm-lengths $L$, arm-widths $W$ perforated in a silver film having thickness $h$. (b) Parametric sweep over the arm-length with initial parameters of $P = 250$ nm, $h = 40$ nm and $W = 40$ nm, sets the optimum arm-length for LSPR at $\lambda = 700$ nm to $L = 145$ nm. Resonance shift vs. (c) $W$ and (d) $h$, for $L = 145$ nm.

Figure 46: Optical image of arrays of cross aperture fabricated with EBL. Each array is 12x12 $\mu$m$^2$.

Figure 47: (a) SEM images of the crosses with arm-lengths ranging from 128 nm to 143 nm which corresponds to the target arm-length $L = 150$ nm as specified in GDF files. (b) Inhomogeneity in shapes and dimensions of the crosses is an indication of the poor quality when fabricating aperture arrays using EBL.

Figure 48: Image representation of the SPE files. The $x$-axis represent the wavelength and the $y$-axis represent the spatial position of the pixel. Pixel intensity at (x,y) position is the measure of amplitude at a specific wavelength and position. a) from top to bottom, 170, 160, 150 nm arm-lengths, b) from top to bottom, 140, 130, 120 nm arm-lengths.

Figure 49: $L = 170$ nm.

Figure 50: $L = 140$ nm.

Figure 51: $L = 160$ nm.

Figure 52: $L = 130$ nm.

Figure 53: $L = 150$ nm.

Figure 54: $L = 120$ nm.

Figure 55: Consolidated plot: Transmitted intensities averaged over all spectral lines (i.e. over the length of an array), normalized to the maximum pixel intensity.

Figure 56: (a) bitmap representation of the cross-shaped aperture antenna array. Dimensions in pixels: arm-length $L = 15$ pixels, arm-width $W = 5$ pixels and periodicity
\( P = 25 \) pixels. (b)-(c) SEM images taken at 52° tilt angle corresponding to milled arrays with dwell times 2 ms, 2.2 ms and 2.5 ms respectively. .............................................. 72

Figure 57: bmp pattern used, note the black inner corners........................................... 73

Figure 58: (a)-(b) Current \( I = 1.5 \) pA, dwell time = 2 ms, 10% relative interaction diameter, total beam diameter of 7.7 nm, (c)-(d) relative interaction diameter reduced to 0, total beam diameter of7.5 nm. (e)-(f) dwell time set to 3 ms. .............................................. 74

Figure 59: (a)-(c) Current \( I = 1.5 \) pA, dwell time = 4 ms, 0% relative interaction diameter, total beam diameter=7.5nm. ......................................................... 75

Figure 60: bitmap patterns for crosses 150 nm in arm-length. Each arm is designed a two successive line segments that measures 15 pixels in total. A central area measuring cell/6 at the centre of each cross is left blank to prevent over milling. ...................... 75

Figure 61: Top view SEM image of the arrays. ................................................................. 76

Figure 62: (a) Test run using a 10x10 cross cavity aperture array bitmap. Current was set to 1.5 pA. Dwell time was set to 4 ms and relative interaction diameter was set %0 with serpentine. The actual arm-length=154.9 nm & 134.7 nm with periodicity 252.3 nm, (b) top view. ................................................................. 77

Figure 63: 50×50 cross-shaped aperture arrays, with target arm-lengths \( L = 150 \) nm, milled with dwell times of (a) 3.5 ms and (b) 3 ms. (c) Dispersed spectra associated with (a) and (b), (d)-(e) top view with tilt angels 0° and 52° respectively. Serpentine milling pattern introduces defects in the form of asymmetry in the milled crosses. The asymmetry changes orientation in alternating rows. .............................................. 78

Figure 64: 50×50 cross-shaped aperture arrays with target arm-length \( L = 130 \) nm milled with dwell times of (a) 3.5 ms and (b) 3 ms. (c)-(d) Dispersed spectra associated with (a) and (b) respectively................................................................. 79

Figure 65: \( L = 150 \) nm DT = 3.5 ms............................................................................. 80

Figure 66: \( L = 150 \) nm DT = 3 ms............................................................................. 80

Figure 67: \( L = 130 \) nm DT = 3.5 ms............................................................................. 80

Figure 68: \( L = 130 \) nm DT = 3 ms............................................................................. 80
Figure 69: Transmitted intensities averaged over the length of an array normalized to the maximum pixel value vs. the wavelength for the 50×50 cross-shaped aperture arrays fabricated with FIB using bitmap patterns based on line segments. Target arm-lengths are \( L = 130 \) nm and \( L = 150 \) nm. averaged over all spectral lines (i.e. over the length of an array). 80

Figure 70: Absolute transmission spectra (normalized to the incident intensity over a unit cell) through a square array of holes as a function of (a) the periodicity at \( \lambda_0 = 700 \) nm and (b) the wavelength for \( P = 394 \) nm. 83

Table 2: Summary of the results obtained from simulations vs. those obtained from equations (2.25), (2.26) and (6.1). 84

Figure 71: Electric field components of the SPP calculated over the silver/glass surface (top row) and air/silver interface (bottom row) in the vicinity of an isolated hole 200 nm in diameter perforated in a 100 nm thick silver film. The aperture was illuminated with a normally incident x-polarized light from the air/silver interface. Note that \( E_x \) for the air/silver interface (bottom-left) depicts the superposition of the incident field and the SPP field. (red = 1, blue = -1 and green = 0). 86

Figure 72: Schematics of interaction between two holes via the z-component of the SPP. 86

Figure 73: (a) Squares of the modulus, \( |\psi_{x,y}|^2 \), amplitude squares of the real, \( \text{Re}(\psi_x)^2 \), and the imaginary parts, \( \text{Im}(\psi_y)^2 \), vs. the wavelength. (b) Response functions for \((0,1)_{\text{SPP-glass}}\) and \((1,1)_{\text{SPP-glass}}\) modes. (c) Modulus of the combined \( x \) and the \( y \) components of the waves, \( |\psi_x + 0.15 \psi_y|^2 \). 89

Figure 74: Transmission (normalized to the incident intensity over a unit cell area) spectra of a square array of holes, 200 nm in diameter and periodicity of 394 nm, perforated in 100 nm silver film, for various combinations of refractive indices of the substrate, superstrate and the hole. 90

Figure 75: Electric field components of the SPPs calculated over the silver/glass surface (top row) and air/silver interface (bottom row) over the array of holes 200 nm in diameter perforated in a 100 nm thick silver film. Period of the array is \( P = 433 \) nm. The array was illuminated with a normally incident x-polarized light from the air/silver.
interface. Note that $E_x$ for the air/silver interface (bottom-left) depicts the superposition of the incident field and the SPP field. All images were produced on the same scale with (red = 1, blue = -1 and green = 0).

Figure 76: Absolute transmission (normalized to the intensity over a unit cell) vs. the angle of incidence when the device is illuminated from the glass/silver side. TE mode, with $x$-z being the plane of incidence.

Figure 77: Absolute transmission (normalized to the intensity over a unit cell) vs. the angle of incidence when the device is illuminated from the glass/silver side. TM mode, with $x$-z being the plane of incidence.

Figure 78: Transmission spectra through a hole array vs. the incident angle reported by Ebbesen et al. [79]

Figure 79: Absolute transmission (normalized to the intensity over a unit cell) vs. the angle of incidence when the device is illuminated from the air/silver side. TE mode, with $x$-z being the plane of incidence.

Figure 80: Absolute transmission (normalized to the intensity over a unit cell) vs. the angle of incidence when the device is illuminated from the air/silver side. TM mode with $x$-z being the plane of incidence.

Figure 81: The square of the amplitude and the relative phase vs. the $P$ was analytically calculated for $k_{SPP} = 2\pi/433$ (nm$^{-1}$) corresponding to $\lambda_0 = 700$ nm.

Figure 82: Schematic of a bi-periodic array of cylindrical holes producing transmitted CPL when illuminated with a linearly polarized light.

Figure 83: (a) the relative phase differences between the $x$ and $y$ components of the transmitted electric field and (b) Absolute transmission (normalized to the intensity over a unit cell) as a function of $P_y$ for $P_x = 394$ nm.

Figure 84: Spectra of the hole array having periodicities $P_x = 368$ and $P_y = 407$ nm. (a) Stokes parameters vs. the incident polarization. (b) Absolute transmission vs. the wavelength, when incident field is at polarizations $0^\circ$, $90^\circ$ and $47^\circ$. (c) Stokes parameters vs. the wavelength when incident polarization is $47^\circ$. 
Figure 85: Surface charge density (red = 1, blue = -1 and green = 0) and the transmitted electric field vector (70 nm from the glass/silver interface, represented by the red arrow), were calculated at $t = \{0, T/8, T/4, 3T/8\}$, where $T$ is the period.

Figure 86: Simulated (a) transmitted $S_3$ and (b) transmitted $S_1$ parameters vs. the wavelength for incident polarizations $10^\circ \leq \alpha \leq 80^\circ$ range.

Figure 87: (a) Top view SEM image of the milled device. (b) Close-up SEM image showing the hole apertures. (c) Cross-sectional SEM image from the film.

Figure 88: Experimental data: (a) Transmitted $S_3$ spectra for incident polarizations $5^\circ \leq \alpha \leq 85^\circ$. (b) Absolute transmission when incident field is at polarizations $90^\circ, 43^\circ$ and $0^\circ$. (c) Stokes parameters vs. the wavelength for incident polarization $43^\circ$. (d) Stokes parameters vs. the wavelength for un-polarized normally incident light. Simulated data: (e)-(f) Simulated $S_3$ and $S_1$ parameters. (Red lines) simulated results performed for the fabricated geometries. (Black lines) experimental results as in (c). Dashed lines show calculations of devices assuming the extreme range of geometric uncertainties.

Figure 89: Cross-sectional schematics of the array (a) before and (b) after etching with HF. (c) cross-sectional SEM image of the freestanding array in a homogenous dielectric environment (immersion oil). (d) Close-up of (c).

Figure 90: Simulated results for absolute transmission (obtained from the scattering parameter $S_{21}$) showing the shift in $(1,0)$ SPP resonance vs. the change in the refractive index $n$ for (a) $P_x = 366$ nm (i.e. incident polarization angle $\alpha = 0^\circ$) and for (b) $P_x = 417$ nm (i.e. incident polarization angle $\alpha = 90^\circ$).

Figure 91: (a) Absolute transmission vs. the film thickness $h$ for $\alpha = 90^\circ$ and $\lambda = 790$ nm (i.e. $\lambda(1,0)_{\text{glass}}$ for Py). (b) Calculated $|E|^2$ for $h = 80$ nm, $\alpha = 90^\circ$ and $\lambda = 790$ nm.

Figure 92: Simulated results for incident polarizations $0^\circ \leq \alpha \leq 90^\circ$. Absolute transmission vs. the wavelength for (a) $n_1 = n_2 = n_3 = 1$ and (b) $n_1 = n_2 = n_3 = 1.52$. $S_3$ vs. the wavelength for incident polarizations $0^\circ \leq \alpha \leq 90^\circ$ for (c) $n_1 = n_2 = n_3 = 1$ and (d) $n_1 = n_2 = n_3 = 1.52$.

Figure 93: Experimentally obtained results for incident polarization $0^\circ \leq \alpha \leq 90^\circ$. Absolute transmission vs. the wavelength for (a) $n_1 = n_2 = n_3 = 1$ and (b)
$n_1 = n_2 = n_3 = 1.5$. $S_3$ vs. the wavelength for (c) $n_1 = n_2 = n_3 = 1$ and (d) $n_1 = n_2 = n_3 = 1.52$. .......................................................... 114

Figure 94: (a) Normalized Stokes parameters for a rectangular array of symmetric cross-apertures, 165 nm in arm-length and 40 nm in arm-width, perforated in a 100 nm thick silver film when illuminated by a normally incident plane wave at $\lambda = 700$ nm polarized at $45^\circ$. The periodicities of the array are $P_x = 272$ nm and $P_y = 366$ nm. (b) $S_3$ parameters for the rectangular array of symmetric cross-cavity apertures with various arm-lengths. .......................................................................................... 116

Figure 95: A cylindrical cavity in a metallic film surrounded by concentric circular corrugations to control the emission pattern from a dipole positioned at its centre [73]. .................................................................................................................................. 119

Figure 96: BE grating etched on the surface surrounding a nitrogen vacancy [242]... 120

Figure 97: (a) Components of a bullseye structure. (b) Formation of a hotspot with a dipole moment $\vec{p}_i$ on the upper corner of a groove at coordinates $(x_i, z_0)$, due to the surface charge oscillations $\sigma_z(x, z_0)$ and $\sigma_x(x, z)$. (c) Electric field components launched by a resonant cross-shaped aperture at $\lambda_0 = 700$ nm.......................................................... 123

Figure 98: Amplitudes of the $x$, $y$ and $z$ components of the electric field obtained from 3D models, calculated over the exit surface surrounding (a)-(c) the cross aperture and (d)-(f) the circular aperture. (g) $z$ component of the electric field calculated in the vicinity of the cross aperture: (red) amplitude, $|E_z(x)|$ (grey and blue) $E_z(x, t)$ at two arbitrary times $t_1$ and $t_2$. (h) 3D calculations of the time averaged radial component of the Poynting vector vs. the angular direction in the $x$-$z$ plane for (red) circular aperture and (blue) cross aperture.......................................................................................... 127

Figure 99: (a) The $x$-component of the far-field intensity integrated over an arc encompassing the aperture and the corrugations vs. the period, normalized to the maximum intensity. (b) Calculated $|P_x|$ inside the film and $|E_x|$ in the surrounding dielectrics for $h = 50$ nm and (inset) close-up of the corrugations showing $E_x$ penetrating the film. (red = 1 and blue = 0)...................................................................................................... 129

Table 3: Summary of the configurations for C6 and $C_{ref}$ configurations. .......................... 129
Figure 100: Electric field components on the surfaces surrounding (a)-(c) the resonant cross aperture and (d)-(f) the circular hole obtained from 3D models. Calculations of the time averaged radial component of the Poynting vector vs. the angular direction, calculated in the x-z plane at the boundaries of a hemisphere encompassing the device for (g) $\alpha = 199$ nm.

Figure 101: $|E|^2 \times 62.5$ (V/m)$^2$ of (a) C6 and (b) C$_{\text{ref}}$, with both film thicknesses set to 300 nm and the same $d/\lambda_0$ ratios.

Figure 102: $|E|^2 / 1.3 \times 10^{17}$ (V/m)$^2$ for (a) C$_{\text{ref}}$ with 50 nm in diameter nan-diamond positioned inside the aperture, (b) C$_{\text{ref}}$ in the presence of the 50/25 system and (c) C6 C$_{\text{ref}}$ in the presence of the 50/25 system.

Figure 103: $|E|^2 / 1.3 \times 10^{17}$ (V/m)$^2$ by (a) C6$_{t=365\text{nm}}$ and (b) C$_{\text{ref}}, t=350\text{nm}$- (c) The x-component of the electric field at the silver/air interfaces of both devices.

Figure 104: (a) $|E|^2 / 1.3 \times 10^{17}$ (V/m)$^2$ scattered by C6$_{t=120\text{nm}}$ when excited by the 50/25 source system with $t = 120$ nm and $h = 70$ nm. (b) Simulated directional gain of the antenna $|E|/|E_0|$ as a function of angle, $\theta$, from the optical axis. (c) 3D simulation of the $|E|^2 / 1 \times 10^{31}$ (V/m)$^2$ scattered by C6$_{t=120\text{nm}}$ with the cross arm-lengths $L = 140$ nm and arm-widths $W = 30$ nm. Film thickness, the corrugations, nano-diamond diameter and its position inside the aperture were set according to the 2D model.

Figure 105: x-component of the electric field calculated for C6 with at an arbitrary time for (a) $t = 365$ nm and (b) $t = 120$ nm. (Red =1, blue = -1, green =0).

Figure 106: (a) Power ratio vs. the $z$ calculated as $P_{\text{air-C6}}/P_{\text{sub-C6}}$ where $P_{\text{air-C6}}$ and $P_{\text{sub-C6}}$ are the total power scattered into the air and the substrate respectively. (b)-(g) $|E|^2 / 1.3 \times 10^{17}$ (V/m)$^2$ vs. the dipole distance from the surface. (h) weakly coupled regime occur between the surface modes and the dipole’s emission occurs at $z \leq - 200$ nm, where the film becomes reflective.

Figure 107: $|E|^2 / 1.3 \times 10^{17}$ (V/m)$^2$, for a diamond substrate with an NV- positioned 10 nm below the surface with (a) no silver film, (b) 100 nm thick silver film, (c) 100 nm thick silver film with a 50 nm wide slit, (d) C6 configuration with $t = 200$ nm, (e) C6 when corrugations are milled all the way through the film and (f) C6 having refractive index of the material filling the corrugations set to 2.41.
Figure 108: $|E|^2/1.3 \times 10^{17} \text{ (V/m)}^2$ calculated for the C$x$ configuration that has no aperture with periodicity matching the SPP wavelength at (a) silver/air interface $\lambda_a = 2\pi/k_a = 667$ nm, (b) silver/diamond interface $\lambda_g = 2\pi/k_g = 230$ nm and (c) $P = (\lambda_a + \lambda_g)/2$. Device was excited with a NV- positioned at $z = -10$ nm from the silver/diamond interface.

Figure 109: a) Ion beam 7.5 nm in diameter produces a gap of 2.5 nm between the neighbouring pixels when the FIB resolution is set to 10 nm per pixel. b) With the resolution of the FIB set to half the beam diameter, i.e. 3.25 nm per pixel, ion beam overlaps between the two neighbouring pixels on the film surface.

Figure 110: SEM images for the fabricated C6 configuration. (a) Top view. (b)-(c) close-up images for the aperture. (d) close up of the top view.

Figure 111: A set of 2D images obtained by raster scanning in the $x$-$y$ plane at various distances from the device. 2D images were stacked to form a high-resolution 3D volumetric map of the transmitted intensity.

Figure 112: Simulated results for the C6 configuration using a 2D model. (a) Maxima positions and their corresponding depth of focuses identified on $|E|^2$ along the optical axis. (b) $|E|^2$ in the $x$-$z$ plane. (c)-(d) $|E|^2$ calculated along the $x$-axis at their maxima.

Figure 113: Experimental results for the C6. (a) 3D iso-surface corresponding to the $0.5 \times I_{max}$. (b) Normalized photon counts along the central lobe. (c) A 2D slice in the $y$-$z$ plane obtained from the volumetric map. (d)-(e) Normalized photon counts in the $x$-$y$ plane at $z_1 = 5 \mu$m and $z_2 = 26 \mu$m.

Figure 114: Design proposed by Gorodetski et. al. [113] that utilizes elliptical corrugations in their Bullseye structure to produce the transmitted CPL.

Figure 115: Assymetric cross shaped aperture with target arm-lengths $L_x = 150$ nm and $L_y = 220$ nm.

Figure 116: Experimental results: (a) A 2D slice in the $y$-$z$ plane obtained from the volumetric map. (b) Normalized photon counts in the $x$-$y$ plane at $z = 35 \mu$m, (c) beam intensities along the x and the y axis at $z = 35 \mu$m shows a symmetric profile.

Figure 117: Transmitted $S_3$ parameter for various incident polarizations.
Figure 118: (a) Surface charge density, $\sigma(x,t_0)$, at an arbitrary time $t_0$, calculated at the air/silver and glass/silver interfaces. (b) The envelope, $|\sigma(x)|$, at the air/silver interface. The corresponding fast Fourier transforms of (c) the wave $f[\sigma(x, t_0)]$ and (d) the envelope $f[|\sigma(x)|]$. ................................................................. 154

Figure 119: $f[\sigma(x, t_0)]$ and $f[|\sigma(x)|]$ calculated for (a)-(b) $h = 50$ nm on glass substrate, (c)-(d) $h = 25$ nm on glass substrate and (e)-(f) $h = 25$ nm on diamond substrate. Note that subscript ‘g’ is used to label the substrate in general. ............................................... 157

Figure 120: Surface charge densities, $|\sigma(x)|$, over the air/silver surface for $h = \{100, 50, 25\}$ on glass substrate, $h = 25$ nm on diamond substrate and for PEC....................... 158

Figure 121: Snapshot of electric field $E_x$ passing through a periodic charge screen (with periodicity $1/K_{\text{beat}}$) formed inside the 25 nm thick silver film for (a) glass and (b) diamond substrates. Note that $E_x$ was calculated at an arbitrary time with the maximum of its amplitude falling over the silver film, hence highlighting the periodic arrangement of the field inside the film. ................................................................................. 160

Figure 122: $|E|^2 \times 10(\text{V/m})^2$ Diffraction patterns of a transmitted Gaussian beam through (a) 25 nm silver film perforated with a slit, supported on a diamond substrate. (b) same as (a) with the maximum intensity of the Gaussian beam displaced to $x = 680$ nm away from the centre of the slit. (c) In the absence of the slit. ............................................. 161

Table 6: Summary of target dimensions for the BE configurations .............................................. 165

Figure 123: Simulated far-field intensities, $|E_{\text{far}}|^2$ and $|E_{\text{far}}|^2 / |E_{\text{far}}^{\text{ref}}|^2$ as a function of angle, $\theta$, from the optical axis for C1-C6 and C_{ref} configurations. .................................................. 166

Table 7: Summary of fabricated dimensions for BE configurations, C1-C5 .................. 166

Figure 124: SEM image for the fabricated C1. ................................................................. 167

Figure 125: (a) the simulated $|E|^2$ and (b) the measured intensity, $I$, with $\Delta z = 0.1$ $\mu$m and (c) the intensity, $I(x,y)-I_{\text{min}}/(I_{\text{max}}-I_{\text{min}})$ at $z = 0$ associated with C1 configuration. 167

Figure 126: SEM image for the fabricated C2. ................................................................. 168

Figure 127: (a) Simulated and (b) measured ($\Delta z = 1$ $\mu$m) radiation patterns. (c) Normalized intensity at $z = 0$ plane. (d) Measured ($\Delta z = 0.1$ $\mu$m), and (e) close-up of the
simulated radiation pattern for C2 configuration. Divergence angle was estimated to be 26°.

Figure 128: SEM image for the fabricated C3.

Figure 129: C3, simulations.

Figure 130: C3, experimental.

Figure 131: SEM image for the fabricated C4.

Figure 132: C4, simulations.

Figure 133: C4, experimental.

Figure 134: SEM image for the fabricated C5.

Figure 135: C5, simulations.

Figure 136: C5, experimental.

Figure 137: Step 1- Markers are milled on the surface of a silicon substrate using Focused Ion beam.

Figure 138: Step 2- Droplets of a solution containing nano-diamonds are placed over the markers and let dry.

Figure 139: Step 3- Using a confocal microscope, nano-diamonds with nitrogen vacancies that exhibit anti-bouncing (i.e. Single Photon Emission) properties are located and identified.

Figure 140: Step 4- Anti-bunching properties of nano-diamonds are measured and recorded at various pump powers prior to the integration with plasmonic devices.

Figure 141: Step 5- The micromanipulator instrument in an SEM machine may be used to integrate a nano-diamond with a plasmonic device using a technique called “pick and place”. Yellow circle shows a nano-diamond attached to the tip of the micromanipulator’s probe.
1. Literature Review

This section is a review of the most relevant topics covering qualitative descriptions of surface plasmons, their utilization in optical antenna designs in the form of metallic nanoparticles/nanorods or subwavelength metallic holes surrounded by periodic surface gratings, their role in changing the radiative decay rate of a quantum emitter and plasmonic metasurfaces, all of which were the focus of this thesis.

Classical optics is concerned with the wave nature of light and its manipulation by means of optical devices such as lenses, mirrors, wave plates, etc. Unlike radio and microwave technology, which make use of antennas that are capable of controlling the electromagnetic wave on subwavelength scales, classical optical devices suffer from the diffraction limit which prevents the confinement of light to dimensions smaller than half the operating wavelength. While Radio Frequency (RF) technology is well matured and implemented in applications such as mobile telephony, radars, radio, television, etc., their optical counterparts are being developed. Furthermore, unlike the RF antennas, classical optical devices are incapable of facilitating the interaction of the localized field with propagating electromagnetic waves. Consequently, fundamental plasmonic effects such as Surface Plasmon and Localized Surface Plasmons on metallic surfaces and subwavelength cavities/nanoparticles have become the corner stones of the search for functional optical antennas and have hence attracted considerable interest among researchers today.

One of the main obstacles preventing optical nano-antennas from technological advances was their characteristic length, which is defined to be of the order of a wavelength. The length of an optical antenna is limited to only a few hundreds of nanometres, fortunately, advances in nanotechnology have made it possible to fabricate and characterise metallic features on nano-scales with a fabrication accuracy of ±5 nm. There are major challenges, however, that need to be met before a concept is transformed into an applicable technology. Localized surface plasmons associated with shape resonances are highly sensitive to the geometry of the metallic nano-apertures. With their wide range of potential applications, research in plasmonics is gaining
momentum internationally[1]. Single photon sources that emit one photon at a time may play a significant role in the future communication systems and quantum computing [2, 3]. Controlling the polarization state of a single photon source also plays an important role in the quantum information as it provides a mechanism to define the \textit{computational basis states} [4-7]. However, due to their low rate of photon emission, detection of such light sources in isolation remains a major obstacle to the technological advances that they may bring about otherwise. Plasmonic nano-antennas may provide a solution to this dilemma. It has been shown that plasmonic effects can alter the radiative decay rate, lifetime and quantum efficiency of an emitter, which takes single photon emitters, such as quantum dots, rare-earth ions, fluorescent molecules and nitrogen vacancy diamonds, one step closer to their technological realizations. Plasmonics is a branch of physics concerning phenomena such as

- Surface Plasmon Polaritons (SPP).
- Localized Surface Plasmon Resonance (LSPR).

The first category is related to effects manifested on the bulk metal/dielectric interfaces whereas the second phenomenon occurs in metallic nanoparticles or subwavelength metallic features. Each term will be examined and elucidated in the following sections. Although the fundamental plasmonics effects are mainly those stated above, these phenomena give rise to other fascinating effects such as:

- Subwavelength confinement in metallic waveguides, which is thought to assist in the miniaturization of optical circuits.
- Enhanced transmission through nano-scaled metallic apertures that has many applications such as near-field spectroscopy, sensors, wave plates…etc.
- Local or near-field enhancement in metallic nanoparticles with applications in optical antennas, Surface Enhanced Raman Spectroscopy, solar cells…etc.
- Plasmonic meta-materials which offer the possibility of designing and fabricating structures with a specific physical properties, which otherwise do not exist in nature, (e.g. negative index meta-materials).
1.1. Surface Plasmon Polaritons

Surface Plasmons (SP) are the coherent surface charge density oscillations that are manifested as longitudinal waves travelling along the metal/dielectric interface[8], see Figure 1. The excitation of SPs is due to the electronic negative and ionic positive charges to be segregated into separate charge bundles at metal/dielectric interface. These charge groups oscillate back and forth along the metal surface collectively and produce a longitudinal surface wave. Electric field lines between the positive and negative charges shows the coupling between fields and the surface charges. Surface plasmons together with all the coupled electromagnetic fields constitute a quasi-particle called Surface Plasmon Polaritons (SPP)[9]. SPs play an important role in the optical properties of metals in their bulk form. Mechanisms behind the excitation of SPPs are explained in sections 2.4 and 2.5 in detail.

Figure 1: Surface polarization at metal/dielectric interface [10].

1.2. Localized Surface Plasmon (Resonance)

By definition, nanoparticles in their various forms, such as the colloidal, nanocrystals or clusters, see Figure 4, are characterized as particles having a diameter between 1 to 1000 nm. In most cases, especially in case of noble metals, nanoparticles exhibit chemical and physical properties that are vastly different from those of their bulk counterparts. In this thesis, the properties of interest are mainly the optical ones. In the bulk form, the optical properties of matter remain independent of size and shape. However, in the case of metallic nanoparticles, surface charges are confined to a closed surface, hence the term Localized [9] For sizes in the same order of magnitude as the skin depth of the metal, when exposing a nanoparticle to an external electric field, the electronic cloud surrounding the particle experiences a displacement from its equilibrium position, with a tendency to oscillate back to their equilibrium position, see Figure 2.
In the case of nanoparticles larger than the skin depth, electronic oscillations are confined to the surface of the nanoparticle. In both cases, the oscillatory charge relaxation has a natural frequency, which depends on the shape, size and the surrounding environment in which the nanoparticle is immersed. When the frequency of the incident field matches the natural frequency, Localized Surface Plasmon Resonances (LSPR), are formed [9, 11, 12], hence the term resonance. The term localized signifies the fact that, unlike the bulk metal/dielectric interface where the SPPs propagate along the semi-infinite 2D boundary, the SPs in nanoparticles may not propagate beyond the nanoparticle’s dimensions. At frequencies other than the resonance, the oscillation of surface charge density is referred to as Localized Surface Plasmons (LSP).

The optical properties of the nanoparticles are highly dependent on their size, shape and material. The Size effect is characterized by a blue shift in SPs’ resonance and an increase in damping with decrease in size [13-19]. Whereas the impact of geometrical shape on resonance follows a more complex pattern, see Figure 3-5.

Figure 3: Size effect on the resonances of a nanoparticle follows a distinct trend. Size reduction results in a blue shift and dampening of the spectra [20].
Figure 4: Schematic illustration of nanostructures with various shapes [10].

Figure 5: Shape effect in nanoparticles follow a complex relation with no apparent trend [20].

It is important to note that the most pronounced LSPR mode that may be excited on a nanoparticle, is that of the fundamental dipole mode regardless of its geometry. From the material point of view, aluminium, copper, silver and gold are the most popular metals for plasmonic applications. Aluminium offers the advantage of designing and fabricating nanoparticles that are larger in dimensions in comparison to silver and gold nanoparticles for a given resonant wavelength. Resistive losses in gold and silver nanoparticles are not dominant in comparison to radiative losses. Therefore, they possess narrower spectral FWHM in comparison to other metallic nano-particles. The oxide layer in copper, which is responsible for damping of LSPR, results in spectral broadening compared to gold and silver. This can also be advantageous in applications requiring broader spectra [20-22].
1.3. Optical Antennas

Classically, an antenna is defined as a device capable of absorbing a free propagating electromagnetic wave and converting it into localized energy and vice versa [23]. Television antennas that are commonly installed on the rooftops of modern houses are an example of radio frequency receiver. As mentioned previously, light incident on nano-scaled metallic features can excite a form of plasmonic oscillation called the Localized Surface Plasmon Resonance, which are responsible for trapping EM fields in their surroundings. As a receiver, an antenna must be able capture the incoming electromagnetic waves efficiently and convert them into localized currents. Localized currents in an RF antenna are analogous to electronic charge oscillations on the surface of an optical antenna. Metallic nanostructures, therefore, certainly qualify as optical receiver antennas.

When operating in the transmission mode, an antenna must be capable of converting the localized electrical activities into free propagating electromagnetic waves. Under the right conditions, the oscillation of surface charges in nanostructures, i.e. LSPs and SPPs, decay into free propagating EM waves. When the size of a nanoparticle is smaller than the wavelength, the dipole mode dominates at the resonance frequency where its radiation pattern becomes very much the same as that of a RF dipole antenna, i.e. toroidal [24]. This also qualifies the metallic nanoparticles as transmitting antennas in optical regimes. Metallic surfaces with subwavelength periodic gratings also qualify as both transmitters and receivers as they convert the incoming EM waves into SPPs and LSPs that decay into free EM waves. Unlike isolated nanoparticles, however, optical antennas designed based on periodic features can offer other incentives, such directionality and gain.

Depending on the distance between a receiving and a transmitting antenna, energy transfer between the two may occur via the near-, intermediate- or the far-field. As the name suggests, the near-field refers to the localized, non-radiative field surrounding the antenna in a region not further than one wavelength away from it. Whereas the far-field refers to the distance away from the transmitter, (usually longer than two wavelengths), where the EM field is purely a travelling wave. The intermediate-field marks the transitory region where both the near-field and the far-field co-exist. In the context of an
optical antenna, the far-field radiation is of interest when the receiving antenna or a detector is positioned in the far-field region of the transmitter. In such cases, to improve the transmission efficiency one must aim at maximizing the directional power exchange between the transmitter and the receiver and this is where the antenna’s efficiency, directivity and gain come into effect. The efficiency of an antenna is defined as a ratio of the radiated power to the received power, \( \epsilon_a = \frac{P_{\text{out}}}{P_{\text{in}}} \). The directive gain of an antenna is defined as the ratio of its directional radiated power to the average radiated power in all directions, i.e. \( D(\theta, \varphi) = \frac{P(\theta, \varphi)}{P_{av}} \), where

\[
p_{av} = \frac{1}{4\pi} \iint_{\theta=0}^{\pi} \iint_{\varphi=0}^{2\pi} P(\theta, \varphi) \sin \theta d\theta d\varphi.
\]

The directive gain of an antenna is then defined as \( G(\theta, \varphi) = \epsilon_a D(\theta, \varphi) \) [25, 26].

A more rigorous analysis of optical antennas that includes the Radiative Decay Rate (RDR) of quantum emitters in terms of Local Density of States (LDOS) [27-30] is also presented in [23]. Optical antennas are also suitable for designing miniaturized optical elements such as filters, lenses, wave plates and meta-materials that offer the possibility of designing and fabricating structures with specific optical properties in the far-field.

Nevertheless, most plasmonic devices utilize both the near- and the far-field and act as both a receiver and a transmitter to achieve their intended goals. For example, perturbing the near-field surrounding a nanoparticle and forcing it to decay into a free propagating EM waves that are detectable in the far-field region, is the basis of the Near-field Scanning Optical Microscopy (NSOM) [31]. The first optoelectronic antenna reported by Elchinger et. al. [32], converted the incoming light to electrical currents. Nowadays, waveguides with dimensions shorter than the operating wavelengths, (a much needed ingredient in miniaturization of the all optical circuit), are becoming a reality by capturing the far-field radiation and producing localized near-fields. The light-matter interaction of a quantum emitter with its surrounding via near-field, not only leads to an enhanced radiative decay rate but may also shape its radiation pattern in the far-field. Localized near-field surrounding metallic nanostructures may be harvested by molecules located in their vicinity. The near-field effect, has found applications in Surface Enhanced Raman Spectroscopy (SERS), solar cells, microscopy,
subwavelength optical interconnects, shaping the emission of quantum emitters and many more. Metallic nanostructures absorb the incident EM waves efficiently, producing localized near-fields that can be up to 10,000 times higher than the intensity of the incident light \[20\]. These high intensity fields can contribute to the Raman scattering of the molecules positioned in their vicinity, resulting in a giant increase in Raman signals \[33-35\].

1.3.1. Metallic Nanorods and Nanoparticles

Classical dipole antenna invented by Heinrich Hertz around 1886 \[36\], which is probably the oldest example of the radio frequency transmitter, consists of two collinear conducting rods separated by a small gap where dipole oscillation along the rods results from an alternating current established on the conductor rods, see Figure 6.

![RF dipole antenna](image)

**Figure 6:** RF dipole antenna and its radiation pattern showing the electric and magnetic field lines and the direction of their propagation \[36\].

The radiation pattern of a RF dipole antenna is that of a toroid having its axis parallel to the dipole moment of the antenna, see Figure 7.
In the optical regime, metallic nanoparticles and nanorods qualify as dipole antennas. Although metallic nanoparticles were originally used in sensing and spectroscopy due to their near-field enhancement effect, soon their near-field absorption and far-field scattering generated immense interest in their use as optical antennas. An example is the angular emission of an emitter, e.g. molecules, atoms or quantum dots, which could drastically be altered when positioned in close proximity of a nanorod, see Figure 8. The resulting angle is a function of, among other factors, the nanoparticle’s size, relative positions and orientations of both emitter and nanoparticle [38-40].

Downscaling existing antenna technologies to the optical regime is of immense interest as it may lead to higher bandwidth, data rate and miniaturization. Simply downscaling existing radiofrequency antenna designs to optical regime, however, has its own challenges. For instance, it was shown [41] that in contrast to the classical antenna theory, the dipole length of optical antennas is much shorter than half the resonant wavelength. A downscaled nanowire antenna exhibits extra modes of radiation in comparison to that of a corresponding RF antenna. This is due to the formation of Fabry-Pérot resonances leading to higher harmonic oscillations along the nanowire which are otherwise absent in RF dipole antennas [42].
Figure 9: RF antenna theory with only a surface current (left) and plasmonic antenna theory with a volume current (right) that leads to a much shorter (plasmon) wavelength as it can be seen in the dispersion relation (centre). Transferring a fixed length antenna from the RF regime to the plasmonic regime leads to fundamentally different radiation patterns (bottom) [42].

An analytical model for plasmonic nanowire antenna [42], having a diameter $2R$, was developed from the existing RF antenna theory. Due to the diameter of the nano-antenna being in the order of skin depth, the surface current that could no longer be a useful parameter, was replaced with a homogeneous volume current. The model was verified both numerically and experimentally. An attempt to downscale the existing antenna designs into the optical regime using a linear wavelength scaling rule under the assumption $R \ll \lambda$ by Novotny, led to the formulation of the effective wavelength associated with metallic nanorods as $\lambda_{eff} = \left[\frac{k_0}{\gamma}\right]^{-4} R$ [43]. Here, $k_0 = \frac{2\pi}{\lambda}$, is the free-space wavenumber and $\gamma$ is the propagation constant of the SPP along the nanorod. In RF antennas, tuning to a desired resonant frequency is achieved through loading the antenna with lumped circuit elements. An interesting modelling technique where nanowires and nanoparticles were analysed in terms of lumped circuit elements such as nano-inductors, nano-capacitors and nano-resistors was proposed [44-47]. The idea is to identify the functionalities of nano-sized elements in terms of their equivalent lumped circuit that could then be used as a building block in optical antennas.
Figure 10: Analogy between loading of a regular RF dipole and an optical nanodipole antenna. a) A regular RF dipole antenna made of highly conductive metal is loaded with lumped circuit elements. b) An optical nano-antenna loaded with combinations of nanoparticles acting as nano-circuit elements [46].

Previously [48, 49], it was shown that near-field enhancement localized at a nanoscale conic shaped gold tip may reach ~3000 times the incident field intensity when the tip is illuminated from the side by light polarized parallel to the cone’s axis. Stockman [50, 51] modelled the optical energy in tapered plasmonic waveguides, using the Wentzel-Kramers-Brillouin quasiclassical approximation, and by setting the effective refractive index, $n$, along the tip to be a function of local tip radius, $R(z)$, which itself is a function of distance, $z$, from the tip along cone’s axis. He showed the existence of a singularity at the tip with $n \to \infty$ as $z \to 0$. Due to the singularity, travelling SPPs experience rapid adiabatic slow down and asymptotic stop without ever reaching the tip. Instead, they are adiabatically transformed into LSPs near the singularity producing a giant concentration of EM energy in nanoscale dimensions, hence nanofocusing. In such structure, the highest enhancement is limited by only the size of the tip. In addition, it was also shown that the oscillation of the localized optical field in space surrounding the tip is progressively decreasing in wavelength while its amplitude increases as $z \to 0$. Nanofocusing is particularly appealing to applications requiring precise spatial control. For example, in an array of closely packed nanoparticles, tapered plasmonic waveguides may be used to excite dipole oscillations on a particular nanoparticle, while excluding others. Enhancement to the near-field surrounding abrupt geometries is not exclusive to nanoparticles. Due to its simplicity in planar design the bow-tie gap antenna, formed by two triangular shaped nanoparticles separated by a subwavelength gap, was one of the earliest optical antennas designed to take advantage of this phenomenon [52-57].
11 depicts an integrated bow-tie antenna with a laser cavity in which the near-field experiences a large enhancement and spatial confinement, producing single sharp optical spot [58]. Figure 12 shows a near-field gap enhancement in a bow-tie antenna and its fractal variations where the increased gap enhancements are proportional to the number of abrupt geometry.

Figure 11: Application of bow-tie antenna in lasers [58]

Figure 12: Gap enhancement in (a) bow-tie antenna, (b) first-, (c) second- (d) and third-iteration Sierpinski fractal bow-tie antenna [55].

Such enhancements are the results of surface charge accumulations/relaxations in the vicinity of abrupt metallic geometries (including cavities and apertures perforated in metallic films), and play an important role in the design of plasmonic nanostructures. Other aspects of LSPR can be exploited when engineering a particular application. LSPRs are functions of both the constituent metal and the surrounding dielectric. In plasmonic biosensors, the change in the resonance wavelength is interpreted as a change in the surrounding permittivity induced by molecules present in the nearby environment [59]. A novel approach for highly sensitive plasmonic sensor capable of detecting a single large molecule was proposed based on a plasmonic Wheatstone bridge circuit [60], see Figure 13.
The presence of a single molecule attached to any of the nanorods, induces an imbalance in permittivity surrounding the sense and reference arms, which then is manifested as a phase differences between the two LSPRs. Under such conditions, charge distributions at a given time along each arm are different. Due to the capacitance formed between the Output-Reference and Output-Sense, dipole oscillations are modulated on the Output nanorod whose far-field radiation can be detected. Plasmonic nanorods offer great versatility in their applications. For example, the electromagnetically induced transparency (EIT) was experimentally observed in 1991 [61] and an early theoretical model describing the effect was put forward. Perhaps it is in order to draw attention to the paper on EIT [62], which makes use of metamaterials whose constituent elements are the very configuration mentioned above.

1.3.2. Subwavelength Metallic Apertures

Depending on applications, sometimes it is desirable to utilize directional antennas that exhibit beaming qualities hence highly sensitive to the direction in which they collect or emit radiations. An example of a RF directional antenna is shown in Figure 14, which consists of a reflector (a parabolic dish in this case) and an antenna feed position at the focal point of the reflector. As a transmitter, radiations emanating from the feed are
reflected by the parabolic dish and are converted into a narrow beam EM waves that are detectable along the parabola’s axis. As a receiver, incoming plane waves from the far that travel parallel to the axis of the parabola are focused into the feed, hence a directional receiver. A subwavelength metallic hole surrounded by periodic surface gratings is analogous to such directional antennas in the optical regime, where the hole plays the role of the feed and the surrounding corrugations that of a reflector.

Figure 14: An example of a RF directional antenna consisting of a parabolic reflector and an antenna feed position at the focal point of the parabola [63]

Electromagnetic activities inside a subwavelength plasmonic hole are complex. A comprehensive qualitative explanation on electromagnetic activities in the vicinity and inside of an elliptical aperture in a metallic thin film illuminated by a linearly polarized incident light was provided in [64]. Electromagnetic field lines, dipole oscillations and their loci for two distinct incident polarizations were identified, see Figure 15 and Figure 16.
An interesting and equally important effect discussed here, was the formation of electric dipole moment brought by surface currents accumulating charges at surface discontinuities, see Figure 15(a) and Figure 16(a). Having simulated a number of aperture antennas with a dipole located at their centre, the observed enhancement in radiated power at the resonance may be explained by synchronous coupling of the cavity’s dipole to that located at its centre. Such coupling changes (and in some cases enhances) the Radiative Decay Rate (RDR). A brief history on RDR is provided in the following section.
1.3.3. Radiative Decay Rate

Prior to Purcell’s discovery of the enhanced spontaneous decay rate of a magnetic dipole in an electronic environment \[27\], the spontaneous decay rate was believed to be an intrinsic property of quantum emitters, such as atoms and molecules. It became clear that the spontaneous decay rate also depends on the environment within which a quantum emitter is embedded. The enhancement and the inhibition of spontaneous decay rate associated with an emitter placed in a cavity, \[65-67\], are already demonstrated. Strong localization of photons in dielectrics was first studied by John \[68\] and later by Joannopoulos et. al. showing that the rate of photon emission is proportional to the free-photon density of state per unit volume \[69\]. For enhancements to the decay rate in various nanophotonic structures one can refer to a review paper by Pelton \[70\]. The enhancement (or the inhibition) of the decay rate is due to the matched (or mismatched) energy levels of the emitter to those of the cavity modes. In general, coupling between the two are divided into two regimes, weak and strong. In the weak regime, the change in decay rate is due to the existence/non-existence of a cavity mode matching that of the emitter. However, in strong coupling regime, there is an exchange of energy between the cavity and the emitter \[71\]. Akselrod et. al. showed an enhancement to the spontaneous emission rate exceeding 1,000 when emitters were positioned in the dielectric gap between a flat gold film silver nanocube \[72\]. The radiation pattern of the system resembles a plume formed on top of the nanocube. Directivity of the emission can be improved by incorporating the emitter inside a cavity that is surrounded by periodic surface features \[73\]. The use of non-resonant cavity, however, is not wise. The enhancement at resonance may be explained by synchronous coupling of the cavity’s dipole to that located at its centre that gives rise to the LDOS at resonance mode, thus increasing the spontaneous emission. Therefore, the existence of cavity LSPRs play an important role in the enhancement of the RDR. The shape and the dimensions of a cavity/aperture are some of the factors that may be tailored to match their LSPR to the emission wavelength of the dipole located at their centre. As mentioned above, periodic surface gratings surrounding a subwavelength hole may act as a reflector that (if designed properly) reshapes the dispersed radiation emanating from the hole into a beam of light. Surface gratings in a periodic setting, implies the presence of evanescent Bloch waves which is covered in the following section.
1.3.4. Surface Corrugations

Discovery of SP Bloch waves may be attributed to R. W. Wood. In 1902, [74] R. W. Wood, stumbled upon a phenomenon, the so called Wood- Rayleigh anomaly, that could not be explained at the time. While examining the optical properties of diffraction grating using an incandescent light source with various incident angles and producing the first order reflection spectra, he noticed the shift in positions of the maxima and minima along the spectrogram by changing the angle of incidence. The shift in position exhibited a very complex pattern, e.g. two different spectral lines merging each other by decreasing the angle of incident, or change in spectral line colours when changing the polarization of the incident light from p- to s-. An attempt to explain the physics behind this phenomenon was initially made by Lord Rayleigh and Wood without arriving at a full theoretical model [75-77].

Figure 17: spectrogram of a diffraction grating produced using various incident angle [78].

The anomaly is concerned with metallic films with narrow lines ruled on the surface illuminated by polarized light such that the electric field vector was perpendicular to the rulings. Spectrograms of the first order reflected light, produced for various incidence angles, \(0^\circ \leq \theta \leq 46^\circ\), exhibited sharp minima and maxima in intensities whose positions shifted with respect to incident angle, see Figure 17, [78]. The dark bands were explained to be the result of destructive interference, however, when the grating space is equal to an integral multiple of wavelengths, dark bands corresponded to constructive interference at grazing angle, i.e. light propagating parallel to the surface. In 1998, Ebbesen et. al. [79, 80] demonstrated enhancement in transmitted light through an array of subwavelength apertures perforated in silver film. Transmission spectra obtained for
various film thicknesses, cavity dimensions and array periodicity indicated that the resonance and the magnitude of transmission is a function of all aforementioned parameters. Two very important observations were made: 1) The linear dependence between array periodicity and resonance wavelength and, 2) The dependence of the resonance on the thicknesses/diameter aspect ratio, where aspect ratio of $\sim 1$ produces the narrowest width. Ghaemi et al. [81], suggested that maximum transmission is the result of coupling of the incident light with surface plasmons on both metal surfaces obeying the conservation of momentum $k_{spp} = k_x + k_R$, where $k_{spp}$ is the surface plasmon wave vector, $k_x$ is the component of the incident wave vector parallel to the metal surface and $k_R$ is the reciprocal lattice vector, $k_R = \frac{2\pi}{d}$, where ‘d’ is the surface lattice periodicity formed by surface corrugations [81]. An important distinction is to be made here regarding the excitation of SPPs on flat metallic surfaces, i.e. $k_R = 0$, which requires a prism to generate evanescent surface waves such as those in Kretschmann or Otto configurations [8, 82]. Further investigations on the subject using a metallic grating [83], see Figure 18, revealed the existence of two distinct mechanisms responsible for this enhancement: 1) excitation of SPP modes on one side of the metallic grating which couples and excite SPP modes on the other side when $\lambda \approx d$ with the film being sufficiently thin and 2) coupling of incident EM waves to the slits’ cavity modes when $\lambda \gg d$.

Figure 18: Schematic demonstration of a metallic diffraction grating [83].

The enhancement in extraordinary transmission via coupling of incident light to cavity modes using a single metallic slit was theoretically analysed [84]. It was shown that the transmission spectrum of a single metallic slit possesses Fabry-Pérot characteristics. However, when experimental data were compared to the theory, a noticeable red-shift in the experimentally obtained resonances were observed, see Figure 19.
Figure 19: Transmission spectrum of a slit with \( w = 0.150 \, \mu m \) and \( t = 1 \, \mu m \). Vertical solid lines: Wavelengths satisfying Fabry-Pérot resonances. Vertical dashed line: Actual cavity resonances \[84\].

The inconsistency between the two sets of data was attributed to the higher diffracted orders caused by the evanescent surface waves whose contributions were not taken into account previously, hence the significance of surface modes. Nevertheless, the magnitude of the transmission through a single metallic slit remains low.

Figure 20: Transmission spectrum of a periodic slit-array with \( w = 0.150 \, \mu m \), \( t = 1 \, \mu m \) and pitch of the grating \( d = 0.9 \, \mu m \) has the same number of Fabry-Pérot resonances, as seen in Figure 19, are clearly visible \[84\].

One of the earliest theoretical models concerning a circular holes in perfect electrical conductor (PEC) films attributes the low transmission to the attenuation of the evanescent modes inside the aperture \[85\]. On the other hand, an infinite slit-array in a diffraction grating formation, produces near perfect transmission at peak resonances, see Figure 20. The enhancement in transmission through slit-arrays was attributed to the constructive interference of the localized Fabry-Pérot resonances of each cavity.
Corrugations surrounding a single metallic slit [86-88], see Figure 21, were shown to have an enhancing effect on the transmission of light through the slit.

![Figure 21: schematic of a single slit in metallic thin film surrounded by surface grooves [86].](image)

Considering a normally incident plane wave, these corrugations may be etched on the top/input surface, on the bottom/output surface or on both surfaces of the metallic film. Note that the top or the input interface is defined as the interface on which the plane wave is incident. With no grooves on either side, i.e. (0,0), the spectrum showed two distinct peaks at $\lambda = 400$ nm and $\lambda = 850$ nm corresponding to the aforementioned Fabry-Pérot resonances of the periodic structure. As the number of grooves are increased on the top/input surface, (x,0), a third peak appears at $\lambda = 550$ nm. For a sufficiently large number of grooves, (10,0), the highest transmittance is achieved at $\lambda = 560$ nm. It was widely accepted that the transmission enhancement observed in cavity apertures with periodic corrugations on the input surface, is purely due to excitation of SPP modes inside the corrugations [89]. However, it was argued that experimentally observed similar transmission enhancement in a non-metallic hole-array structures which does not support SPP is inconsistent with the above model. A new model, called Composite Diffracted Evanescent Wave (CDEW) was instead proposed [90, 91]. In this model, the SPP modes are just one contributing factor in the enhancement. The other contributing factors are the individual in-plane evanescent waves produced by the surface corrugations (or the neighbouring cavities in case of the hole-array). Superposition of these evanescent waves results in a composite wave
which then interferes with the SPP waves from a nearby cavity/corrugation. Depending on the constructive or the destructive interference of the two waves, transmission enhancement at some wavelengths and suppression at others is achieved. Although, the variation in the number of grooves on the output/bottom surface, \((0, x)\), has no significant impact on the transmittance, they act as collimators. Periodic texturing of the output surface surrounding a single aperture, collimates the output beam with angular divergence of \(\pm 3^\circ\) at resonance wavelength \([92]\).

Yu et. al\([93, 94]\) incorporated a carefully tailored periodic grating structure on the output surface of a Quantum Cascade Laser, surrounding the active region. Reduction in beam’s divergence angle by a factor of 30 in vertical and 10 in lateral directions were reported, see Figure 23.

Figure 22: Periodic texturing of the output surface surrounding a single aperture collimates the output beam with angular divergence of \(\pm 3^\circ\) at resonance wavelength \([92]\).
Figure 23: application of the periodic grating structure on the output surface of a Quantum Cascade Laser to reduce the beam’s divergence angle [93, 94].

Figure 24: Intensity spectra as a function of beaming angle: a) at $\lambda = 560$ nm maximum intensity is at $0^\circ$ b) $800$ nm has two maxima at $\pm 30^\circ$ [95].
Another important property one must carefully consider when designing and characterizing such structures, is wavelength dependent beaming effect due to the phase relation between the slit’s and the grooves’ far-field radiations. It was shown that the far-field intensity of a particular aperture antenna is maximum at 0° for an incident light with $\lambda = 560$ nm, whereas the spectra for $\lambda = 800$ nm has its maxima at ±30° with respect to the optical axis [95]. This phenomena has already been exploited in identifying various molecular species in a solution [96].

### 1.4. Plasmonic Metamaterial

The term “Metamaterial”[97-99] refers to an artificially produced material with properties that do not exist in nature occurring materials. Constituent microscopic elements in metamaterials are usually arranged in a periodic fashion. Various materials, such as metals and dielectrics, may be used to form the microscopic elements. However, a metamaterial as a whole can be treated macroscopically where its overall physical properties differ from the constituent materials significantly [100, 101]. Of interest to this project, are the optical properties of the planar plasmonic metamaterials. These are nanoscale metallic structures in periodic setting. In fact any periodic subwavelength metallic structure may be considered to be a plasmonic metamaterial. Arrays of nanoparticles, metallic screens with periodic subwavelength apertures (or grooves) and a single subwavelength aperture perforated in metallic film surrounded by periodic corrugations are few examples of the planar plasmonic metamaterials. These three categories, coined as *metasurfaces* in the literature, can have diverse optical properties. Due to their resonant SPPs and/or LSPs, probably the most obvious application of plasmonic metamaterials is that of an optical filter [83, 102]. Arrays of cross-shaped apertures [103, 104], simple cylindrical hole arrays, [105], annular aperture arrays, [106-108], and 1D ultrathin slit arrays, [109], are few examples. It is also possible to design nano-scale optical elements based on the plasmonic metamaterials. An optically thin quarter-waveplate based on a phase antenna array was shown to produce a high degree of Circularly Polarized Light (CPL) in the range $5 < \lambda < 12 \, \mu m$ range in transmission mode [110], where the array consisted of simple v-antennas with various angles. The broad operating wavelength is attributed to the optical properties of the v-
antennas. However, a drawback associated to this design is the wavelength dependent scattering angle of the transmitted CPL and its low efficiency which limits the transmittance to only 10% of the incident power. CPL may also be produced on transmission through an array of subwavelength apertures. A novel design based on two orthogonal LSP modes supported by asymmetric cross-shaped apertures in array formation was shown to produce a high degree of zero order transmitted CPL [111, 112]. Depending on the application, it is sometime desirable to obtain CPL through a single subwavelength aperture. Gorodetski et al. [113], demonstrated that a single metallic aperture surrounded by concentric elliptical grooves at its exit interface, produced CPL on transmission. The two orthogonal modes needed to support CPL were produced by the SPPs along the minor and the major axes of the ellipses.

Plasmonic lenses, based on slit arrays [114-116], single slit with corrugations [117, 118], bullseye structures [119-123] or a simple disk [124] are also of immense interest to scientists and engineers due to their ability to overcome the diffraction limit. Fundamental basis behind the plasmonic lens design is the distinct phase retardation experienced by the surface plasmons as they pass through each nanoslit in the array, decaying into free propagating electromagnetic waves and consequently making a constructive interference collectively at a focal point, $f$, away from the metasurface. Both the depth and the width of the slit influence the phase in which the light emanates. Based on the assumption that resonant coupling between the neighbouring slits are negligible, an analytical model was presented for a plasmonic lens, where the phase retardation of an individual slit was controlled by its depth [125]. Controlling the depth of each slit during the fabrication of such device, however, is a challenge. Shi et al. [126] showed both analytically and numerically, the possibility of focusing the transmitted light through an array of subwavelength slits, at a desired distance away from the exit surface. For practical purposes, the film thickness was kept constant. In this case, the phase modulation was achieved by varying the width of the slit, see Figure 25. Being a one dimensional slit array, however, it is conspicuous that the device performs best with TM polarized incident light and yet its focusing capabilities is only limited to one dimension thus failing to produce a focal “point”. In a similar fashion, a plasmonic microzone comprising of concentric circular metallic rings was proposed which did not suffer from such shortcomings [127]. Similarly, a series of concentric
metallic rings can also act as a polarization converter, when a radially polarized light illuminates them. Transmitted field through the device showed enhancement and a focal spot no larger than $\lambda/2.88$ in diameter. The transmitted electric field was polarized in the $z$-direction, i.e. perpendicular to the metasurface [128].

![Figure 25: Variation of slit width along the $x$-axis brings about the required phase retardation for constructive interference at a desired focal point in the $z$ direction [126].](image)

Roy et al. [129] proposed a design where a group of 5 annular ring apertures arranged in a cross formation formed the basis of a 2D square lattice in a gold film, see Figure 26. Transmission response of the device, when illuminated with circularly polarized incident light at $\lambda = 800$ nm, produced an array of foci, 14.7$\lambda$ away from the exit surface, where the FWHM diameter of each focal point measured no more than $\lambda/5$. Asymmetric crosses, instead of annular aperture, and/or variation in periodicities in orthogonal directions may create a desirable polarization response in conjunction to its meta-lens functionalities.

![Figure 26: a group of 5 annular ring apertures arranged in a cross formation, a meta-atom, forms the basis of a 2D square lattice.](image)
Performance of a classical optical lens can be modelled using the ray optics where each point in the focal plane can be traced back to a point on the image plane. Surely, some of the examples mentioned above, are capable of focusing the transmitted intensity to a subwavelength spot. Is this enough to categorise them as lenses? Unlike the optical lens, a plasmonic lens does not transmit the incident light, but rather converts its energy to form SP oscillations, which in turn decay, into free propagating EM waves. The focusing capability comes as a result of constructive interference of these secondary waves. In the case of a classical lens, it is possible to reconstruct the original image by means of other lenses. Certainly, the resonant excitation/suppression associated with the plasmonic metasurfaces will filter out some vital information.

1.5. Objectives

In chapter 4, we examine the possibility of rotating the radiation pattern scattered by a pair of nano-rods (or a single asymmetric nano-cross) by controlling the wavelength of the incident wave. We show that a pair of nano-rod can potentially function as a wavelength division multiplexer/demultiplexer and if so, under what conditions. An analytical solution is proposed followed by numerical models that confirm the theory. In chapter 5 comparisons are made between two different techniques in fabricating arrays of subwavelength cross-shaped apertures in metallic screens. This is to identify the most appropriate fabrication technique (considering the cost, ease of fabrication and accuracy in which the apertures are created) to be used for the rest of the project. In chapter 6 theoretical studies of SPP Bloch waves in arrays of cylindrical holes is carried out. A simple analytical model is presented (not for design purposes) only to identify the origin of the alleged (1,1) and (0,2) modes and the results are compared to the numerical ones. Changes in the transmission spectrum vs the refractive index of the surrounding dielectric when illuminated with a linearly polarized light at normal incidence is studied. Circumstances in which an obliquely incident light changes the SPP momentum are also identified. In chapter 7 possibilities of a simple bi-periodic array of cylindrical holes in producing circularly polarized lights are examined and requirements for the periodicities to do so, are determined. A free-standing array of cylindrical hole to achieve a tuneable waveplate, tuneable colour filter and sensor without refabricating arrays with different dimensions is proposed and confirmed both theoretically and
experimentally. In chapter 8, certain claims regarding the design criteria and characterizations of plasmonic bullseye (BE) structures are investigated. Matching of the grating’s periodicity to the wavelength of surface waves (as proposed by other authors) are examined against claims that such a condition results in a strong collimated beam along the optical axis of the device. Conditions in which the device can enhance the radiative decay rate are studied with newly proposed design criteria. The new bullseye structure, optimized based on maximization of the far-field intensity along the optical axis, is compared to the most popular design published in the literature. Techniques proposed by other authors are debated and a technique based on 3D confocal microscopy is proposed and applied experimentally to the newly proposed device. In chapter 9 it is shown that a bullseye structure with concentric circular surface gratings can change the state of polarization of the transmitted light when integrated with an asymmetric cross-aperture at its centre. Finally, in chapter 10 the impact of film thickness on travelling SPPs is examined. However, before going into the original work of this project, a review of the physics behind most relevant concepts is in order.
2. Concepts

The following introduction on electrodynamics is a consolidated review [8, 82, 130-134] of the most relevant topics to this report. In this chapter, the following topics are covered:

- The time-harmonic form of Maxwell’s equations.
- Poynting theorem related to the calculations of the total power by integrating the time-averaged Poynting vector over a given surface,
- Changes to the radiative decay rate of a dipole, when embedded in an inhomogeneous environment.
- Surface plasmons, that are the fundamental physical effects governing the functionality of optical antennas.
- Finite element method, which is the main numerical approach used throughout this report.

2.1. Time-Harmonic Maxwell’s Equations

Maxwell’s equations are the complementary set of equations, unifying and completing the work previously established by Faraday, Ampere, Gauss, Poisson and others. In classical electrodynamics, the concept of ‘field’ was introduced to explain the forces observed at the distance from charged bodies that could not be explained otherwise. It is appropriate to state the very basic definitions concerning electromagnetism that constitute the building blocks of Maxwell’s equations beforehand. Electric polarization \( \mathbf{P} \) is defined as the electric dipole moment per unit volume, \( \mathbf{P} = N\mathbf{p} \), where \( \mathbf{p} = q\mathbf{d} \) is the electric dipole moment established between two point charges, \( +q \) and \( -q \), separated by a distance \( \mathbf{d} \). Here, \( N \) is the total number of dipoles per unit volume. The electric displacement inside a medium in the presence of an electric field is defined as:

\[
\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}
\]  

(2.1)
In the case of linear, isotropic and homogeneous media, the polarization and the electric field are related via \( \mathbf{P} = \varepsilon_0 \chi_e \mathbf{E} \), which implies \( \mathbf{D} = \varepsilon_0 (1 + \chi_e) \mathbf{E} = \varepsilon_0 \varepsilon \mathbf{E} = \varepsilon \mathbf{E} \). Here, \( \varepsilon_0 \) is the permittivity of free space, \( \chi_e \) is the electric susceptivity and \( \varepsilon \) is the relative permittivity (or the dielectric constant) and \( \varepsilon \) is the absolute permittivity of the media.

Magnetic flux density or the magnetic induction, \( \mathbf{B} \), inside a magnetic medium in the presence of an applied external magnetic field, \( \mathbf{H} \), is given by:

\[
\mathbf{B} = \mu_0 [\mathbf{H} + \mathbf{M}]
\]

(2.2)

where \( \mu_0 \) is the permeability of free space and \( \mathbf{M} \equiv \mathbf{N} \mathbf{m} \), is the magnetization (or magnetic dipole moment per unit volume) and \( \mathbf{m} \) is the magnetic dipole moment. In the case of linear, isotropic and homogeneous media, magnetization is related to the applied field via magnetic susceptibility, \( \chi_m \) by \( \mathbf{M} = \chi_m \mathbf{H} \). The magnetic flux density may be written in terms of the applied field as \( \mathbf{B} = \mu_0 (1 + \chi_m) \mathbf{H} = \mu_0 \mu_r \mathbf{H} = \mu \mathbf{H} \), with \( \mu_r = 1 + \chi_m \) and \( \mu \) being the relative and absolute permeabilities of the medium respectively.

In most conductors and semiconductors, the conduction current density \( \mathbf{J}_c \), is proportional to the applied electric field \( \mathbf{E} \) via \( \mathbf{J}_c = \sigma \mathbf{E} \). The proportionality constant \( \sigma \) is referred to as the conductivity of the medium. Above relations, which form the so called constitutive relations collectively, describe the behaviour of materials under the influence of electric and magnetic fields. In summary the constitutive relations are:

\[
\begin{align*}
\mathbf{D} &= \varepsilon_0 \varepsilon \mathbf{E} \\
\mathbf{P} &= \varepsilon_0 \chi_e \mathbf{E} \\
\mathbf{B} &= \mu_0 \mu_r \mathbf{H} \\
\mathbf{M} &= \chi_m \mathbf{H} \\
\mathbf{J}_c &= \sigma \mathbf{E}
\end{align*}
\]

(2.3)
Most relevant form of Maxwell’s equation here is that of time-harmonics. In time-harmonic oscillations, i.e. monochromatic waves, it is possible to separate the time dependency and represent the wave equation in the phasor notation, \( \mathbf{E}(r, t) = \text{Re}\{\mathbf{E}(r)e^{-i\omega t}\} \). This allows the spatial term, \( \mathbf{E}(r) \), to be solved for independently of time. In the time-harmonic representation, therefore, it is convenient to write the Maxwell’s equations in terms of the complex phasors:

\[
\nabla \times \mathbf{E}(r) = i\omega \mathbf{B}(r) \tag{2.4}
\]

\[
\nabla \times \mathbf{H}(r) = -i\omega \mathbf{D}(r) + \mathbf{J}(r) \tag{2.5}
\]

\[
\nabla \cdot \mathbf{D}(r) = \rho(r) \tag{2.6}
\]

\[
\nabla \cdot \mathbf{B}(r) = 0 \tag{2.7}
\]

where \( \mathbf{J} \) and \( \rho \) are the current and charge densities. Time-harmonic wave equations may then be derived from the corresponding Maxwell’s equations:

\[
\nabla \times \mu^{-1} \nabla \times \mathbf{E}(r) - \frac{\omega^2}{c^2} \varepsilon \mathbf{E}(r) = i\omega \mu \mathbf{J}_s(r) \tag{2.8}
\]

\[
\nabla \times \varepsilon^{-1} \nabla \times \mathbf{H}(r) - \frac{\omega^2}{c^2} \mu \mathbf{H}(r) = \nabla \times \varepsilon^{-1} \mathbf{J}_s(r) \tag{2.9}
\]

where \( \mathbf{J}_s \) is the source current density. Once solutions to the complex field phasors are determined, the time-dependent field amplitudes can be reconstructed by post-multiplying them by \( e^{-i\omega t} \). Frequency dependent representations of the wave equations are most useful in solving electromagnetic problems where the steady state response to a particular frequency is under investigation. They may also be numerically solved using Finite Element Method (FEM) such as that implemented in COMSOL Multiphysics, which uses equation (2.8) with \( \mathbf{J}_s = 0 \), in the “Frequency Domain” interface to solve the full Maxwell’s equation when “Electric displacement field model” is set to “relative permittivity”. In this case, care must be taken in order not to introduce any contribution from the electrical conductivity by setting, \( \sigma = 0 \). The other option is
to set the “Electric displacement field model” to “Refractive index” which solves the model assuming non-conductive and non-magnetic materials where COMSOL sets $\mu = 1$ and $\sigma = 0$ automatically.

Boundary conditions derived from Maxwell’s equations across a surface between media 1 and 2 are:

1. The tangential components of the electric field must be continuous across any interface
   \[ \hat{n} \times (\mathbf{E}_1 - \mathbf{E}_2) = 0 \] (2.10)

2. The tangential components of the magnetic field must be continuous across any boundary except where surface current is present. Surface currents are present at the surface of a conductor due to free/conduction electrons. Magnetic field inside a perfect conductor is 0.
   \[ \hat{n} \times (\mathbf{H}_1 - \mathbf{H}_2) = 0 \]
   \[ \hat{n} \times \mathbf{H}_1 = J_{\text{surf}} ; \quad \mathbf{H}_2 = 0 \] (2.11)
   where $\mathbf{J}_{\text{surf}}$ is the surface current density.

3. The normal components of the electric flux density, i.e. electric displacement, are continuous across interfaces except where the surface charges are present.
   \[ \hat{n} \cdot (\mathbf{D}_1 - \mathbf{D}_2) = \rho_{\text{free}} \] (2.12)

   where $\rho_{\text{free}}$ is the free charge density at the surface. When $\mathbf{E}_1$ and $\mathbf{E}_2$ are known at the interface, \[ \hat{n} \cdot (\mathbf{E}_1 - \mathbf{E}_2) = \frac{\rho_{\text{total}}}{\varepsilon_0} \], may be used to calculate the total charge density, $\rho_{\text{total}}$, that includes free and bound charges. This is a useful relation in calculating the charge distribution associated with SPPs and LSPs.
4. Normal components of the magnetic flux density are continuous across the interface.

\[ \hat{n} \cdot (\mathbf{B}_i - \mathbf{B}_o) = 0 \]  \hspace{1cm} (2.13)

### 2.2. Poynting Theorem

Maxwell’s equations may be used to establish the relationship governing the fields, currents and their respective energies by combining Ampere’s and the Faraday’s laws which results in [8]:

\[
\int_s (\mathbf{E} \times \mathbf{H}) \cdot d\mathbf{s} = -\int_V \left[ \mathbf{H} \cdot \frac{\partial \mathbf{B}}{\partial t} + \mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial t} + \mathbf{E} \cdot \mathbf{J} \right] dV 
\]  \hspace{1cm} (2.14)

The integral on the LHS, represents the total power flow in and out of the volume V. The term under the integral, \( \mathbf{S} = \mathbf{E} \times \mathbf{H} \), known as Poynting vector, represents the power flux density and it is required for evaluating radiation patterns. The term \( \left( \mathbf{H} \cdot \frac{\partial \mathbf{B}}{\partial t} + \mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial t} \right) = \frac{d \mathbf{w}_m}{dt} + \frac{d \mathbf{w}_e}{dt} = \frac{d \mathbf{w}}{dt} \) is interpreted as the volume energy density per unit time required to establish the electromagnetic field in the medium having a volume V, with the first term being associated with the magnetic field and the second term with the electric field. The Poynting vector is particularly useful in evaluating the radiation pattern of an antenna in the far-field limit, where the electromagnetic fields are in phase and transverse to the propagation direction, z. Hence, considering the simplest case \( \mathbf{E} = (E_x,0,0) \) and \( \mathbf{H} = (0,H_y,0) \), the characteristic impedance of an isotropic, linear and homogenous medium in the far-field is given by \( Z = \frac{E_x}{H_y} = \sqrt{\frac{\mu}{\varepsilon}} \). Therefore, the time-averaged of the Poynting vector may be expressed as:

\[
\left\langle \mathbf{S} \right\rangle = \frac{1}{2} \text{Re} \left[ \mathbf{E} \times \mathbf{H}^* \right] = \frac{1}{2} [E_x, H_y, \hat{z}] = \frac{1}{2} \sqrt{\frac{\varepsilon}{\mu}} \left| \mathbf{E} \right|^2 \hat{z} = \frac{1}{2} \sqrt{\frac{\mu}{\varepsilon}} \left| \mathbf{H} \right|^2 \hat{z} 
\]  \hspace{1cm} (2.15)

The radiation pattern, \( R(\Omega) \) where \( \Omega \) is the solid angle, is related to the Poynting vector via the total outward power, \( P \), crossing a spherical surface that surrounds the source.
In our simulations, the radiation patterns of metallic nanoparticles, or nanocavities, are calculated from the magnitude of the time-averaged radial component of Poynting vector vs. the angular direction.

2.3. Radiative Decay Rate Enhancement

The spontaneous decay rate of a two level quantum system where $|i\rangle$ & $|f\rangle$ denote the initial and final states, is given by the Fermi’s Golden rule:

$$
\gamma = \frac{2\pi}{\hbar^2} \sum |i\rangle \langle H | f\rangle \langle H | i\rangle \delta (\omega_i - \omega_f)
$$

Note that in the spontaneous emission, unlike stimulated emission, emitted photons are not limited to a particular direction/polarization, consequently, the summation is performed over all non-degenerate final states, where each mode is identified by $\omega_k$, i.e. the frequency of the $k^{th}$ mode, characterized by a particular polarization and wavevector. Although spontaneous emission is purely a quantum mechanical effect, it is possible to derive a relation for the decay rate in the terms of the dipole emission. In the dipole approximation, the interaction Hamiltonian operator of a dipole located at $r_0$, is defined by $\hat{H} = -\hat{p} \cdot \hat{E}$, where electric field operator at $r = r_0$ is given by $\hat{E} = \sum_k E_k^+ \hat{a}_k(t) + E_k^- \hat{\alpha}_k(t)$. The creator and annihilator operators are defined as $\hat{a}_k(t) = \hat{a}_k(0)e^{-i\omega_k t}$ & $\hat{\alpha}_k(t) = \hat{\alpha}_k(0)e^{i\omega_k t}$, with $\omega_k$ denoting the frequency of the $k^{th}$ mode. Dipole moment operator is defined as $\hat{p} = \hbar\hat{r}$ with $\hat{r}^+ = |e\rangle \langle g|$ & $\hat{r}^- = |g\rangle \langle e|$ where $|g\rangle$ & $|e\rangle$ are the orthogonal basis indicating the ground and the excited states respectively. Here the cap, $\hat{}$, signifies an operator. Expanding the Hamiltonian in terms of the quantities mentioned above results is:

$$
\hat{H} = -\hbar \sum_k E_k^+ |e\rangle \langle g| \hat{a}_k(0)e^{-i\omega_k t} + E_k^- |e\rangle \langle g| \hat{\alpha}_k(0)e^{i\omega_k t} + E_k^- |g\rangle \hat{a}_k(0)e^{i\omega_k t} + \cdots
$$
The initial and the final states in terms of ground, excited and photon states are given by:

\[ |i\rangle = |e\rangle |0\rangle = |e,0\rangle \]
\[ |f\rangle = |g\rangle |1_{\alpha_k}\rangle = |g,1_{\alpha_k}\rangle \]

Here \( |0\rangle \) denote the zero-photon state whereas \( |1_{\alpha_k}\rangle \) corresponds to a state where the system has one photon associated to the \( k \)th mode. Evaluating the decay rate as stated in equation (2.17) leads to:

\[ \gamma = \frac{2\pi}{h^2} \sum_k \left[ \mathbf{p} \cdot \left( \mathbf{E}_k^* \mathbf{E}_k \right) \right] \delta (\omega_k - \omega_0), \]

where

\[ \mathbf{E}_k^* = \sqrt{\frac{\hbar \omega_k}{2\varepsilon_0}} \mathbf{u}_k \quad \text{and} \quad \mathbf{E}_k = \sqrt{\frac{\hbar \omega_k}{2\varepsilon_0}} \mathbf{u}_k^*, \]

\( \mathbf{u}_k \) are the normal modes and \( \mathbf{p} = \rho \mathbf{u}_p \) with \( \mathbf{u}_p \) being the unit vector in the direction of the dipole moment. Note that \( \mathbf{E}_k^* \mathbf{E}_k \) designates an outer product resulting in a 3x3 matrix. Finally the decay rate can be simplified to [8]:

\[ \gamma = \frac{\pi \omega}{3h \varepsilon_0} |\mathbf{p}|^2 \rho (\mathbf{r}_0, \omega_0), \]

where

\[ \rho (\mathbf{r}_0, \omega_0) = \frac{3}{\pi} \sum_k \left[ \mathbf{u}_p \cdot (\mathbf{u}_k^* \mathbf{u}_k) \cdot \mathbf{u}_p \right] \delta (\omega_k - \omega_0) \]

\{in the case of free space\} \quad \rightarrow \quad \rho_0 = \frac{\omega_0^2}{\pi^2 c^3} \quad \rightarrow \quad \gamma_0 = \frac{\omega_0^3}{3\pi h \varepsilon_0 c^3} |\mathbf{p}|^2

Equation (2.19) shows that the spontaneous decay rate is the function of the local density of states \( \rho \). Furthermore, the density of states depends on the interaction of the dipole moment with its own secondary field arriving at the dipole’s location after being scattered by the environment (such as a cavity surrounding it). Microscopic representation of the decay rate based on quantum mechanics, however, does not account for losses. Alternative representation, based on classical electrodynamic, correlates the spontaneous decay rate, \( \gamma \), to a macroscopically observable quantity such as the power, \( P \), in an inhomogeneous environment by [8]:

\[ \text{decay rate enhancement} \equiv \frac{\gamma}{\gamma_0} = \frac{P}{P_0} \] (2.20)

For derivation see 8.3.3 and 8.5.2 in [8]. Here, \( P_0 \) is the power emitted by a dipole in a homogeneous environment (such as vacuum) and \( \gamma_0 \) is the FWHM of the Lorentzian line associated with the dipole’s frequency dependent emission in the same
environment, see section 8.5.1 in [8] for the origin of the damping factor, $\gamma_0$. Depending on the inhomogeneous medium containing the dipole, some or all of the power emanating from the dipole maybe absorbed leading to an expression of the total power $P = P_{\text{far}} + P_{\text{abs}}$ where $P_{\text{far}}$ and $P_{\text{abs}}$ are the power detected in the far-field and the power absorbed by the medium (e.g. due to losses, surface mode excitations, quenching effects and more). Similarly, spontaneous decay rate should be expressed as $\gamma = \gamma_r + \gamma_{nr}$, where $\gamma_r$ and $\gamma_{nr}$ are the radiative and non-radiative terms [8]. Since $\gamma_r$ is proportional to $P_{\text{far}}$ the power ratio $\frac{P_{\text{far}}}{P_0}$ is a valid quantity, representing the enhancement or inhibition of the radiative part of the spontaneous decay rate, provided that $P_0$ be also measured in the far-field over the same area as $P_{\text{far}}$. The power ratio $\frac{P_{\text{far}}}{P_0}$ is used in this thesis to evaluate radiative decay rate enhancement of a dipole located in a cavity or an aperture relative to that in vacuum.

### 2.4. Surface Plasmon Polaritons

Surface Plasmons, or Surface Plasmon Polaritons (SPP), is a designation given to the quasi-particles representing the collective oscillation of the surface charges in the realm of quantum mechanics. The necessary condition for the excitation of the SPPs modes, from the material point of view, is the existence of a 2D electronic gas formed at the interface between a lossless dielectric having a positive permittivity, e.g. the air, and another material with complex permittivity having a large negative real and small positive imaginary parts. Collective oscillations of surface charges manifest themselves in the form of longitudinal surface waves that propagate along the metals/dielectric interface. Here, the term ‘Polaritons’ suggests the coupling between the polar excitations (i.e. positive/negative charge bundles) of the surface waves to the near surface electromagnetic field that also travel with the SPP. Such waves have a well-defined wavelength. In order to excite a SPP mode by an incident EM wave, the energy and momentum of the incident wave must match those of the SPP. The energy requirement is easily achieved by adjusting the frequency of the incident light. Adjusting the momentum, however, is not as trivial. Consider an interface between a
metallic surface with a complex permittivity \( \varepsilon_1 = \varepsilon'_1 + i \varepsilon''_1 \) and a lossless dielectric medium having a real permittivity \( \varepsilon_2 \).

It can be shown that when \( \varepsilon'_1 \gg \varepsilon''_1 \), components of the wavevector parallel surface satisfy [8]:

\[
k_{spp} \approx \sqrt{\frac{\varepsilon'_1 \varepsilon_2}{\varepsilon'_1 + \varepsilon_2}} k_0 + i \frac{\varepsilon'_1 \varepsilon_2}{\sqrt{\varepsilon'_1 + \varepsilon_2} \ 2\varepsilon'_1 (\varepsilon'_1 + \varepsilon_2)} k_0 \equiv k'_{spp} + i k''_{spp} \quad (2.21)
\]

Here, \( k_0 = \omega / c \) is the free space wavevector and \( k'_{spp} \) is the propagation constant, hence must be real. This condition is satisfied only when \( \varepsilon'_1 \varepsilon_2 < 0 \ & \varepsilon'_1 + \varepsilon_2 < 0 \). It is obvious that \( k_0 \neq k'_{spp} \) even if the momentum of incident light is entirely parallel to the surface. SPPs, therefore, cannot be excited on a flat metallic surface by any incident light from vacuum, see Figure 27(a). To excite a particular SPP mode, an evanescent wave with a matching energy and momentum is required. One approach is the use of a prism, see Figure 27(b)-(d), where the incident light passing through the prism launches the evanescent wave with a wavevector parallel to the surface. This will certainly bring the momentum of the evanescent wave closer to that of the SPP’s. Furthermore, since the incident light is passing through a prism, momentum of the evanescent wave becomes larger by a factor of the prism’s refractive index. The magnitude of the evanescent wavevector may be adjusted by altering the incident angle. The SPP mode are excited when this wavevector matches the real part of the \( k_{spp} \) defined by the equation (2.21).
Figure 27: (a) Dispersion curves for SPP (the curved line), light in vacuum \( \omega = ck_0 \) and the evanescent light produced by the prism \( \omega = ck_0/n \). (b) Otto configuration for generating SPPs. (c) Formation of evanescent wave at base of a prism when incident angle is larger than the critical angle. (d) Kretschmann configuration is an alternative way in generating SPP in metallic film having thicknesses in the order of skin depth [8].

When the wavevector of the evanescent wave matches a particular SPP mode, incident energy couples to the surface and the reflected intensity drops to zero. The latter is known as Surface Plasmon Resonance (SPR) and the incident angle at which this occurs is designated as \( \theta_{SPR} \), see Figure 32 [8].

Figure 28: Reflectivity vs. various incident angles in metallic films with various thicknesses (nm) [8].

In the previous chapter a qualitative explanation of the propagation length of SPPs were provided. Quantitatively, this value can be calculated from the imaginary part of the \( k_{SPP} \)
as $1/k_{spp}^*$, for which the electric field’s magnitude decays to 1/e of its original value. The SPPs’ wavevector components normal to the interface are given by \[8\]:

$$
\begin{align*}
    k_{1,z} &\approx \sqrt{\frac{\varepsilon_1}{\varepsilon_1' + \varepsilon_2}} \left[ 1 + i \frac{\varepsilon_1^*}{2 \varepsilon_1'} \right] k \equiv k_{1,z}' + i k_{1,z}'' \tag{2.22} \\
    k_{2,z} &\approx \sqrt{\frac{\varepsilon_2^*}{\varepsilon_1' + \varepsilon_2}} \left[ 1 - i \frac{\varepsilon_1^*}{2 (\varepsilon_1' + \varepsilon_2)} \right] k \equiv k_{2,z}' + i k_{2,z}'' \tag{2.23} 
\end{align*}
$$

Here, $k_{1,z}$ & $k_{2,z}$ represent the components in metal and dielectric respectively. The propagation length normal to the surface is obtained from the imaginary part of the wavevector. The decay lengths (or penetration depth) normal to the surface in silver and gold, which are also the material of interest in this thesis, are calculated to be 23 nm and 28 nm respectively. Kretschmann and Otto configurations are not the only techniques to generate SPPs. Surface plasmons may also be launched by a subwavelength aperture/cavity perforated in a metallic film. As mentioned previously, subwavelength cavities and apertures in metallic films support LSPR. In the case of a ‘single’ hole perforated in a metallic film LSP within the interior cavity walls can decay not only into free propagating EM waves, but also into cylindrical SPP waves propagating away from the hole along the metallic surface \[135\]. The cylindrical nature of the SPP waves generated by a single cavity makes the cylindrical frame of reference, with unit vectors $(\hat{\rho}, \hat{\phi}, \hat{z})$ where $\hat{z}$ is the normal vector to the metallic surface, ideal for investigating the SPPs launched from the rims of nanoholes. SPP fields propagating away from a single hole follows the complex phasor \[135\]:

$$
E^{\text{spp}} = A \left( \hat{z} - \frac{i}{K_{\text{spp}}} \rho \right) H_1^{(1)}(K_{\text{spp}} \rho) \cos(\phi) \exp(-\alpha z) \exp(-i \omega t) \tag{2.24}
$$

where $A$ is a constant, $H_1^{(1)}(K_{\text{spp}} \rho)$ is the Hankel function of first kind with $m=1$, $\alpha$ is the decay length in the $z$ direction, where $z = 0$ represents the metal/dielectric surface \[8\]. For a given $K_{\text{spp}}$, the amplitude of the phase varies with the distance from the centre of the hole cavity. An azimuthal dependence of the SPP field is a result of the variation in the amplitude of the fields in the holes when excited by linearly polarized light. The strength of the SPPs are governed by $\hat{n} \cdot K_{\text{spp}}$, due to the dependency on $\cos(\phi)$ \[136\].
where $\hat{n}$ is the normal vector from the cavity to an observation point on the metal surface. Therefore, the strength is maximized along the $K_{SPP}$ direction and this direction is established by the component of the incident electric field parallel to the surface of the metal, and hence the polarization angle of the incident light, see Figure 29.

![Figure 29: Surface charge density (red = 1, blue = -1 and green = 0) at glass/silver interface when illuminated by a normally incident light, linearly polarized at -45°, indicates that the strength of travelling SPP waves launched from the rims of the cavity at the centre, follows the complex phasor relation (2.24). $K_{SPP}$ is parallel to the in-plane component of the incident electric field. This direction is defined as $\varphi = 0$. The strength of the SPPs decreases as $\rho$ increases and/or as $|\varphi|$ approaches 90°.](image)

### 2.5. Surface Plasmon Polaritons and Bloch Waves

SPP Bloch waves are exclusive to metallic surfaces with periodic features. These are the plasmonic standing waves [137] formed between the periodic surface features resulted from abrupt changes to the surface geometry. One dimensional metallic gratings is already is discussed in section 1.3.2. Here, the concept is extended to two dimensional periodic structures using arrays of subwavelength holes perforated in a metallic film. SPP Bloch waves on the surface of a metallic hole array follow the conservation of momentum [79, 81, 138]:

$$k \pm iG_x \pm jG_y = k_{SPP}$$

$$k_{SPP} = \text{Re} \left( \frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d} \frac{2\pi}{\lambda_o} \right)$$

(2.25)

where $k$ is the component of the incident wavevector parallel to the surface of the film, $G_x = 2\pi/P_x$ and $G_y = 2\pi/P_y$ are the reciprocal unit vectors of a rectangular array with periodicitics of $P_x$ and $P_y$, $\varepsilon_m$ and $\varepsilon_d$ are the complex permittivities of the metal and the
dielectric respectively, and $i$ and $j$ are integers. For a square lattice the relationship between the SPP Bloch modes and the lattice periodicity is given by [81]:

$$P = \frac{2\pi}{k_{SPP}} \sqrt{i^2 + j^2}$$

(2.26)

Table 1 depicts some of the possible reciprocal vectors supported by a square lattice. Each wavevector is related to a different permuted value of $(i,j)$ in $G = iG_x + jG_y$, e.g. $G_1 = iG_x$ or $jG_y$, $G_2 = iG_x + jG_y$, $G_3 = i2G_x$ or $j2G_y$ and so on. For simplicity, degenerate modes resulting from $-i$ and $-j$ are omitted.

Table 1: SPP Bloch modes in reciprocal space. Possible modes supported by a square lattice. Each wavevector is related to a different combination of $(i,j)$ in $G = iG_x + jG_y$, e.g. $G_1$ is related to the degenerate modes $G_1 = iG_x$ or $jG_y$, $G_2 = iG_x + jG_y$, $G_3 = i2G_x$ or $j2G_y$ and so on. Degenerate modes resulting from $-i$ and $-j$ are omitted.

Equations (2.25) and (2.26) imply that multiple resonant SPP-Bloch modes can coexist in a symmetric hole array. However, whether these modes interplay and influence each other, remains to be seen experimentally.

2.6. Finite Element Method

When dealing with complex electrodynamics problems, exact analytical solutions are either non-existent or oversimplified by generalizations, assumptions and approximations. In most cases, solving real-life scientific problems requires a numerical approach, which produces an approximate solution. Finite element method (FEM) is
one of the popular techniques for solving differential or integral equations numerically. It was initially developed by structural engineers in the late 50s but in the late 60s it was generalized into a technique for numerically solving any partial differential equations (PDE) or integral equations. Recent increases in computational power have made the FEM a popular choice for solving scientific problems numerically. Two main reasons for the FEM’s popularity over other techniques are the flexibility in modelling complex geometry by utilizing polygonal/polyhedral mesh elements and the higher computation accuracy by calculating the functional value over individual mesh elements. Figure 30(a) depicts the discretization of a 2D object into a mixture of different types of polyhedral mesh elements. A single triangular mesh element with vertices labelled 1, 2 and 3 is shown in Figure 30(a). Unlike the finite difference method where the functional value is evaluated only at the vertices, FEM is capable of calculating the values anywhere within the element (and hence their contribution to the whole domain) by interpolating the values at the vertices.

Figure 30: (a) Unlike the finite difference method, FEM mesh elements can be a mixture of different types of polyhedrons with various sizes. (b) Single triangular mesh element within the domain.

With the variety of scientific software available in the market today, the burden of programming is shifted to the third party companies, allowing the scientists and engineers to focus on the modelling and analysis of the core scientific phenomena. COMSOL is an example of such software that offers a variety of solvers for a given problem. Nevertheless, understanding the method and the problem at hand helps in optimization of the model by allowing the researchers to make an informed decision in selecting the most suitable solver for a given problem. For example, the second order linear PDEs in two independent variables of general form
\[
\frac{\partial^2 u}{\partial x^2} + 2b \frac{\partial u}{\partial x \partial y} + c \frac{\partial^2 u}{\partial y^2} + d \frac{\partial u}{\partial x} + e \frac{\partial u}{\partial y} + fu = 0,
\]
may be divided into three categories [139], elliptic (e.g. Laplace, Poisson’s or Helmholtz equations), hyperbolic (e.g. wave equations) and parabolic (e.g. heat equations), depending on the coefficients \(a, b, c, d, e\) and \(f\), i.e.:

- **elliptic**: \(b^2 - 4ac < 0\)
- **hyperbolic**: \(b^2 - 4ac > 0\)
- **parabolic**: \(b^2 - 4ac = 0\)

One of the fastest and most memory-efficient algorithms to solve the elliptic and parabolic PDEs is the geometric multigrid iterative solver [140, 141]. There are ample of published books on FEM and related solvers/algorithms, [130, 131, 142-144], and a complete description of the method under this subheading is not feasible. To gain insight into modelling techniques and strategies, it is best to consult FEM software manuals. In particular, COMSOL is shipped with an extensive model library for users to explore. Sample models are well documented and explained. COMSOL’s user manual is also a rich source of documents describing the building blocks of FEM models. A brief mention of the simulation strategies, however, is made throughout this thesis in the context of a particular design when appropriate.
3. Experimental Setups

Devices proposed in this project were planar in design, involving metallic films with surface features fabricated using the two most popular techniques/instruments namely Electron Beam Lithography (EBL) and Focused Ion Beam (FIB). Throughout this project, various characterization techniques were deployed to determine the optical response of the fabricated devices. Spectral measurements were mainly based on the standard inverted microscopes and spectrographs. Polarization measurements required a slight modification to the standard microscope configuration, whereas the lifetime measurements and the beam profiling required an existing custom-made confocal microscope that was already available in the basement of the David Caro building. A brief introduction to each fabrication technique/instrument and an overview for each microscope setup is provided in this chapter.

3.1. Electron Beam Lithography

Electron beam lithography is a technique for transferring 2D nanoscaled geometric patterns onto a film coated with an electron sensitive resist by exposing it to electron beam current. Depending on the type of resist, i.e. positive or negative, the exposed regions of the resist become more or less soluble than the unexposed regions. Developing the resist removes the selected area, which exposes the film beneath it. The film is then etched until the desired depth is reached. The main advantage of the EBL over the optical/UV lithography is the higher resolution. The resolution in optical lithography depends on the wavelength $\lambda$, and suffers from the diffraction limit, $d = \frac{\lambda}{2n\sin \theta}$, where $d$ is the smallest spot size achieved by the light passing through a medium with a refractive index $n$ and emerging with an angle $\theta$. With UV as the light source, the smallest possible spot size is few hundreds of nanometers. Whereas the wavelength associated with an accelerated electron beam, calculated based on the de Broglie’s wave-particle duality, may only be few pico-meters resulting in a much higher resolution. However, due to the contributions from the secondary electrons, the smallest
spot size achievable in practice is reported to be 4-10 nm [145, 146], i.e. an improvement over the optical lithography’s resolution.

3.1.1. The Instrument

There are strong resemblances between the working components of a scanning electron microscope (SEM) and an EBL. The electron beam column of a SEM is described in section 3.2.1. The only significant difference between a SEM and an EBL is the way electron beam traverses the sample. In a SEM the beam traverses the sample’s surface in a raster scan pattern that covers a rectangular area under examination, whereas in an EBL the beam targets only specific regions of the sample based on the 2D patterns fed into the computer that controls the beam position on the surface. Figure 31 shows the “Vistec EBPG5000 plus” which is available at Melbourne Centre for Nanofabrication (MCN) whose facilities were used throughout this project. It is one of the most advanced EBL instruments available to date, capable of generating structures with a minimum feature size of 8 nm at rapid exposure rate of 50 MHz.

![Vistec EBPG5000plus](image)

**Figure 31: Vistec EBPG5000plus**

3.1.2. Process

First step in EBL fabrication process is the design of the intended structure. EBL is a planar fabrication technique therefore a 2D projection of the structure must first be produced using specialized software such as KLayout. Output generated by KLayout is
a graphic database system (GDS) file. GDS is the de facto industrial standard for the integrated circuit layouts. GDS files are further processed into EBL’s GPF files using Layout Beamer software, incorporating dose matrices, proximity error corrections and other instruction needed to drive the EBL.

Figure 32 illustrates the step by step process involved in fabricating metallic features using EBL. Starting with a substrate such as glass, a layer of gold or silver film is deposited on the top. A resist is then evenly spread over the film using spin coating technique. The final thickness of the resist depends on its chemical composition, spin frequency and the duration of spin. Depending on the recipes, the resist may also require dehydration baking which also influences its thickness and sensitivity to electron charge exposure. Selective area of the sample is then exposed to electron charges based on the patterns defined in the GPF file. The total charge delivered onto the target area depends on the acceleration voltage and the duration that the beam stays on. In this diagram it is assumed the use of positive resist, i.e. areas exposed to the beam will become soluble. Developing the film involves removing the part of the resist exposed to the beam using chemical solutions. Once developed, the film is ready for dry etching process by means of Inductively Coupled Plasma (ICP)/Reactive Ion Etching (RIE). Finally, once the etching process is completed, the leftover resist is striped off leaving the metallic feature on the substrate. The resolution, hence the fabrication quality, depends on complex interplay of various factors. For example, increasing the bias voltage reduces the electron beam wavelength, which presumably improves the resolution. However, increase in electron velocity may also give rise to the number of secondary electrons, and consequently the proximity error, which has adverse effect on the resolution.
3.2. Focused Ion Beam

Focused Ion Beam (FIB) refers to a technique or instrument that utilizes accelerated ions of moderately heavy atoms, such as Gallium, for either analysis or ablation of a material. Apart from being popular among the material scientists, biologist and semiconductor engineers, FIB is one of the main tools deployed in nano-science for fabricating features that are nano-scale in dimensions. During the course of this project, FIB was utilized as the main fabrication tool for milling sub-wavelength apertures or cavities in metallic films. The two organizations that provided access to such instrument, Bio21 and MCN, are in ownership of the Helios NanoLab FEI FIB, see Figure 33. A general description of the main components of FIB instruments is provided in the following section, 3.2.1. A more elaborated description related to the fabrication steps, such as bitmap file preparation and run time parameters, follows in the subsequence section 3.2.2.
3.2.1. The Instrument

Figure 34 depicts the typical components inside a FIB chamber. The two components fundamental to FIB instruments are labelled “E-beam”, which generates the electron beam for SEM functionality, and “I-beam”, which generates the ion beam for FIB functionality. Working components of an E-beam column closely resembles the I-beam column, compare Figure 35(a) and (b). Therefore describing the I-beam column and its interior components applies to the E-beam column.
Figure 34: A Helios NanoLab FEI FIB instrument. Components inside the chamber.

The interior of a typical ion-beam gun (or column) is shown in Figure 35(b)-(c), [147]. To achieve high resolution ion beams, FIB uses techniques referred to as liquid metal ion sources (LMIS) in conjunction with field ionization source (FIS). The idea is to minimize the ion source size, i.e. to reduce the bulk Gallium to nano-scale dimensions, prior to ionization and particle acceleration. Reduction in the ion source size leads to an ion beam that can be focused into smaller diameters.

During the LMIS process, the Gallium reservoir is first coil heated to its melting temperature of \( \sim 30^\circ \), producing a liquid Gallium that flows from the reservoir into a funnel with an exit diameter of \( \sim 20 \mu m \). As the liquid Gallium exits the funnel, it gets moulded into a needle with a diameter no wider than that of the funnel’s exit. At this point the ion source size is reduced from the bulk to a micro-scale dimension. The Gallium needle experiences further reduction in diameter due to the presence of a strong negative potential of \( \sim 30 \) keV between the tip and the extractor, (i.e. an electrode plate with an aperture at its centre), which exerts enough down force to taper the tip of the Gallium needle into Taylor-Gilbert cone with a tip diameter of \( \sim 10 \) nm, hence the LMIS. The strong electric potential between the tip and the extractor also ionizes the Gallium atoms at the tip and generates the necessary acceleration required for field emission, hence the FIS. As the ionized Gallium particles travel down the column, they pass through an electrostatic lens, that acts as a condenser, and then through a variable aperture that limits the current and defines the beam spot size. A fast operating aperture called the beam blanker is also positioned on the path which acts as an on/off switch,
controlling the duration on which the beam is on, thus the dwell time. The beam blanker
has no impact on the current when the beam is on. The beam blanker is followed by a
second electrostatic lens that focuses the beam into an even smaller spot. By this stage
the ion beam is on a straight collision path with the sample. To control the beam
position on the target sample, the beam must pass through an octopole that, besides
adjusting the stigmatism, is responsible for steering the beam. [148-150]. An electron-
beam column shares the same components described above. The main difference,
however, is the particle source which generates free electrons, rather than ions, using
thermionic emission [151].

Figure 35: (a)-(b) A typical electron-beam and ion-beam gun (or column). (c) close up of
the Ion source. [147]

3.2.2. Bitmap File Preparation

Helios NanoLab 600 Focused Ion Beam software is capable of milling patterns,
represented by a 24-bit bitmap file, onto the target sample. The following specifications
must be considered when preparing the bitmap file. Each pixel in a bitmap file
represents a point-location on the sample to be milled. Various parameters such as
dwelling time, ion beam current, degree of ion beam defocusing and relative interaction
diameter (RID) can affect the spot size of the ion beam on the target surface, hence
improving or worsening the resolution and accuracy. These parameters can be set using
the FEI’s FIB software’s GUI prior to the milling, however, it is also possible to control
the dwelling time (which affects the milling depth) at runtime by specifying the pixel
value (i.e. pixel colour) in the bitmap file. In a 24-bit bitmap file, each pixel consists of
red, green and blue components, (i.e. RGB). After the bitmap file is loaded into the
memory, each of these colour components is represented by a byte, (i.e. 8 bits). Therefore, an RGB representation of a single pixel occupies 3 bytes of memory, (i.e. 24 bits). Value of a single byte ranges from 0 to 255 and FEI’s FIB software interprets each of these RGB colour components as follow:

- Red: The red component is currently not used.
- Green: The green component is used to turn on/off the ion beam, with 0 representing “off”, and any other value between 1 and 255 representing “on”.
- Blue: The blue component controls the dwelling time per pixel. This is the duration for which the ion beam stays on. If the value of the pixel’s blue component is set to 0, the dwelling time will be 100 ns, (i.e. the minimum dwell time achievable by the instrument). A value of 255, sets the maximum dwelling time to the value specified by the user in the GUI. Therefore, although the minimum dwelling time is limited by the instrument itself, the maximum dwell time can be adjusted by the user before the milling starts. Any value between 0 and 255 is linearly interpolated from the minimum/maximum values.

3.3. Confocal Microscope (Lifetime and Depth of Field Measurements)

Figure 36 depicts the schematics for the confocal setup mentioned above. Such simple setup has proven to be extremely beneficial to this project. Section (a) shows the position of the light source with respect to the rest of the instrument when operating in the transmission mode. Section (b) constitutes the core of the microscope. In its standard configuration, it consist of a green laser, $\lambda = 532$ nm, as its light source, a dichroic mirror that reflect $\lambda = 532$ nm and transmits $\lambda > 633$ nm at 45° angle, a LU PLAN Nikon 100× objective with a numerical aperture of $\text{NA} = 0.95$ and a 3D piezoelectric translational stage (100 nm step resolution) where the sample is mounted. Light collected from the sample, after passing through the dichroic mirror, passes through two additional filters, $\lambda > 530$ nm and $650 > \lambda > 750$ nm, in order to block all
UV radiations and allow only the emissions associated with the nano-diamonds photoluminescence into the fibre which acts as a pinhole. Section (c) consists of the hardware related to the photon statistics. Collected photons pass through a 50/50 split fiber coupler that outputs them into two channels with one going through a 125 ns fiber-optics delay line. The photon counting module and the two single photon detectors (S.P.D SDPM140 and SDPM100) are integrated with the Picoharp 300 module that carries out the photon counting and time correlation between the events in a 25 ns time window. The confocal setup described thus far operates in the reflection mode where the $\lambda = 532$ nm light source, marked as (1) in the diagram, provides the excitation pump power for characterizing the NV centres. Section (a) was added during the course of this project to cater for the beam profiling that required the collection of the transmitted power at various distances away from the sample. Incident light on the back of the sample, marked as (2) in the diagram, could be sourced from a laser or an incandescent light source such as a halogen or Supercontinuum Fianium SC-450-2.
Figure 36: Schematics for the confocal setup
3.4. Inverted Microscope (Spectroscopy and Polarization Measurements)

Figure 37 shows a typical inverted microscope, consisting of a halogen light source, a condenser that can either focus or collimate the light, a stage where the sample resides. The quarter wave plate, QWP, and the two polarizers, P1 and P2, and the spectrometer attached to the outport, which had a sole purpose of spectroscopy in bright and/or dark field modes. To customize the microscope for polarization measurements a special mount was fabricated to house P1. However, the custom-built mount can also house additional optical components in series, e.g. P1 in conjunction to a QWP or an optical filter. To accommodate the QWP and the P2, the spectrograph was simply pulled back to allow space for the rotation mounts housing the P1 and the QWP. Pulling the spectrometer away from its designated position, however, introduces a change in the optical path leading to the slit plane and the image plane not to coincide. To correct this, after pulling back the spectrometer, a sample was mounted on the microscope stage and, using the microscope’s oculars, it was positioned at the focal plane of the objective. Spectrometer’s CCD was then activated and the image displayed on the PC screen (which was initially blurred) was monitored while fine adjustments were made to the stage until a clear image was obtained. To ensure correct alignment of the spectrometer with the optical axis of the outport, spectrum of the light source was measured through a transparent glass (as the sample) with the spectrometer at its original and then at its pulled back position. Spectra were found to be identical in both positions. The optimum pulled back position was then marked to ease the reconfiguration process. Such reconfiguration, however, is not advisable for a spectrometer without a built-in CCD.

For this project a Nikon inverted microscope (Eclipse Ti-U) with a Nikon 40x NA = 0.6 objective was used. The condenser/collimator was a Nikon C-C achromatic condenser operating in collimator mode. P1 and P2 were both ThorLabs LPVIS050-MP (550-1500 nm) linear polarizers. QWP was a ThorLabs AQWP05M-600. The spectrometer was an Andor Shamrock 303i-A spectrometer with Andor iDus DU920P-BR-DD CCD, controlled by Andor Solis 4.21 software.
Figure 37: An inverted microscope customized for polarization measurements.
4. Wavelength Dependent Optical Steering

In this chapter, we examine the possibility of rotating the radiation pattern scattered by a pair of nano-rods (or a single asymmetric nano-cross) by controlling the wavelength of the incident wave. The question is whether a pair of nano-rod can potentially function as a wavelength division multiplexer/demultiplexer and if so, under what conditions. An analytical solution is proposed followed by numerical models that confirm the theory.

Optical dispersion in materials leads to the spatial or angular separation of light rays based on their frequency, or wavelength. Dispersion phenomena have their physical origin in the presence of one or more optical resonances due to the frequency-dependent refractive index, resulting from the phase gradient along an interface, with a length scale of the order of the wavelength of light or larger. The different frequency components of a light wave propagate with different phase velocities which separate when incident at an angle on an interface between two optical materials. Strong localized optical resonances also occur in metallic nanoparticles due to the excitation of LSP. The nanoparticles can be significantly smaller than the wavelength of light and can be thought of as optical antennas, [152, 153]. The direction of radiation of the optical antenna can be controlled by the antenna design, suggesting the possibility of localized optical beaming at the subwavelength scale without the need for spatial phase gradients [154]. However, in absence of phase gradient, previous attempts to demultiplex the incident light in subwavelength scales, were mainly limited to the separation of the multi-wavelength SPPs and direct or focus the SPPs towards an observation point associated to a specific wavelength,[96, 155, 156].

4.1. Theory

Here we show theoretically the possibility of localized optical beaming using a pair of orthogonal metallic nanoparticles, and a resonant mode of each to control the intensity and direction of scattering as the incident radiation sweeps though the LSP resonances. This effect represents a new form of angular beaming of light that does not rely on propagation phase shifts or diffraction, with applications in wavelength division
demultiplexing for optical computing and communication, \[157\]. We consider the configuration shown in Figure 38a where two nanorods are placed and excited such that their resonant modes are orthogonal and different in frequency. A linearly polarized plane wave incident from above has a polarization angle $\psi$ chosen to excite both modes simultaneously. The direction of scattering depends on the orientation of the induced net dipole moment. If the applied light is resonant only with the first mode in Figure 38a, then the light is scattered predominantly in the $y$-$z$ plane since the mode dipole moment is oriented in the $x$ direction. As the frequency moves off resonance with the first mode, but into resonance with the second, the radiation is scattered in the $x$-$z$ plane since the second mode has a dipole moment in the $y$ direction. This effect leads to angular beaming of the incident light.

Figure 38: (Color online) a) Two nanorods with distinct orthogonal resonant modes that radiate light perpendicular to their dipole moments (shown by the dashed arrows). The angle $\psi$ defines the direction of polarization of the incident light. b) The radiated power (solid line) and the range of angles over which radiation is dispersed (dashed line) as functions of the normalized frequency difference between the nanorods’ resonances. The radiated power is expressed as a ratio of $dP/d\Omega$ at the frequency of one of the resonances to $dP/d\Omega$ at a frequency midway between the two resonances.

Ideally we require the resonant frequencies such that the scattered power is constant and the direction is proportional to the applied frequency. We derive the key relationships required to satisfy these conditions using an electrostatic method based on an eigenmode representation of the LSP for coupled nanoparticles, \[158\]. This method is largely independent of the shape of the nanoparticle allowing a general description without requiring the details of the geometry. The excitation of a given resonance mode
$j$ in nanoparticle $p$ excited by an incident field $\mathbf{E}_0 \exp(-i\omega t)$ is described by an amplitude $a'_p(\omega)$ which can be written in an approximate form [159] as

$$a'_p(\omega) \approx -\frac{A'_p \mathbf{p}'_p \cdot \mathbf{E}_0}{\omega - \omega'_p + i\Gamma'_p / 2}$$

(4.1)

where $\mathbf{E}_0$ is approximately constant over the surface of the nanoparticle and the vector $\mathbf{p}'_p$ is proportional to the dipole moment of the resonant mode, [160]. Here $\omega'_p$ is the resonance frequency of the mode, $A'_p$ is a constant and $\Gamma'_p$ is the loss term at the resonance of mode $j$ arising from the Drude model for the metal dielectric.

From Maxwell’s equations, it can be shown that the lowest order contribution to the emitted radiation is from the oscillating dipole moment $\mathbf{p} \exp(-i\omega t)$ of the nanoparticle. The time averaged power per unit solid angle radiated into the far-field in direction $\hat{n}$ is given by, [161],

$$dP / d\Omega = (ck^4 / 32\pi^2\epsilon_0)(\hat{n} \times \mathbf{p} \cdot (\hat{n} \times \mathbf{p}))$$

where $c$ is the speed of light, $k$ is the wavenumber and $\epsilon_0$ is the permittivity of free space. The radiation pattern from an ensemble of interacting nanoparticles will, to lowest order, depend only on the sum of the induced dipole moments $\mathbf{p}(\omega) = \sum_p \mathbf{p}_p(\omega)$, therefore the polarization of the scattered radiation follows the resultant dipole moment. The dipole moment for each particle is given by a sum over the resonant modes $\mathbf{p}_p(\omega) = \sum_j a'_p(\omega) \mathbf{p}'_p$, however, for the pair of nanoparticles shown in Figure 38a, we only consider the dominant resonance mode in each nanoparticle hence eliminating the sum over the modes.

The conditions required for a constant scattered power and a scattering direction proportional to the applied frequency is found for the pair of nanoparticles in Figure 38a, labelled 1 and 2, with two modes in the frequency range of interest. The two nanoparticles lie in the $x$-$y$ plane and their modes have dipole moments $\mathbf{p}'_1 = p'_1 \hat{x}$ and $\mathbf{p}'_2 = p'_2 \hat{y}$, and the incident field has a polarization $\mathbf{E}_0 = E_0(\hat{x}\cos\psi + \hat{y}\sin\psi)$. In spherical coordinates, the point of observation is in the direction $\hat{n} = \sin\theta_o(\hat{x}\cos\phi_o + \hat{y}\sin\phi_o) + \hat{z}\cos\theta_o$. For our analysis we simplify the amplitudes, as
in equation (4.1), by assuming that \( A_1(p_1^1)^2 = A_2(p_2^1)^2 = Ap^2 \), \( \Gamma_1 = \Gamma_2 = \Gamma \). With the incident polarization \( \psi = \pi / 4 \), the power radiated parallel to the x-y plane has the form

\[
\frac{dP}{d\Omega} = \frac{ck^4A^2p^4E^2}{32\pi^2\epsilon_0} \left( \frac{A - B\sin 2\phi_c + C\cos 2\phi_c}{2(A + C)(A - C)} \right)
\]

(4.2)

where \( A = \tilde{\omega}^2 + \Gamma^2 / 4 + \Delta^2 \), \( B = \tilde{\omega}^2 + \Gamma^2 / 4 - \Delta^2 \), \( C = 2\tilde{\omega}\Delta \), and \( \omega_\lambda = (\omega_2 - \omega_1) / 2 \) is the average of the two resonances, \( \Delta = (\omega_2 - \omega_1) / 2 \) is half the difference and \( \tilde{\omega} = \omega - \omega_\lambda \).

Because of the overlap between the light emitted from the two modes, there will be some direction of observation \( \phi_{\text{max}} \) in x-y plane where the radiated power is a maximum. Differentiating equation (4.2) with respect to \( \phi_c \) and setting the result to zero gives

\[
\phi_{\text{max}} = -\frac{1}{2} \arctan \left( \frac{\tilde{\omega}^2 + \Gamma^2 / 4 - \Delta^2}{2\tilde{\omega}\Delta} \right)
\]

(4.3)

which depends only on the frequencies of the LSP resonances and on their damping (linewidths). When the frequency of the incident light is tuned to one of the resonances, \( \tilde{\omega} = \Delta \), then the maximum radiated power is in a direction \( \phi_{\text{max}} = -(1/2)\arctan(\Gamma^2 / 8\Delta^2) \). This depends on the ratio of the resonance widths, as determined by \( \Gamma \), and the resonance separation 2\Delta. For resonances far apart in frequency, then \( \Delta / \Gamma \gg 1 \) and the maximum radiated power is in a direction \( \phi_{\text{max}} = -\pi / 2 \) or 0. Although this gives the largest angle change of the scattered radiation with frequency, the situation is not optimum because the radiated power will vary with angle.

We want to choose the ratio \( \rho = 2\Delta / \Gamma \) so that the power remains approximately constant with frequency. A measure of the change in the radiated power over a frequency interval is the ratio

\[
\frac{dP(\tilde{\omega} = \Delta) / d\Omega}{dP(\tilde{\omega} = 0) / d\Omega} = \frac{(\rho^2 + 1)^2(4\rho^2 + 1 + 2\rho^2 + 1)}{2(4\rho^2 + 1)}
\]

(4.4)
based on equation (4.2) evaluated at \( \phi_{\max} \) obtained from (4.3). This ratio compares the maximum radiated power at the resonance of one mode \( \omega = \omega_A + \Delta \) with the maximum power at the frequency midway between the resonances, \( \omega = \omega_A \).

Plots of the power ratio and the angle \( \pi / 2 + 2 \phi_{\max} \), which is the angular range over which beaming occurs, are shown in Figure 38b as functions of \( \rho = (\omega_2 - \omega_1) / \Gamma \). The angular range varies from 0, for \( \omega_2 = \omega_1 \) where \( \rho = 0 \), to a maximum of 90° for large differences \( \Delta \) in the resonances. As discussed above, the radiated power per solid angle varies greatly for large \( \Delta \) as shown in the figure. This power variation is less than about 12% for \( \rho \leq 0.6 \). This is the key relationship that we need to satisfy to minimize the power variation with frequency. In this regime, changes in the frequency of the incident light direct radiation over an angular range of 35°.

4.2. T configuration: Detached Two Nanoantennas

Using the above design criteria, we have modelled two gold nanorods using the finite element method as implemented in COMSOL Multiphysics 4.2a utilizing the scattered field formulation with plane wave illumination propagating in the \( z \)-direction and linearly polarized in the \( x-y \) plane. A spherical region with diameter 750 nm was modelled and the outer surface terminated in a perfectly matched boundary layer. The relative permittivity of gold was obtained from tabulated data [162] and the background permittivity was \( \epsilon_b = 2 \). Each mode is associated with one nanorod where the resonant frequency is controlled by its length. For the first rod 57 nm long and 24 nm in diameter, the simulated scattering spectrum has a peak at \( \lambda_{\text{res1}} = 697 \text{ nm} \) and a FWHM of 25 nm, see Figure 40(a). Using \( \rho \leq 0.6 \) requires the second arm to have a resonance at \( \lambda_{\text{res2}} = 712 \text{ nm} \). This condition was achieved with a nanorod having the same diameter but 61 nm long, see Figure 39.
Figure 39: Schematic representation of the two nanorods in T configuration. Surface charge density (red = 1, blue = -1 and green = 0) clearly indicates the dipole formation along each nanorod.

To match the scattering magnitude from each mode at each resonance, as assumed in our derivation, the polarization of the incident light was set to $\psi = 44^\circ$, favouring the excitation of the first mode in nanorod-1 to compensate for its smaller geometrical cross section.

Figure 40: Based on a numerical solution of Maxwell’s equations: (a) The scattering power at $\psi = 0^\circ$ and $\psi = 90^\circ$ polarizations. At $\psi = 0^\circ$, excitation is limited to nanorod-1 (57 nm long), whereas at $\psi = 90^\circ$, excitation is limited to nanorod-2 (61 nm long). Simulated scattering spectra have peaks at $\lambda_{res1} = 697$ nm and $\lambda_{res2} = 712$ nm. Both spectra have FWHM of 25 nm. (b) The scattering power maxima and the relative angles at which they occur. The two sets of curves in (b) relate to the lower and upper scattering maxima shown in (a).

These conditions produced a band-pass spectrum with an almost flat-top feature in the region $\lambda_{res1}/\lambda_{res2}$ leading to angular beaming with constant far-field strength, Figure 40b. These far-field radiation patterns show a beaming angle range of up to $25^\circ$ for a wavelength change of 12 nm where intensity remains constant, see Figure 41.
This is consistent with our analytical result. It should be noted that this subwavelength system does not lead to a perfect separation of light frequencies with angle but results in a significant overlap which is determined by the natural widths of the LSP resonances. Alternatively, this system can be thought of as a subwavelength scale optical antenna that exhibits a frequency-dependent beaming of the incident radiation. That is, the direction of scattering is wavelength-dependent. This system provides a mechanism for controlling the direction of radiation through small changes in the frequency of the light.

4.3. Cross configuration: Single Nanoantenna

As mentioned previously, theory derived in the subheading 4.1 is independent of the shape of nanoparticles, yet largely dependent on the two orthogonal LSP modes satisfying the equation (4.4) for \( \rho \leq 0.6 \). To demonstrate the validity of the concept in the case of a single nanoparticle, an asymmetric cross-shaped copper nanoparticle having an arm-width \( W = 24 \) nm was modelled in a similar fashion. For an arbitrary centre wavelength \( \lambda = 835 \) nm, the two fundamental LSP modes \( \lambda_{\text{res1}} \) and \( \lambda_{\text{res2}} \) were found to be 815 nm and 885 nm corresponding to the arm-lengths \( L_1 = 80 \) nm and \( L_2 = 95 \) nm respectively. FWHM of \(~95\) nm associated to copper satisfies the \( \rho \leq 0.6 \) over a wider wavelength range in comparison to the gold nanoparticles previously modelled. Needless to say that the choice of material is purely driven by the application. Figure 42 depicts the radar cross section spectra vs. the incident polarization for the asymmetric
cross-shaped nanoparticles describe above at various incident polarization angle. At incident polarization, $\psi = 32^\circ$ LSPs along both arms have equal strengths and the scattered power is almost constant over the $\lambda_{\text{res1}} - \lambda_{\text{res2}}$ continuum.

![Graph showing radiation cross section spectra vs. incident polarization for asymmetric cross-shaped nanoparticles](image)

**Figure 42:** Radiation cross section spectra vs. the incident polarization for the asymmetric cross-shaped nanoparticles with arm-lengths $L_1 = 80$ nm, $L_2 = 95$ nm and arm-width $W = 24$ nm. $\lambda_{\text{res1}} = 815$ nm and $\lambda_{\text{res2}} = 885$ nm and FWHM is $\sim 95$ nm. At $\psi = 32^\circ$, (solid green line), scattered power is almost constant between $\lambda_{\text{res1}}$ and $\lambda_{\text{res2}}$.

Far-field radiation pattern in the x-y plane showed a total beam steering angle of 55° is achievable for $\lambda_{\text{res1}} < \lambda < \lambda_{\text{res2}}$, see Figure 43. However, unlike the gold T nanorods, radiated power drops between the 2 maxima. The other noteworthy feature is the 2-fold symmetry in the radiation pattern attributed to the cross shape, which is not present in the case of the gold T nanorods.

![Graph showing far-field radiation pattern in x-y plane](image)

**Figure 43:** Far-field radiation pattern in the x-y plane. Total beam steering angle of 55° is achieved for $\lambda_{\text{res1}} < \lambda < \lambda_{\text{res2}}$.  

62
Figure 44 depicts the range of wavelengths over which the beam steering occurs. The ratio $\Theta = \frac{\Delta \phi_{\text{max}}}{\Delta \lambda}$, i.e. the angular beam steering per unit wavelength, can be used as a quantitative measure for the device performance. For the asymmetric copper cross this quantity measures 2.75 degrees/nm over the beaming range, implying a higher beam steering with respect to changes in wavelength compared to that of the gold T nanorods with $\Theta = 1.6$ degrees/nm. However, the increase in $\Theta$ is mainly due to the larger FWHM associated to the copper.

**Figure 44:** Angular positions of the maxima of the far-field vs. the wavelength for the asymmetric copper cross nanoparticle.

### 4.4. Conclusion

In conclusion, we have shown that a pair of nanoparticles, as well as a single cross-shaped nanoparticle, exhibiting two distinct resonant modes can be optimized to achieve "beaming" of incident light for a particular spectral bandwidth. The antenna exploits the LSP resonances within the metal nanorods, exhibiting a new form of angular beaming that does not rely on propagation phase shifts or diffraction effects. The optimization of the subwavelength nanoantennas is based on an analytical method describing the LSPs of metallic nanoparticles. By direct comparison with full-field numerical solutions, we found that, in spite of the number of simplifications implied in our electrostatic approach, the method captures the fundamental aspects that determine the angular radiation pattern of the nano-antennas.
5. Fabrication of Resonant Aperture Antenna Arrays

In this chapter comparisons are made between two different techniques in fabricating arrays of subwavelength cross-shaped apertures in metallic screens. The aim is to determine the most appropriate fabrication technique to be used for the rest of the project (considering the cost, ease of fabrication and accuracy in which the apertures were created). To put the fabrication and the characterization of such array into perspective, an array of symmetric cross-shaped apertures is first modelled using FEM prior to fabrications. This is an important step as it determines the target dimensions to be fabricated for a given transmission spectrum.

Ever since the enhancement of extraordinary optical transmission (EOT) through arrays of sub-wavelength cylindrical apertures perforated in a silver film was reported [79], 2D aperture arrays in metallic films have attracted immense interests among scientists and engineers. They have already been utilized in nano-scale colour filters [104, 105], sensor arrays [163, 164], Raman spectroscopy [165], optical elements [129, 166-168]…etc. With their wide range of applications in physics, chemistry, biology and material science [169], 2D plasmonic aperture antenna arrays play an important role in the future of nanoscience. In this section, attention is drawn to the fabrication techniques of arrays of symmetric cross-shaped apertures. Unlike a simple nanoslit, the transmission response of a sub-wavelength symmetric cross-shaped aperture is independent of the incident polarization. Furthermore, due to the aperture’s geometrical symmetry in the x-y plane, it does not act as a polarization converter, which makes it ideal for nano-scaled optical filter design [104].

5.1. Symmetric Cross-shaped Aperture Antennas (Simulation)

In general, to investigate LSPR modes in apertures reliably, one must first eliminate any possible SPP-LSP couplings. LSP modes are function of the cavity dimensions whereas
the SPP modes depend mainly on the array periodicities. To segregate the resonant SPP modes away from the target wavelength \( \lambda = 700 \text{ nm} \), array periodicity of \( P = 250 \text{ nm} \) is chosen. According to equations (2.21) and (2.25), at this periodicity, the cut-off wavelengths for the \((1,0)_{\text{SPP-air}}\) and \((1,0)_{\text{SPP-glass}}\) for silver fall at \( \lambda = 256 \text{ nm} \) and \( \lambda = 403 \text{ nm} \) respectively, and are well away from the target wavelength. Subsequently, a unit cell within the array was modelled in COMSOL, see Figure 45(a). The top and the bottom interfaces were terminated with ports and the side interfaces were set to periodic boundary conditions to mimic an infinite array. The incident wave was polarized at 45°, although any polarization angle would produce the same response. With the initial parameters of \( P = 250 \text{ nm}, h = 40 \text{ nm}, W = 40 \text{ nm} \) and \( \lambda = 700 \text{ nm} \), a parametric sweep over the arm-length, sets the optimum value to \( L = 145 \text{ nm} \) where the maximum EOT is observed, see Figure 45(b). The impact of variations in the arm-width and the film thickness was also investigated for \( L = 145 \text{ nm} \), see Figure 45(c)-(d). An increase in arm-width is accompanied by a noticeable blue-shift in LSPR, at a rate of \( \Delta \lambda / \Delta W = -3.834 \), and a slight increase in transmitted power. A similar trend in resonance shift is observed with respect to the increase in film thickness at a rate of \( \Delta \lambda / \Delta h = -1.25 \) and a more significant drop in the transmitted power.

Figure 45: (a) schematic of a unit cell with periodicity \( P \), used for modelling the cross-shaped aperture array having arm-lengths \( L \), Arm-widths \( W \) perforated in a silver film having thickness \( h \). (b) Parametric sweep over the arm-length with initial parameters of
5.2. Fabrication - Electron Beam Lithography

The sample was prepared as part of a training session at Melbourne Centre for Nanofabrication (MCN), where a microscope slide, (i.e. glass), was used as a substrate. The substrate was cleaned in a successive ultrasonic bath of acetone and isopropanol for 1 min/solvent followed by a 30 sec rinse in deionized water before being air-dried. Preparation of the thin film was carried out at MCN using IntlVac Nanochrome II electron beam evaporator, by first depositing a 2 nm Germanium layer, which acts as an adhesion layer between the glass and the silver, followed by 40 nm silver and 6 nm SiO$_2$ layers. The SiO$_2$ layer acts as a protective layer against the abrasion and/or oxidization of the silver film, with no significant impact on the overall optical properties. The sample was spin-coated with a diluted version of ZEP, (ANISOLE:ZEP 2:1), at 2000 RPM for 1 minute and baked at 180 degrees for 2 minutes, which yield a 120 nm thick ZEP layer on top of the silver film. Note that ZEP was chosen due to its reasonably high post-spin thickness, hence a higher resolution after the exposure and higher resistance to the etching processes. EBL patterns for a set of cross-shaped aperture arrays, with arm-lengths $L$ ranging from 120 nm to 170 nm, arm-width $W = 50$ nm and array periodicity $P = 250$ nm were prepared in the form of GDS files using KLayout software. GDS files were further processed into EBL’s GPF files, incorporating dose matrices, using the “Layout BEAMER” software at MCN. The optimal amount of energy, in the form of incident charge per unit area on the resist, may vary depending on the type of resist and geometrical aspects of the target feature. A dose matrix is particularly useful in determining the amount of energy required to optimally produce sub-wavelength features. For our target structures, 860 $\mu$C/cm$^2$ produced the best outcome. After the exposure, the film was developed in ZED solution, (AZ726), at 53 degrees for 1 minute. Etching process was carried out using a Reactive Ion Etching (RIE) with Argon gas for 5 minutes. Gas flow was set to 40 Standard Cubic Centimetres per Minute (sccm), with RIE power set to 200W.

Post-fabrication characterizations were carried out by first examining the back-scattered light from each antenna array under an optical microscope, see Figure 46. Variations in
observed colours are consistent with the variations in arm-length. Cavities with longer arm-lengths transmit the red component of the incident white light and back-scatter the rest, hence appear green due to the presence of blue, green and yellow components. Cavities with shorter arm-lengths, on the other hand, transmit the shorter wavelengths and reflect the longer wavelengths of the spectrum, giving them their reddish appearance. However, under a careful observation one notices a disparity in colour texture within each array caused by irregularity in the apertures’ shape and dimensions. SEM images, see Figure 47, confirms the inhomogeneity of the crosses within a single array. Besides the irregularities in shapes, fabricated arm-lengths ranged from 128 to 143 nm vs. the target arm-length of $L = 150$ nm as specified in GDF files. The array periodicity, surprisingly, measured $P \approx 250$ nm, that is very close to the design parameter.

Figure 46: Optical image of arrays of cross aperture fabricated with EBL. Each array is 12x12 μm$^2$. 
Figure 47: (a) SEM images of the crosses with arm-lengths ranging from 128 nm to 143 nm which corresponds to the target arm-length \( L = 150 \) nm as specified in GDF files. (b) Inhomogeneity in shapes and dimensions of the crosses is an indication of the poor quality when fabricating aperture arrays using EBL.

Arrays were further characterized using an inverted microscope in dark-field mode with a dry dark-field condenser having a numerical aperture \( NA = 0.95-0.8 \). The sample was illuminated from the substrate by an incandescent unpolarized light. Transmitted intensities were collected from the air side by a 40× objective and analysed by a Princeton Instruments spectrometer. Using the WinSpec software that controlled the spectrometer, dispersed spectra were recorded into two “.SPE” files, (a Princeton Instruments’ proprietary format) where each image was taken with a single exposure for 10s. Two SPE files (each containing three arrays) required post-processing in order to extract the spectra but offer an advantage of recording additional information such as exposure time, wavelength resolution, background emission, dark current, temperature…etc. Image representations of dispersed spectra are shown in Figure 48. Each bright band corresponds to an array. The \( x \)-axis represents the wavelength, (averaged over the width of the array in the horizontal direction) and the \( y \)-axis represents the spatial position in the vertical direction. Therefore the pixel intensity at \((x,y)\) is the measure of the spectral amplitude at a specific wavelength \(x\) at position \(y\) along the length of the array.
Figure 48: Image representation of the SPE files. The x-axis represents the wavelength and the y-axis represents the spatial position of the pixel. Pixel intensity at (x,y) position is the measure of amplitude at a specific wavelength and position. a) from top to bottom, 170, 160, 150 nm arm-lengths, b) from top to bottom, 140, 130, 120 nm arm-lengths.

Examining the wavelength distribution over the length of each array allows us to probe the homogeneity of the cavities within a particular array, hence the accuracy and consistency of the fabricated. Figure 49 to Figure 54 show different aspects of the results for each array. In each figure, (a) is the image cut out from Figure 46. (b) Shows the post-processed normalized dispersed spectrum \((I-I_{\text{min}})/(I_{\text{max}}-I_{\text{min}})\) with red = 1 and blue = 0. Here, \(I\), \(I_{\text{min}}\) and \(I_{\text{max}}\) are the intensity measured, the minimum local intensity (or the background) and the maximum local intensity respectively. (c) Depicts critical lines where maximum intensities are observed within the array. Additionally, a single spectrum produced by averaging the intensities over the length of the array (labelled as “Lines \(y_{\text{start}}-y_{\text{end}}\)”), is also included. (d) Represents the averaged intensity distribution over the length of the array.

Figure 49: \(L = 170\) nm

Figure 50: \(L = 140\) nm
The resonance of the array with arm-lengths $L = 170$ nm, was closer to the target wavelength $\lambda = 700$ nm, see the consolidated plot in Figure 55. This is in contrast to our simulations that set the arm-length to $L = 145$ nm. Uneven intensities and the presence of double peaks within each array is a testimony to the inhomogeneity in apertures’ dimensions. The inhomogeneity in shapes and dimensions of cross-shaped apertures are clearly due to the fabrication process. Next section entails an entirely different fabrication process based on Focused Ion Beam (FIB).
5.3. Fabrication - Focused Ion Beam

In this section, a different technique for fabricating arrays of cross-shaped apertures is employed, namely the Focused Ion Beam (FIB). Film deposition and the sample preparation for a 40 nm thick silver film is carried out in the same manner as explained in section 5.2. Fabrication process using a FIB instrument, however, requires the preparation of bitmap files comprising of the patterns to be milled, as discussed in section 3.2.2. Possibilities in bitmap preparation for an array of cross-shaped apertures with certain dimensions are endless. Combination of bitmap resolution, the use of bitmap colours, ion current, beam overlaps, dwelling time...etc. all impact the quality of the milled feature. During the course of my project, I investigated various techniques in fabricating cross-shaped apertures, some of which led to a higher fabrication quality. This chapter is divided into three sections, each corresponding to a different technique.

5.3.1. Black and White, 10 nm per Pixel

For the first trial, a 24 bit bitmap file, in black and white, was prepared for an array of cross-shaped apertures having arm-lengths $L = 150$ nm and arm-widths $W = 50$ nm and a periodicity of $P = 250$ nm. With ion current set to $I = 1.5$ pA and the relative interaction diameter of 35%, the ion beam spot size is approximately 10 nm in diameter.
Based on this, cavity dimensions were scaled into their pixel representation. The arm-length $L = 150$ nm is represented by 15 pixels, arm-width $W = 50$ nm by 5 pixels and periodicity $P = 250 \times 250$ nm by a $25 \times 25$ pixels square area, see Figure 56(a). Milling was performed with various dwelling time to determine the optimum value responsible for the highest quality. For our target pattern, the optimum dwell time of 2.2 ms produced features resembling cross-shaped apertures, however, the fineness was less than the desirable level of precision. Clearly, the crosses were over-milled at the centre, see Figure 56(b)-(d).

![Figure 56](image)

Figure 56: (a) bitmap representation of the cross-shaped aperture antenna array. Dimensions in pixels: arm-length $L = 15$ pixels, arm-width $W = 5$ pixels and periodicity $P = 25$ pixels. (b)-(c) SEM images taken at $52^\circ$ tilt angle corresponding to milled arrays with dwell times 2 ms, 2.2 ms and 2.5 ms respectively.

5.3.1.1. Grey Scale, 10 nm per Pixel With

In the previous attempt, fabricate crosses were over-milled at the centres. The anisotropy is partially due to the ion beam having a Gaussian profile and partially due to the secondary ions bouncing off the edges of the cavity’s boundaries towards the centre. Cavity centres, consequently, receive more charge per unit area, hence the over-milling. To remedy the problem a new bitmap file was prepared which took advantage of the
correlation between the pixel values (i.e. the colour) and the dwelling time. In the new
bitmap file, the area occupied by a cross was divided into four different regions each
with a different grey scale value, see Figure 57. The centre lines are drawn using the
darkest grey as the cavity centre needs less exposure. Moving away from the centre,
lines become lighter until the outermost boundaries which are in pure white (i.e.
[R,G,B]=[255,255,255]). Furthermore, the pixels at the inner corners of the crosses are
set to black to switch the ion beam off at those locations. This helps the milled geometry
to resemble a cross more pronouncedly.

![Figure 57: bmp pattern used, note the black inner corners](image)

With the ion current set to \( I = 1.5 \) pA, the impact of variation in the RID and dwelling
time on the milling quality were examined. A 10% RID results in a beam diameter of
7.7 nm. Milling the crosses with 2 ms dwell time, marked the geometries on the metal
surface at a depth less than the film thickness, see Figure 58(a)-(b). A second attempt
with 0% RID reduced the effective beam diameter to 7.5 nm and increased the delivered
charge per unit area and consequently the mill depth, see Figure 58(c)-(d). Increasing
the dwell time to 3 ms, Figure 58(e)-(f), and then to 4 ms, Figure 59, gradually
improved the fabrication quality. The end results, however, showed irregularities in
milled crosses.
Figure 58: (a)-(b) Current $I = 1.5$ pA, dwell time = 2 ms, 10% relative interaction diameter, total beam diameter of 7.7 nm, (c)-(d) relative interaction diameter reduced to 0, total beam diameter of 7.5 nm. (e)-(f) dwell time set to 3 ms.
5.3.2. Lines Segments

An alternative approach to the bitmap preparation for the cross-shaped apertures is the use of line segments to define the arms. Assuming a 10 nm per pixel resolution, an arm-length of 150 nm translates to a single line, (or number of successive line segments), that measures 15 pixels in total. In 5.3.1 the over-milled centres were attributed to the secondary ions bouncing off towards the centre. To avoid this intrinsic FIB behaviour, a new bitmap file was prepared where each cross is represented by four nonintersecting line segments, Figure 60. In this technique, the effective diameter of the ion beam determines the arm-width.

**Figure 59**: (a)-(c) Current \( I = 1.5 \) pA, dwell time = 4 ms, 0% relative interaction diameter, total beam diameter=7.5nm.

**Figure 60**: bitmap patterns for crosses 150 nm in arm-length. Each arm is designed a two successive line segments that measures 15 pixels in total. A central area measuring cell/6 at the centre of each cross is left blank to prevent over milling.
As a test run, a 10×10 version of the above bitmap was prepared. The beam current and the dwell time were set to 1.5 pA and 4 ms respectively. Other parameters were set to their default values by the Ag_photonics script. The instrument set the effective diameter of the ion beam to 7.5 nm. Note that Ag_photonics is a script developed at MCN, comprising a number of parameters optimized for milling silver films. A test run with a single pass, produced an array with a relatively high quality in comparison to previous attempts, see Figure 62. The arm-lengths measured ~155 nm and ~135 nm. The arm-width of ~50 nm, (although very close to the design parameter), is wider than the ion beam diameter. A close examination of the SEM images, see Figure 62(a), revealed that the apertures are over-milled deeper than intended and subsequently into the substrate. This is the consequence of a longer than needed dwell time which can be adjusted accordingly. A remarkable observation here is the periodicity of ~252 nm.
which is very close to the target $P = 250$ nm. This is noteworthy because the previous techniques discussed in sections 5.2, 5.3.1 and 5.3.1.1 also led in arrays where the fabricated periodicities reflected the design parameter accurately.

Figure 62: (a) Test run using a 10x10 cross cavity aperture array bitmap. Current was set to 1.5 pA. Dwell time was set to 4 ms and relative interaction diameter was set %0 with serpentine. The actual arm-length=154.9 nm & 134.7 nm with periodicity 252.3 nm, (b) top view.

With the promising results obtained in the test run above, a 50×50 symmetric cross-shaped aperture array with target arm-length $L = 150$ nm, was milled with slightly lower dwell times of 3.5 and 3 ms to further refine the fabrication quality, see Figure 63. Fabricated array periodicities measured $P = 248$ nm in both cases and close to the target $P = 250$ nm. Arm-lengths measured $L_x = \{150, 143\}$nm and $L_y = \{120, 133\}$nm for dwell times $t = \{3.5, 3\}$ms respectively, i.e. an unexpected asymmetry, see Figure 63(a)-(b). Dispersed spectrum, Figure 63(c), for both arrays were obtained using the same technique described in section 5.2. Top view images taken at 0° tilt angle, Figure 63(d), revealed that the fabricated crosses are asymmetric and anisotropic in a peculiar way. One half-arm is shorter, narrower and shallower than the other half-arm along the same arm-length, e.g. in the $x$ direction. When milling a feature based on a single line segment, the starting pixel on the target surface receives more charge per unit area in comparison to the subsequent pixels. The excess charges are due to the diffracted secondary ions from the neighbouring pixels. This delivers more charge per unit area at the starting pixel giving it a deeper and wider profile in comparison to the end pixel. Fabrication defect due to secondary ions, however, may not be eliminated. In this exercise, a single pass in conjunction with the serpentine milling pattern was used, where each row is milled in the opposite direction to the previous one. This results in defects that are aligned in opposite directions in alternating rows, see Figure 63(d)-(e).
Figure 63: 50×50 cross-shaped aperture arrays, with target arm-lengths \( L = 150 \) nm, milled with dwell times of (a) 3.5 ms and (b) 3 ms. (c) Dispersed spectra associated with (a) and (b), (d-e) top view with tilt angles 0° and 52° respectively. Serpentine milling pattern introduces defects in the form of asymmetry in the milled crosses. The asymmetry changes orientation in alternating rows.

Above process was repeated with bitmap patterns with a target arm-length \( L = 130 \) nm. With 3 ms dwell time, the fabricated arm-lengths measured 126×119 nm, whereas the 3.5 ms dwell time produces crosses with arm-lengths 122×121 nm, see Figure 64.
**Figure 64**: $50 \times 50$ cross-shaped aperture arrays with target arm-length $L = 130$ nm milled with dwell times of (a) 3.5 ms and (b) 3 ms. (c)-(d) Dispersed spectra associated with (a) and (b) respectively.

SPE files were post-processed and are depicted in Figure 65-Figure 68. In each figure, (a) shows the post-processed normalized dispersed spectrum $(I-I_{\text{min}})/(I_{\text{max}}-I_{\text{min}})$ with red = 1 and blue = 0. Here, $I$, $I_{\text{min}}$ and $I_{\text{max}}$ are the intensity, the minimum local intensity (or the background) and the maximum local intensity respectively. (b) Depicts critical lines where maximum intensities are observed within the array. A single spectrum produced by averaging the intensities over the length of the array (labelled as “Lines $y_{\text{start}}$-$y_{\text{end}}$”), is included as before. (c) Represents the averaged intensity distribution over the length of the array. SPE files revealed a more evenly distributed spectral lines in comparison to those fabricated with EBL, compare Figure 65-Figure 68 to Figure 49-Figure 54. Unlike the double peaks observed in section 5.2, spectra in this exercise exhibit a smoother line with a single peak at $\lambda \approx 750$ nm and $\lambda \approx 700$ nm for $L = 150$ nm and $L = 130$ nm respectively. The consolidated plot of the transmission spectra is depicted in Figure 69 (read the caption). This is an indication that milled crosses within a single array are more homogenous in shapes and sizes in comparison to those fabricated with EBL. Relevant post-process SPE files are shown in Figure 67 and.
Figure 65: $L = 150$ nm DT = 3.5 ms

Figure 66: $L = 150$ nm DT = 3 ms

Figure 67: $L = 130$ nm DT = 3.5 ms

Figure 68: $L = 130$ nm DT = 3 ms

Figure 69: Transmitted intensities averaged over the length of an array normalized to the maximum pixel value vs. the wavelength for the 50×50 cross-shaped aperture arrays fabricated with FIB using bitmap patterns based on line segments. Target arm-lengths are $L = 130$ nm and $L = 150$ nm. Averaged over all spectral lines (i.e. over the length of an array)
5.4. Conclusion

Although EBL is more economical in mass production of large arrays, shapes and the dimensions of the fabricated apertures deviates from the design when EBL is used as a preferred fabrication technique. FIB on the other hand, was proven to produce subwavelength apertures with dimensions that are more aligned with the design. Note, the accuracy and precision of fabricated features depend partially on the skill and proficiency of an individual with each technique. Another disadvantage of EBL, in comparison to FIB, is the cost associated with extra steps involved in the process, namely the preparation of GDS and GPF files, spin coating the resist, developing, etching, separate SEM session for measurements and sometimes removal of the left over resist by means of chemical etching. FIB (besides having none of the overheads associated with EBL) has a built-in SEM that facilitates a trial and error approach during the fabrication. Therefore, until a more efficient EBL techniques and recipes are investigated/discovered, one must resort to FIB when fabricating arrays cross-shape apertures. One remarkable outcome of these exercises was the observed accuracy in fabricated periodicities that was common among various techniques deployed. Plasmonic effects based on periodic surfaces, although entailing a more complex light-matter interaction, are versatile. A seemingly simple surface wave may be responsible for a variety of optical effects. SPP Bloch waves are explored further in the next chapter.
6. Cylindrical Hole Arrays

This chapter concerns with the theoretical studies of SPP Bloch waves in arrays of cylindrical holes. Most analytical solutions are simplified, approximate and fitted, thus leading to wrong design parameters. A rigorous analytical solution that can accurately predict the desired dimensions associated with hole arrays is complex to derive. Finite element analysis (FEA), on the other hand, offers extensive capabilities for modelling and simulating the response of periodic structures such as that of an infinite array of holes. In this chapter, a square array of cylindrical holes is modelled numerically. A simple analytical model is presented (not for design purposes) only to identify the origin of the alleged (1,1) and (0,2) modes and the results are compared to the numerical ones. Changes in the transmission spectrum vs the refractive index of the surrounding dielectric when illuminated with a linearly polarized light at normal incidence is studied. Circumstances in which an obliquely incident light changes to the SPP momentum are also identified.

6.1. SPP Bloch Modes and Wood Anomalies

Various attempts have been made to explain the resonance shift and the Fano profile associated with the extraordinary optical transmission (EOT) through subwavelength hole arrays in terms of the coupling of SPP Bloch waves to the non-resonant scattering process from the holes [170-173]. The role of resonant and non-resonant Wood anomalies in shaping the spectral line was also investigated [174]. Recent investigations on the origin of EOT in metallic hole arrays suggest that the contributions of SPPs are limited to only 50% of the total EOT, with quasi-cylindrical waves (QCW) being responsible for the other 50% [175-177]. The optical transmission associated with hole arrays in metallic films can be described in terms of resonant excitation of SPP modes responsible for enhanced transmissions and Wood’s anomalies [135] that are responsible for supressed transmissions. An array of cylindrical holes was modelled using a 100 nm silver film set in the x-y plane, sandwiched between two semi-infinite dielectric slabs with refractive indices $n_1$ and $n_2$. The dielectric filling the holes has an
index of $n_3$. The structure was normally illuminated with a plane wave propagating in the $+z$ direction, polarized at $0^\circ$ to the $x$ axis at $\lambda_0 = 700$ nm. To prevent transmission through the film and propagating modes through the holes at $\lambda_0 = 700$ nm, the thickness of the silver film was set to $h = 100$ nm and the diameter of the holes to $d = 200$ nm [178]. Note that the skin depth for silver is $\delta \approx 25$ nm in the visible regime. The relative permittivity of bulk silver at the design wavelength of $\lambda_0 = 700$ nm was calculated from tabulated experimental data to be $\varepsilon_{Ag}(\lambda = 700nm) = \varepsilon_{Ag}^0 - i\varepsilon_{Ag}^\prime = -20.43279 - 1.2862i$ [179]. Various techniques, such as perfectly matched layers, scattering boundary conditions and ports were employed to eliminate the back reflection of the diffracted wave from the upper and the lower boundaries of the model. Terminating the upper and the lower boundaries with ports was found to be the most appropriate approach for square arrays. In this section, the silver film was assumed to be supported by a glass with a refractive index of 1.52. The simulated absolute transmission (normalized to the incident intensity over a unit cell) as a function of the periodicity at $\lambda_0 = 700$ nm for $n_1 = 1.52$ and $n_2 = n_3 = 1$, (i.e. silver hole array on a glass substrate), is shown in Figure 70(a). A maximum is observed at the periodicity of $P = 394$ nm. The absolute transmission vs. the wavelength for numerically obtained periodicity $P = 394$ nm is depicted in Figure 70(b). The numerical solution produced the modes at $(1,0)_{W-glass} = 650$ nm, $(1,1)_{W-glass} = 500$ nm and $(1,0)_{SPP-glass} = 700$ nm. Although the peaks at 540 nm and 475 nm are being attributed to the $(1,1)_{SPP-glass}$ and $(2,0)_{SPP-glass}$ respectively, it can be shown that this is not the case.

![Figure 70: Absolute transmission spectra (normalized to the incident intensity over a unit cell) through a square array of holes as a function of (a) the periodicity at $\lambda_0 = 700$ nm and (b) the wavelength for $P = 394$ nm.](image-url)
SPP Bloch modes is already covered in section 2.5. In a square array the condition for Wood's anomalies is given by [135]:

\[ P = \frac{\lambda_W}{\sqrt{\varepsilon_d}} \sqrt{i^2 + j^2} \]  

(6.1)

where \( \varepsilon_d \) is the permittivity of the dielectric that interfaces the metal and within which the Wood's anomalies manifest and \( \lambda_W \) is the free space wavelength associated with the Wood’s anomaly. Using equations (2.25) and (2.26), for \( i = 0 \) and \( j = 1 \), the periodicity that supports the fundamental SPP Bloch mode at \( \lambda_0 = 700 \text{ nm} \) is analytically calculated to be \( P = 2\pi/G = 433 \text{ nm} \). Using equations (2.25), (2.26) and (6.1), analytical values for the free space wavelengths associated with the resonant SPP Bloch modes and Wood’s anomalies that fall within the optical spectrum for \( P = 433 \text{ nm} \) were calculated to be:

\[
\begin{align*}
(1,0)_{\text{SPP-glass}} &= 700 \text{ nm}, \\
(1,1)_{\text{SPP-glass}} &= 530 \text{ nm}, \\
(2,0)_{\text{SPP-glass}} &= 433 \text{ nm}, \\
(1,0)_{\text{W-glass}} &= 658 \text{ nm}, \\
(1,1)_{\text{W-glass}} &= 465 \text{ nm}.
\end{align*}
\]

Although the simulated \( (1,1)_{\text{SPP-glass}} \) mode seems to agree with the corresponding analytical value, large difference observed between the simulated \( (2,0)_{\text{SPP-glass}} \) mode and that obtained analytically, raises suspicions that other phenomena may be involved in shaping the spectral lines.

Table 2: Summary of the results obtained from simulations vs. those obtained from equations (2.25), (2.26) and (6.1)

<table>
<thead>
<tr>
<th></th>
<th>( P ) (nm)</th>
<th>( (1,0)_{\text{SPP-glass}} )</th>
<th>( (1,1)_{\text{SPP-glass}} )</th>
<th>( (2,0)_{\text{SPP-glass}} )</th>
<th>( (1,0)_{\text{W-glass}} )</th>
<th>( (1,1)_{\text{W-glass}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Simulations</td>
<td>394</td>
<td>700</td>
<td>540</td>
<td>475</td>
<td>650</td>
<td>500</td>
</tr>
<tr>
<td>From Equations</td>
<td>433</td>
<td>700</td>
<td>530</td>
<td>433</td>
<td>658</td>
<td>465</td>
</tr>
</tbody>
</table>

de Abajo [172] modelled the hole array in real space based on the actual transmittance through the holes, where each hole is considered to be a combination of electric and magnetic dipoles. In doing so, he applies the Babinet’s principle to equations derived for array of circular disks. He then derive the dispersion relation governing lattice surface modes for various types of hole, e.g. circular and square. In a similar fashion, Garcia-Vidal et. al. [173] started with modelling the transmission through a single hole (circular and rectangular) in k-space leading to a relationship governing their
transmittance. He then extends the concept to arrays. SPPs, however, are highly confined to bulk metallic surfaces in general [180]. Here, I attempt to single out the contribution of SPPs in the transmission spectrum based on the interaction between two holes. It is assumed here that SPPs in a periodic setting are confined to the surface. This hypothesis was formed based on a numerical solution (result not shown here) revealing that the maximum transmission in the air corresponding to the (0,1) mode, coincided with standing SPP waves on the surface of the array. It was hypothesized that although it is valid to consider contributions of every hole to the overall transmission in the air, it would be incorrect to assume long range involvement over the surface. Although SPP fields extend from the rims of the hole to almost the centre of the hole, surface charges do not jump across a hole to the other side. Long range surface wave that graze the periodic surface are responsible for the wood anomaly and suppressed transmission at certain wavelengths [135, 174]. The nearest neighbour interaction, on the other hand, was hypothesized to be responsible for scattering of the SPPs into free electromagnetic waves. Inferring that, although both effects can coexist, only the latter must influence the EOT. But first consider Figure 71 that depicts the electric field components of the SPPs launched by an isolated hole with \( d = 200 \) nm perforated in a 100 nm thick silver film set on the x-y plane. \( E_x \) is an even function of \( x \) and \( y \). \( E_y \) is an even function of \( x = y \) and \( x = -y \). The normal to the surface component of the electric field, \( E_z \), that are responsible for shaping the surface plasmons is an odd function \( x \) which explains the vanishing intensity along the y-axis due to destructive interference, hence the \( \cos(\phi) \) dependency in equation (2.24)[135]. Note that one must distinguish between surface plasmons (SP) and surface plasmon polaritons (SPP). Strictly speaking, SPs are launched by the electric field normal to the metal surface (i.e. \( E_z \) in this case). Whereas, SPs and all the electromagnetic fields that couple to them (e.g. \( E_x \) and \( E_y \)) constitute the SPPs, hence the term polaritons.
Figure 71: Electric field components of the SPP calculated over the silver/glass surface (top row) and air/silver interface (bottom row) in the vicinity of an isolated hole 200 nm in diameter perforated in a 100 nm thick silver film. The aperture was illuminated with a normally incident $x$-polarized light from the air/silver interface. Note that $E_x$ for the air/silver interface (bottom-left) depicts the superposition of the incident field and the SPP field. (red = 1, blue = -1 and green = 0)

The model proposed in this section considers only the nearest neighbour lattice points as it represents the simplest possible interaction over the surface of the array. The model proposed here is related to the electric field components of the SPPs. Now, consider the interaction between two virtual holes in 1D (Figure 72). Interaction via the $z$-component of the SPP may be explained in terms of the superposition principle. Let us assume that hole (1) is position at $x = 0$ and hole (2) at $x = P$.

The superposition of the $z$-components of surface waves at $x = 0$ may be written as:

$$
\psi_z\big|_{x=0} = \frac{1}{3} \left[ \psi_1(\varphi_1 + 0) + \psi_1(\varphi_1 + 2k_{spp}P) + \psi_2(\varphi_2 - k_{spp}P) \right]
$$

(6.2)
where \( \psi(kx) = e^{ikx} \). The first term in the RHS corresponds to the wave launched by hole (1) prior to propagation along the surface. The second term represents the time lapsed wave launched by the hole (1) and reflected back by hole (2) arriving at \( x = 0 \), hence travelling a total distance of \( 2P \). The third term is the wave launched by hole2 in the opposite direction arriving at hole (1). The normalization factor 1/3 is set based on the assumption that all waves are launched with equal strength. The condition for resonance dictates that all three waves be in phase at \( x = 0 \). From the first two terms in the RHS it can be inferred that \( k_{SPP} \times P = \pi \). From the first and the third terms, it can be shown that \( \varphi_2 = \varphi_1 + \pi \). Setting \( \varphi_1 = 0 \) for convenience, the equation at \( x = 0 \) can be written as:

\[
\psi_x |_{x=0} = \frac{1}{3}[\psi_1(0) + \psi_1(2k_{SPP}P) + \psi_2(\pi - k_{SPP}P)]
\]  
(6.3)

The \( x \)-component of the SPP, \( \psi_x \), that are responsible for shaping the transmission spectrum, may now be derived. For \( \psi_x \) the phase difference between the two holes must be set to \( \varphi_2 = \varphi_1 = 0 \), simply due to the \( E_x \) (see Figure 7) being an even function of \( x \), leading to:

\[
\psi_x |_{x=0} = \frac{1}{3}[\psi_1(0) + \psi_1(2k_{SPP}P) + \psi_2(-k_{SPP}P)]
\]  
(6.4)

To derive the \( y \)-component of the SPP, \( \psi_y \), one must note that there are no interactions between the holes along the \( x \)-axis via the \( E_y \) since \( E_y = 0 \) along the \( x \)-axis (see Figure 7). When considering a square array of holes, however, interactions along the \( x = y \) line may be written as:

\[
\psi_y |_{x=0} = \frac{1}{3}[\psi_1(0) + \psi_1(2k_{SPP}\sqrt{2}P) + \psi_2(-k_{SPP}\sqrt{2}P)]
\]  
(6.5)

where, \( k_{SPP} \) is obtained from equation (2.21) for a given wavelength. With \( P = 433 \) nm, complex amplitudes \( \psi_x \) and \( \psi_y \) were calculated at the glass/silver interface over the range of wavelengths \( 450 \leq \lambda_0 \leq 850 \) nm using equations (6.4) and (6.5) respectively. Squares of the modulus, \( |\psi_{x,y}|^2 = |\psi_{x,y}|^2 \), amplitude squares of the real, \( |\text{Re}(\psi_x)|^2 \), and the imaginary parts, \( |\text{Im}(\psi_x)|^2 \), were also calculated, see Figure 73(a). Positions of the \((0,1)_{SPP\text{-glass}}\) and \((1,1)_{SPP\text{-glass}}\) modes were determined using the response function:
where \( k_{ij}^2 \) was determined from equation (2.26) for \( P = 433 \) nm. Here, \( \gamma = k_{i,j}/Q \) is the damping factor associated to the \((i,j)\) mode. The quality factor, \( Q \), was set to 5 in order to represent a lightly damped system \([181]\). Peak position observed at \( \lambda_0 = 525 \) nm in the \( |\psi_y|^2 \) spectrum is due to the diagonal distance \( \sqrt{2}P \) from the neighbouring holes in a square array and it agrees with the \( R_{(1,1)} \), Figure 73(b). The \( |\psi_x|^2 \) spectrum shows a maximum at \( \lambda = 700 \) nm corresponding to the \((0,1)_{\text{SPP-glass}}\) mode that is also confirmed by \( R_{(0,1)} \). Given that equation (6.4) is modelled based on a 1D interaction, it is not surprising that the \((1,1)_{\text{SPP-glass}}\) mode at \( \lambda_0 = 525 \) nm is absent in the \( |\psi_x|^2 \) spectrum. What is remarkable is the presence of other peaks at \( \lambda_0 = 540 \) nm and \( \lambda_0 = 475 \) nm. These are the exact locations of the alleged \((1,1)_{\text{SPP-glass}}\) and \((2,0)_{\text{SPP-glass}}\) modes observed in Figure 70(b). With \( |\psi_x|^2 \) spectrum being calculated using a 1D model that does not support the diagonal modes, neither of the two peaks may be attributed to \((1,1)_{\text{SPP-glass}}\). The origin of the peaks at \( \lambda_0 = 540 \) nm and \( \lambda_0 = 475 \) nm in \( |\psi_x|^2 \) spectrum is simply the constructive interference of the superposition of the three complex waves \( \psi_1(0), \psi_1(2k_{\text{SPP}}P) \) and \( \psi_2(-k_{\text{SPP}}P) \). The modulus of the combined \( x \) and the \( y \) components of the waves, \( |\psi_x + 0.15 \psi_y|^2 \), shown in Figure 73(c), bears resemblance to that depicted in Figure 70(b). Here, the amplitude of 0.15 is a fitted parameter and is attributed to the strength of the coupling between the \((1,1)_{\text{SPP-glass}}\) mode to the incident/transmitted field. Note that the model proposed here is not predictive but is a posteriori fitted. Contribution of the \((1,1)_{\text{SPP-glass}}\) mode to the overall transmission spectrum of a square array of holes was found to be insignificant. Nevertheless, throughout this thesis higher order peaks observed in spectra of a hole array are labelled as \((1,1), (0,2) \ldots \) etc. for convenience. The impact of the dielectric material filling the holes on the SPP Bloch modes is presented in the following section.
Figure 73: (a) Squares of the modulus, $|\psi_{x,y}|^2 = \psi_{x,y} \psi_{x,y}^*$, amplitude squares of the real, $|\text{Re}(\psi)|^2$, and the imaginary parts, $|\text{Im}(\psi)|^2$ vs. the wavelength. (b) Response functions for (0,1)$_{\text{SPP-glass}}$ and (1,1)$_{\text{SPP-glass}}$ modes. (c) Modulus of the combined $x$ and the $y$ components of the waves, $|\psi_x + 0.15 \psi_y|^2$.

6.2. Impact of Cavity’s Dielectric Constant on The SPP Bloch Modes

Although it seems that values obtained analytically are in agreements with the simulations, one cannot rely on the analytical solution for design purposes. Figure 74 depicts the absolute transmission spectra for the same array with various combinations of refractive indices of the substrate, superstrate and the hole. To segregate and identify
modes supported at glass/silver interface from those supported at air/silver interface, the transmission spectra vs. the wavelength for $n_1 = n_2 = n_3 = 1$ was simulated. The spectrum shows that SPP Bloch modes at the air/silver interface have a cut-off wavelength of $\lambda = 530$ nm, corresponding to the $(1,0)_{\text{SPP-air}}$, and is well away from the target wavelength $\lambda = 700$ nm. The $n_1 = n_2 = 1.52$ spectrum, with the index of the material filling the holes kept fixed at $n_3 = 1$, shows a redshift in the fundamental mode from $\lambda_0 = 700$ nm to $\lambda_0 = 730$ nm caused by the presence of the glass at both the incident and the exit interfaces. The $(1,1)_{\text{SPP-glass}}$ mode, on the other hand, maintained its original position at 530 nm. This implies that an extra momentum could not have been responsible for shifting the $(1,0)_{\text{SPP-glass}}$ resonance. Changes in the amplitude and the position of the mode, in this case, may be explained in terms of the coupling of surface modes to the Fabry-Pérot (FP) standing waves inside the holes. Resonant SPP Bloch waves as well as the FB resonance of the holes are influenced by the film thickness, array periodicity and the diameter of the holes, hence the Fabry-Pérot Evanescent Wave (FPEV) [182]. The offset between the two resonances influences both the peak wavelength and the enhancement/suppression of EOT. With $n_1 = n_3 = 1.52$ and the superstrate being the air, the $(1,0)_{\text{SPP-glass}}$ is redshifted to $\lambda = 760$ nm. For $n_1 = n_2 = n_3 = 1.52$, a further shift to $\lambda = 800$ nm is observed. In both cases with $n_3 = 1.52$, the $(1,1)_{\text{SPP-glass}}$ mode was also redshifted away from its original peak wavelength of 530 nm.

![Figure 74: Transmission (normalized to the incident intensity over a unit cell area) spectra of a square array of holes, 200 nm in diameter and periodicity of 394 nm, perforated in](image)
100 nm silver film, for various combinations of refractive indices of the substrate, superstrate and the hole.

6.3. Incident Light and SPP coupling

Whether subwavelength cylindrical holes support guided modes, is a matter of dispute. Ebbesen and co-workers are inclined towards a cut-off wavelength of $\lambda_0 = d/2$ for guided modes through cylindrical holes [178] hence attributed the origin of the extraordinary optical transmission to the coupling of the light and the SPPs on the incident surface [79] that initiate an evanescent tunnelling process through the holes. In a later publication [183], they changed their claim to include the exit surface as part of the tunnelling process without any justification. And yet this was done in the same article showing that the coupling between the incident light and the SPP Bloch waves at the silver/air interface has a different EOT dispersion curve in comparison to that of the silver/glass interface [183]. Indeed such claims have created confusion and misunderstandings among the plasmonic community that I intend to clarify here. I tend to agree with Catrysse et al. that proved subwavelength cylindrical holes do support guided modes [180, 184, 185]. For a hole array perforated in an optically thick metallic film the dispersion relation at the metal/air interface as a function of the angle of incidence, is traced along a separate band structure in comparison to that of the metal/glass interface [186]. As previously mentioned: the cut-off wavelength of $\lambda = 530$ nm, corresponding to the (1,0)$_{SPP_{air}}$ Bloch mode, is well away from the target wavelength $\lambda_0 = 700$ nm. This implies that if the array is illuminated by a normally incident light at $\lambda_0 = 700$ nm from the air/silver interface, none of the Bloch modes at the air/silver interface would be excited. Consequently, in the absence of SPPs on the incident surface, the so-called “evanescent tunnelling process through the subwavelength holes” claimed by Ebbesen and co-workers, is never initiated. Therefore, one must not expect the formation of SPP Bloch waves on the exit (glass/silver) surface either. However, my simulations showed the formation of SPPs at the glass/silver interface with no (or faint) SPPs at the air/silver interface when the array was illuminated from the air with a normally incident light at $\lambda_0 = 700$ nm. This is only possible if the holes have transmitted the incident light without the assistance of the (frail or non-existent) SPPs at the incident air/silver interface, Figure 75. This may
provide us with a solution for minimizing the depolarization effect in hole arrays which may result from the off-normal incidence.

![Figure 75: Electric field components of the SPPs calculated over the silver/glass surface (top row) and air/silver interface (bottom row) over the array of holes 200 nm in diameter perforated in a 100 nm thick silver film. Period of the array is $P = 433$ nm. The array was illuminated with a normally incident $x$-polarized light from the air/silver interface. Note that $E_x$ for the air/silver interface (bottom-left) depicts the superposition of the incident field and the SPP field. All images were produced on the same scale with (red = 1, blue = -1 and green = 0).](image)

It is well known that in the case of an off-normal incidence on metallic surfaces, the parallel to the surface component of the incident wavevector makes a contribution to the resultant SPP wavevector, see equation (2.25), which impacts the state of polarization at transmission, hence the term depolarization [187, 188]. Reiterating the fact that the period of the above array was designed based on the fundamental surface mode supported at the glass/silver interface, it is obvious that the parallel-to-the-surface component of the incident wavevector contribute to the momentum of $(i,j)_{SPP,\text{glass}}$ modes only if the light is incident on the glass/silver interface. Figure 76 depicts the absolute transmission (normalized to the intensity over a unit cell) calculated in the air half-space vs. the angle of incidence, $\theta$, when the array is illuminated from the glass half-space with a TE polarized light in the $x$-$z$ plane, i.e. $E_x k_\parallel = 0$ for all $\theta$. Any changes observed in the spectrum, therefore, are purely due to the contribution that $k_\parallel = k_\parallel \sin(\theta)$ makes to the SPP momentum. For $\theta = 10^\circ$, the resonance wavelength associated with the
(1,0)$_{\text{SPP-glass}}$ mode, is blue shifted from $\lambda = 700$ nm to $\lambda = 690$ nm, whereas the analytical solution, see equation (2.25), predicts the resonance shift to $\lambda = 640$ nm or $\lambda = 760$ nm for $(+1,0)_{\text{SPP-glass}}$ and $(-1,0)_{\text{SPP-glass}}$ respectively. A $\pm 10$ nm shift in resonance, instead of $\pm 60$ nm, cannot be explained by the equation (2.25). The coupling of the parallel-to-the-surface component of the incident “electric” field to SPP modes is influential in shaping the spectra of the hole array when excited with an oblique incident light. Figure 77 shows the calculated transmission through the array when it is illuminated from the substrate with a TM polarized light, i.e. $\mathbf{E} \cdot \mathbf{k}_\parallel > 0$, for $\theta > 0$. The redshift in $(-1,0)_{\text{SPP-glass}}$ resonance and blue-shift in the degenerate mode $(+1,0)_{\text{SPP-glass}}$ are now in accordance with equation (2.25). The amplitude of the degenerate mode, however, builds up gradually with increasing $\theta$. The only difference between the TE and TM mode, is the presence of the $z$-component of the incident electric field in the latter. So it seems that it is the $z$-component of the incident field that interacts with the hole, giving rise to $\mathbf{k}_\parallel$.

Clearly equation (2.25) has no provision for TE vs. the TM illuminations since it only addresses the SPP Bloch waves. To show the validity of my simulations Transmission spectra through a hole array vs. the incident angle reported by Ebbesen et.al[79] is also included for comparison. Although the array dimensions are different to those reported here, the trend in resonance shift vs. the incident angle are identical, compare Figure 77 to Figure 78.

Figure 76: Absolute transmission (normalized to the intensity over a unit cell) vs. the angle of incidence when the device is illuminated from the glass/silver side. TE mode, with $x$-$z$ being the plane of incidence.
Figure 77: Absolute transmission (normalized to the intensity over a unit cell) vs. the angle of incident when the device is illuminated from the glass/silver side. TM mode, with $x$-$z$ being the plane of incidence.

Figure 78: Transmission spectra through a hole array vs. the incident angle reported by Ebbesen et.al[79]

The other scenario where the off-normal light is incident on the air/silver interface, the $(i,j)_{\text{SPP-glass}}$ modes (that are excited only by the extraordinary transmitted light through the holes) are shielded from the incident light and desensitized to the angle of incidence. In such a scenario only the $(i,j)_{\text{SPP-air}}$ modes couple to the incident light and since the cut-off wavelength for $(i,j)_{\text{SPP-air}}$ is at $\lambda = 530$ nm, the $(1,0)_{\text{SPP-glass}}$ mode at $\lambda = 700$ nm remains uninfluenced, compare Figure 77 to Figure 80. In the case of the off-normal incidence in TE mode, $k_{\text{SPP}}$ and the parallel-to-the-surface component of the incident electric field are orthogonal, therefore there will be negligible changes to the SPP’s momentum, see Figure 76 and Figure 79.
Figure 79: Absolute transmission (normalized to the intensity over a unit cell) vs. the angle of incident when the device is illuminated from the air/silver side. TE mode, with $x$-$z$ being the plane of incidence.

Figure 80: Absolute transmission (normalized to the intensity over a unit cell) vs. the angle of incident when the device is illuminated from the air/silver side. TM mode with $x$-$z$ being the plane of incidence.

6.4. Conclusion

In section 6.1, the contribution of the actual $(1,1)_{SPP\text{-}glass}$ mode to the overall transmission spectrum of a square array of holes was found to be insignificant. According to the analytical mode, the origin of the conventionally labelled $(1,1)$, $(0,2)$ modes were attributed to the constructive interference of the complex superposed standing waves formed between the nearest neighbouring holes that did not include the diagonal modes.

Investigations in section 6.2 were extremely important as they indicate that equation

$$P = \frac{2\pi}{k_{SPP}} \sqrt{l^2 + j^2}$$

lacks generality since it does not include the hole.
From section 6.3 it is safe to say that for an off-normal incident light to have any impact on a particular SPP mode, two conditions, $E_k > 0$ and $E_{k_{SPP}} > 0$ must be satisfied. Understanding SPP-Bloch waves and Wood’s anomalies in aperture antennas is also important when designing antenna arrays which rely solely on the cavity mode. The SPP-LSP coupling may or may not be a desirable effect depending on the requirements. When designing plasmonic antenna arrays, however, one cannot rely on the analytical relations governing the surface modes.
7. Polarization Response of Bi-Periodic Hole Arrays

Can a simple bi-periodic array of cylindrical holes produce circularly polarized lights? Is it possible to produce a free-standing array of cylindrical hole to achieve a tuneable waveplate by changing the surrounding dielectric without refabricating arrays with different dimensions? What techniques have the potential in realizing such arrays? This chapter represents theoretical and experimental studies, answering the above questions.

There is increasing interest in harnessing plasmonic effects that are the manifestation of coherent electron oscillations on a metallic/dielectric interface. With the performance of electronic integrated circuits approaching their limit with respect to their size, optical circuits are promising alternatives as the next generation integrated circuits. However, miniaturization of optical circuits requires ultra-compact optical elements. Scaling down traditional optical elements is not possible due to the diffraction limit and plasmonic effects may offer a solution to this dilemma [189]. The ability to manipulate light with subwavelength optical components is highly desirable for a variety of applications [152]. For example, a controllable active display using light emitting diodes has been integrated with plasmonic polarizers consisting of an array of rectangular apertures [190]. High degrees of circular polarization using asymmetric cavities, such as arrays of asymmetric crosses [111, 112] or a single elliptical cavity in conjunction with elliptical periodic corrugations surrounding it, [113, 191], have been reported. Polarization conversion associated with arrays of L-shaped apertures in metallic films [192, 193] was previously explored as was a theoretical investigation of the polarization response of an array of stereo nano-holes [194]. A periodic array of elliptical apertures in a metal film and their impact on the polarization of the transmitted light has also been investigated [195, 196] along with a number of demonstrations of polarizing devices based on asymmetric nanoscale metallic particles [110, 197-199]. An investigation of the near-field optical phase in the feed gap of an asymmetric cross-shaped dipole antenna and the possibility of its application as a quarter waveplate is presented in [200]. The role of SPPs in polarization conversion from diffraction gratings was
analytically explored in [201]. The radiation pattern and, to a lesser extent, polarization effects of a rectangular array of holes, has been investigated in the context of shaping the scattered emission of a single dipole positioned at the centre cavity of the array [202]. Controlling the polarization state of a Single Photon Source (SPS) provides a mechanism for defining the computational basis states [2, 5, 203]. A single photon source emits energy in the form of one quantized unit of light at a time. A more elaborated account on the photon statistics related to the bunched, coherent, anti-bunched and single photon states, in terms of the second order correlation function $g^2(0)$, can be found in [7, 204]. The driving force behind the development of SPSs and single photon detectors is mainly the quantum information science, including cryptography [205]. Quantum cryptography based on Bell’s theorem was first outlined in [206], where the polarized photons were proposed as a replacement for the $\frac{1}{2}$-spin particles. The quantum mechanics and the algorithms behind the cryptography are elaborated on in [6] and beyond the scope of this report. Suffice to say that the two requirements, i.e. individual quanta and the entangled states could be satisfied by the polarization states of a single photon. Furthermore, it was argued that the vertical or the horizontal polarization states can only be defined relative to the emitter’s and/or detector’s position and orientation, hence not suitable for real-life applications. Therefore, to form the computational basis states, the left-handed and the right-handed circular polarization are more suitable [2].

It is apparent that there are variety of plasmonic structures that can potentially produce a Circularly Polarized Light (CPL). The main question is whether one must rely on Localized Surface Plasmons (LSP) or Surface Plasmon Polaritons (SPP) to achieve this. Can SPPs be the fundamental mechanism behind a design to produce CPL? Do SPPs play any role in the polarisation response of arrays of resonant apertures that possess LSPs? Which approach is more feasible?

### 7.1. Surface Plasmon Wave Plates

It has already been shown that a biaxial nanohole array possesses two distinct orthogonal SPP Bloch modes [207]. SPPs propagate away from a single hole as a cylindrical wave, see section 2.4. For a given $\mathbf{k}_{\text{SPP}}$, the amplitude of the phase varies
with the distance from the centre of the hole, which suggests the possibility of positioning scattering features, such as holes, at a distance from the source so that the SPP waves arrive at the surface features with a desired phase that depends on $k_{SPP}$ and the propagation distance. An azimuthal dependence of the SPP field is a result of the variation in the amplitude of the fields in the holes when excited by linearly polarized light. The strength of the SPP is maximized along the $k_{SPP}$ direction and this direction is established by the component of the incident electric field parallel to the surface of the metal, and hence the polarization angle of the assumed normally incident light. However, in the case of periodic structures, such as hole arrays, standing waves arise in the steady state which are called SPP Bloch waves that oscillate with a relative phase, $\Phi_{SPP}$, to that of the incident field. It is possible to detune the periodicity of the square lattice into a rectangular lattice with $P_x$ and $P_y$ such that $\Phi_{SPP,x} - \Phi_{SPP,y} = \pi/2$, where $\Phi_{SPP,x,y}$ represent the relative phases of the two orthogonal SPP Bloch waves. Drezet et al.\cite{191} proposed a BE structure with periodic elliptical corrugations such that $a_n = b_n + \delta L$, where $\delta L$ is the length difference between the long axis, $a_n$, and the short axis, $b_n$, of the $n^{th}$ concentric ellipse. It was then concluded that for SPP Bloch waves to satisfy $\Phi_{SPP,x} - \Phi_{SPP,y} = \pi/2$, the length difference must simply satisfy $\delta L \times k_{SPP} = \pi/2$.

The analytical value (using equation (2.25)) for the period of a square array of holes that supports SPPs at its glass/silver interface at $\lambda_0 = 700$ nm was found to be $P = \lambda_{SPP} = 433$ nm. Applying Drezet’s suggestion to the square array, the detuning in each direction is given by $\Delta P = \pm(\pi/4)/k_{SPP} = \pm54$ nm. When dealing with any periodic settings, however, one cannot ignore the reflection (hence the standing wave) by the surface features. The square of the amplitude and the relative phase vs. the $P$ was analytically calculated using equations (6.3) for $P_{SPP} = 433$ nm, corresponding to the $(1,0)_{SPP\text{-glass}}$ mode at $\lambda_0 = 700$ nm and are depicted in Figure 81.
Figure 81: The square of the amplitude and the relative phase vs. the \( P \) was analytically calculated for \( k_{\text{SPP}} = 2\pi/433 \text{ (nm}^{-1}) \) corresponding to \( \lambda_0 = 700 \text{ nm} \).

To achieve a phase difference of 90°, two orthogonal lattice constants symmetric about \( P = 433 \text{ nm} \) were chosen such that \( \Phi_{\text{SPP},x} - \Phi_{\text{SPP},y} = \pi/2 \). The detuning, \( \Delta P = \pm 21 \text{ nm} \) results in two orthogonal periodicities \( P_x = 412 \text{ nm} \) and \( P_y = 454 \text{ nm} \). Although the calculated periodicities guarantee a 90° phase difference between the two orthogonal modes, to produce a Circularly Polarized Light (CPL), they must also be equal in amplitudes. The amplitude of each mode can be controlled via their coupling to the parallel-to-the-surface component of the incident electric field. There is an optimum incident polarization angle were the two modes are equal in amplitude, that is \( H^{(1)}_{x}(k_{\text{SPP}} P_x) \cos(\alpha) - H^{(1)}_{y}(k_{\text{SPP}} P_y) \sin(\alpha) = 0 \), see equation (2.24). The optimum incident polarization angle was calculated to be \( \alpha = 46.5^\circ \) from the x-axis of the array. Figure 82 shows the schematic of such a bi-periodic array,
Figure 82: Schematic of a bi-periodic array of cylindrical holes producing transmitted CPL when illuminated with a linearly polarized light.

The analytical solution above does not take into account details such as the influence of the hole geometry, film thickness and the interaction between the incident field and the surface modes that give rise to the Fano line shape of the transmitted spectrum, compare Figure 81(ii) to Figure 70(a). Equation (2.25), therefore, cannot precisely predict the transmission maxima and minima associated with metallic hole arrays. For these reasons, to fine-tune the device, Maxwell equations were numerically solved using the Finite Element Method (FEM), implemented in COMSOL Multiphysics 4.3b. Specifically the interaction between a normally incident, linearly polarized plane wave with an infinite array of cylindrical holes in a thin silver film was modelled. The upper boundary on the exit side was terminated with a perfectly matched layer (PML) to eliminate back reflection of the diffracted wave. For a design wavelength of $\lambda_0 = 700$ nm under the normal incidence, the numerical solution in section 6, identified the periodicity $P = 394$ nm to correspond to the fundamental SPP Bloch mode $(1,0)_{\text{SPP-glass}}$. 
Figure 83: (a) the relative phase differences between the $x$ and $y$ components of the transmitted electric field and (b) Absolute transmission (normalized to the intensity over a unit cell) as a function of $P_y$ for $P_x = 394$ nm.

To identify the periodicities of the rectangular hole array capable of producing transmitted CPL at $\lambda_0 = 700$ nm, a parametric sweep over $P_y$, while setting $P_x = 394$ nm, determines the relative phase differences between the $x$ and the $y$ components of the transmitted electric field, see Figure 83. To achieve a phase difference of $90^\circ$, two orthogonal lattice constants symmetric about $P = 394$ nm were chosen. The nominated periodicities giving a net $90^\circ$ phase difference are $P_x \approx 368$ nm and $P_y \approx 407$ nm. Note that the total detuning $P_x - P_y = 39$ nm obtained from the simulation is close to the analytical value $2\Delta P = 42$ nm. Now that the phase requirements are met, to produce CPL, rather than elliptically polarized light, the $x$ and the $y$ components of the transmitted electric field must have equal amplitudes. This can be achieved by adjusting the SPP-incident field coupling which controls the amplitudes of the SPP Bloch waves. The coupling strength may be controlled by varying the incident polarization. Transmission through a rectangular array of holes with $P_x = 368$ nm and $P_y = 407$ nm was numerically modelled when illuminated with a normally incident light from the air/silver side at various polarization angles. The optimum polarization for the incident light at $\lambda_0 = 700$ nm, was found to be $\alpha = 47^\circ$ from the $x$-axis (and in agreement with the analytical value) resulting in $S_3 = 1$, which is the Stokes parameter for degree of circular polarization, see Figure 84(a). Equations governing the Stokes parameters are listed below:

$$S_0 = |E_{tx}|^2 + |E_{ty}|^2$$
$$S_3 = \frac{|E_{tx}|^2 - |E_{ty}|^2}{S_0}$$

(7.1)
Here, $E_{tx}$ and $E_{ty}$ are the transmitted $x$ and $y$ components of the electric field respectively. Transmission spectra, when the rectangular array was illuminated with incident polarizations $\alpha = \{0^\circ, 47^\circ \text{ and } 90^\circ\}$, are depicted in Figure 84(b). In all cases, the absolute transmission $P_t/P_0$ was calculated, where $P_t$ and $P_0$ are the transmitted power through the device and through the glass substrate in the absence of the device, respectively. $P_t$ and $P_0$ were calculated by integrating the $z$-component of the transmitted Poynting vector over the area covered by one unit cell within the array. Note that this definition of “normalized transmission” is synonymous with “absolute transmission”, hence the transmission less than the unity. Transmission spectra for the $0^\circ$ and $90^\circ$ incident polarizations show two maxima at $\lambda_x = 665$ nm and $\lambda_y = 720$ nm associated with the two orthogonal periodicities $P_x$ and $P_y$ respectively. These values are very close to those obtained analytically, $\lambda_x = 670$ nm and $\lambda_y = 728$ nm, see Figure 81(b). Note that the curves intercept each other at $\lambda_0 = 700$ nm, confirming the equal amplitude requirement at the design wavelength. At $47^\circ$ incident polarization, transmitted Stokes parameters were calculated, confirming transmission of CPL at $\lambda_0 = 700$ nm, (Figure 84(c)). Note further that at $\lambda_0 \approx 663$ nm, the transmitted light is linearly polarized in the $x$ direction, i.e. $S_1 = 1$. The origin of such dichroic behaviour lies in the resonant transmission in the $x$ direction coinciding with the transmission suppression in the $y$ direction at $\lambda_0 \approx 663$ nm. At this wavelength, in the case of the purely $y$-polarized incident light, the total transmission drops to 0. This is confirmed by the spectrum produced at $90^\circ$ incident polarization. The other interesting effect is the similarities in the spectra for scattering parameters in the range of $\lambda = [660\rightarrow 730]$ nm corresponding to $(1,0)_{\text{SPP-glass}}/(0,1)_{\text{SPP-glass}}$ to those in $\lambda = [450\rightarrow 490]$ nm corresponding to $(2,0)_{\text{SPP-glass}}/(0,2)_{\text{SPP-glass}}$ modes. Achieving the same effects at shorter wavelength without reducing the dimensions of the array is a highly desirable effect.
Figure 84: Spectra of the hole array having periodicities $P_x = 368$ and $P_y = 407$ nm. (a) Stokes parameters vs. the incident polarization. (b) Absolute transmission vs. the wavelength, when incident field is at polarizations $0^\circ$, $90^\circ$ and $47^\circ$. (c) Stokes parameters vs. the wavelength when incident polarization is $47^\circ$.

A top view of the simulated surface charge density on the silver/glass interface, when the device was normally illuminated from the air side with $\alpha = 47^\circ$ at $\lambda_0 = 700$ nm, is depicted in Figure 85. The surface charge density and the transmitted electric field vector (represented by the red arrow) 70 nm from the glass/silver interface, were produced at $t = \{0, T/8, T/4, 3T/8\}$, where $T$, is the period of the optical wave. The expected rotation in the surface charge density and electric field can be seen. The results shown above are based on optimizing the performance at a design wavelength of $\lambda_0 = 700$ nm. Simulations calculating the Stokes parameters as a function of wavelength with varying incident polarization angle, $\alpha$, provide a clearer picture of the device performance.
Figure 85: Surface charge density (red = 1, blue = -1 and green = 0) and the transmitted electric field vector (70 nm from the glass/silver interface, represented by the red arrow), were calculated at $t = \{0, T/8, T/4, 3T/8\}$, where $T$ is the period.

Figure 86(a) shows that a high degree of transmitted CPL, i.e. $S_3 \approx 1$, is achievable for $40^\circ \leq \alpha \leq 80^\circ$, however the wavelength associated with the $S_3 \approx 1$, experiences a blue shift from 704 nm to 676 nm respectively. The $S_1$ remains close to 1 at $\lambda_0 = 663$ nm for $\alpha \leq 70^\circ$.

Figure 86: Simulated (a) transmitted $S_3$ and (b) transmitted $S_1$ parameters vs. the wavelength for incident polarizations $10^\circ \leq \alpha \leq 80^\circ$ range.

7.1.1. Fabrication, Results and Discussion

A 2 nm thick germanium film (as an adhesion layer) was first deposited on a glass substrate followed by a 100 nm thick silver film and a 10 nm SiO$_2$ protective layer, using an IntlVac Nanochrome II electron beam evaporator. A rectangular array of holes with design periodicities of $P_x = 368$ nm and $P_y = 407$ nm was milled using a Helios
NanoLab 600 Focused Ion Beam (FIB). Fabricated periodicities (averaged over 20 periods) measured $\bar{P}_x \pm \Delta P_x = 366 \pm 13 \, \text{nm}$ and $\bar{P}_y \pm \Delta P_y = 417 \pm 16 \, \text{nm}$, Figure 87(a), with the array being slightly skew ($< 1^\circ$). The average hole diameter measures $d \approx 180 \pm 11 \, \text{nm}$ with some irregularities in geometry, see Figure 87(b). Visually inspecting the cross-sectional SEM image, layers of SiO2, silver and silver oxide were identified. The silver and the silver oxide layers varied in thickness across the film and measured approximately 80 nm and 20 nm respectively, Figure 87(c). Note that deposition of Platinum was part of the process of taking the cross-sectional images away from the array.

Figure 87: (a) Top view SEM image of the milled device. (b) Close-up SEM image showing the hole apertures. (c) Cross-sectional SEM image from the film.

To measure the transmission coefficients, the device was illuminated (at normal incidence) from the air/silver side with an incandescent (halogen) light using a Nikon inverted microscope (Eclipse Ti-U) and a Nikon 40x NA = 0.6 objective in bright field mode. The incandescent light was passed through a ThorLabs LPVIS050-MP (550-1500 nm) linear polarizer and a Nikon C-C Achromatic condenser operating in collimator mode prior to incidence on the air/silver interface. Transmitted intensities were analysed by an Andor Shamrock 303i-A spectrometer with Andor iDus DU920P-BR-DD CCD
operating in continuous mode, controlled by Andor Solis 4.21 software. The transmitted intensities (normalized to those through the substrate over the same area occupied by the array) were measured with the incident polarization varying between 0° to 90° (where 0° was aligned with $P_x$). To measure the Stokes parameters, the techniques described in [208] were employed where the transmitted light was passed through a circular polarizer constructed using a linear polarizer (ThorLabs LPVIS050-MP) and a quarter waveplate (ThorLabs AQWP05M-600).

Figure 88: Experimental data: (a) Transmitted $S_3$ spectra for incident polarizations $5^\circ \leq \alpha \leq 85^\circ$. (b) Absolute transmission when incident field is at polarizations 90°, 43° and 0°. (c) Stokes parameters vs. the wavelength for incident polarization 43°. (d) Stokes parameters vs. the wavelength for un-polarized normally incident light. Simulated data: (e)-(f) Simulated $S_3$ and $S_1$ parameters. (Red lines) simulated results performed for the fabricated geometries. (Black lines) experimental results as in (c). Dashed lines show calculations of devices assuming the extreme range of geometric uncertainties.
Experimental results are shown in Figure 88(a)-(d). The maximum value of $S_3$ was found to occur for an incident polarization of $43^\circ$ at $\lambda_0 \approx 739$ nm, Figure 88(a)-(b). Here $S_3 = \{0.86$ and $0.83\}$ indicates a high degree of circularly polarized light transmitted at $\lambda_0 = \{739$ and $570\}$ nm, see Figure 88(c). Stokes parameters and degree of polarization (DOP) in Figure 88(d) were obtained when the device was illuminated with an unpolarised light at normal incidence. A high degree of linearly polarized light, i.e. $S_1 \approx 0.75$, is transmitted in the range $650 < \lambda_0 < 700$ nm, due to the overlap of resonant SPP Bloch waves and Wood’s anomalies, (responsible for suppressed transmission), mentioned above.

The discrepancies between the predicted and experimental results are attributed to fabrication artefacts and the use of refractive index data for bulk silver in modelling nanoscale features\[209\]. To demonstrate the impact of fabrication artefacts on the $S_1$ and $S_3$ parameters, simulations were performed at an incident polarization angle of $43^\circ$ for the fabricated geometries, although assuming a perfectly rectangular array for simplicity. Also the 20 nm Ag$_2$O and the 10 nm SiO$_2$ were incorporated into the model. The refractive index for the Ag$_2$O layer was set to $n = 2.4$ \[210, 211\]. In Figure 88(e)-(f), the dotted lines show the results of simulations performed at $(\vec{P}_x - \Delta P_x, \vec{P}_y - \Delta P_y)$ and $(\vec{P}_x + \Delta P_x, \vec{P}_y + \Delta P_y)$ giving an indication of the variability in performance associated with fabrication errors. It is apparent that fabrication uncertainties play a key role in the performance of devices.

### 7.2. Polarization Dependent Refractive Index Based Sensor with Tuneable Quarter Wave Plate and Colour Filter Functionalities

Some plasmonic devices such as quarter wave-plates \[111, 112\] and frequency dependent optical beam steers \[212\] rely on two carefully spaced orthogonal modes to achieve their primary objectives. Arrays of cylindrical holes are relatively simpler to fabricate in comparison to those that rely on shape resonances. Previously, the wave-plate functionality of a bi-periodic array of cylindrical holes was demonstrated at a design wavelength \[213\]. It is highly desirable, however, to make such devices
tuneable without fabricating numerous arrays with different dimensions that operate at other wavelengths. Refractive index based plasmonic sensors rely on the shift in surface plasmon resonance (SPR) in response to a change in the surrounding dielectric environment making them ideal for the next generation of miniaturized sensing devices [207, 214-216] and spectroscopy [217-219]. SPRs are also the fundamental reason behind other effects such as the color filter functionality [220]. Considering optical filters and waveplates, devices that operate in EOT mode are analogous to their classical counterparts where the properties of an incandescent light passing through an optical element are altered on transmission, hence more suitable for miniaturization. In accordance with Krishnan et al. [221], numerical analysis confirmed that the highest resonant transmission through an array of holes perforated in metallic screens occurs in a symmetric setting where the dielectric constant of the substrate, the superstrate and the hole are the same. Changes in the refractive index of the environment also produced the largest shift in resonance in comparison to other scenarios where the substrate differed from the superstrate. Here, I investigate the possibility of the shift in resonances observed in refractive index based sensors being applied to a bi-periodic array of holes surrounded by a homogenous dielectric environment. The proposed device operates in EOT mode, hence achieving a miniaturized tunable quarter waveplate by means of controlling the polarization of the incident light as well as the dielectric medium surrounding the array homogeneously.

Soon after completing the measurements for the surface plasmon wave plates [213], additional measurements was performed on the sample before further oxidization consumes the silver film. The aim was to examine the $S_3$ parameter and the transmittance through the device for $n = n_1 = n_2 = n_3$ scenarios. A report on etching techniques by Hydrofluoric (HF) acid, suggested that HF penetrates through the holes and etches the substrate just under the array while leaving the silver film intact [222]. HF also removes other oxide layers, such as Ag$_2$O, formed over the film during the four months period that the sample was dormant. The sample was etched in a HF solution. The array was then free standing, attached to the substrate only through the rest of the silver film, Figure 89(a)-(b). This represents a scenario for $n_1 = n_2 = n_3 = 1$. Once the measurements were concluded for $n = 1$, a single drop of optical immersion oil with a refractive index matching that of the glass was place over the device and left overnight.
to allow the oil to penetrate the holes, filling the space under the array as well as the cavities. A microscope cover slip was placed on top of the device to flatten the convexed excess oil. Measurements were repeated for \( n_1 = n_2 = n_3 = 1.52 \) using the same procedure. Taking cross-sectional images is a destructive method, naturally, this could only be carried out after the measurements with immersion oil had concluded. Attempts to remove the oil (using acetone, IPO and even detergent) were unsuccessful. Acetone, in particular, took some of the moisture away from the oil upon vaporization. This made the oil even less flowing but this was a blessing as it made the sample more appropriate for SEM imaging. A cross-sectional image was produced using a dual-beam, focused ion beam and scanning electron microscope (FIB-SEM), Figure 89(c)-(d), representing a free-standing array in immersion oil.

\[
\begin{align*}
S_3 &= \frac{I(0°, 45°) - I(0°, 135°)}{I(0°, 0°) + I(90°, 90°)\cos(\Delta)} + S_2 \sin(\Delta) \\
\Delta &= \sin^{-1} \left[ I(0°, 135°)_{ini} - I(0°, 45°)_{ini} \right].
\end{align*}
\]

where \( S_2 = [I(45°, 45°) - I(135°, 135°)] \) and \( \Delta = \sin^{-1} \left[ I(0°, 135°)_{ini} - I(0°, 45°)_{ini} \right]. \) Note that \( I(\beta_2, \alpha_2) \) is the light intensity emerging from the analyser towards the spectrometer with \( \alpha_2 \) and \( \beta_2 \) being angles that

Figure 89: Cross-sectional schematics of the array (a) before and (b) after etching with HF. (c) cross-sectional SEM image of the freestanding array in a homogenous dielectric environment (immersion oil). (d) Close-up of (c).

In both cases the device was characterized using a technique suggested by Kihara [223] that takes into account the phase difference error associated with optical elements using:
transmission axis of the analyser and the fast axis of the quarter waveplate of the microscope makes with the x-axis respectively. $I(0°,135°)_{ini}$ and $I(0°,45°)_{ini}$ intensities were measured with $\alpha = 45°$ in the absence of our array. Being an argument to an arcsine function, $I(\beta, \alpha)_{ini}$ must be normalized to the incident intensity.

7.2.1. Results and Discussion

The array was modelled with the fabricated dimensions, $P_x = 366$ nm, $P_y = 417$ nm, $h = 80$ nm and $d \approx 180$ nm, with incident polarization angles, $\alpha = \{0°, 90°\}$ for $1 \leq n \leq 1.52$. The refractive index data for the silver was taken from a recently published work by Barbar and Weaver [224], that proved to be more aligned with the experiment (compared to Palik). Redshifts in $(1,0)_{SPP}$ resonances vs. the change in the refractive index, see Figure 90, were calculated to be $\Delta \lambda_{0x}/\Delta n = 458$ nm and $\Delta \lambda_{0y}/\Delta n = 496$ nm per refractive index unit (RIU) for $P_x$ and $P_y$ respectively. The enhancement in $(1,0)_{SPP}$ resonances vs. the refractive index changes linearly for both periodicities and was calculated as $\Delta T/\Delta n = (T_1 - T_{1.52})/0.52 \approx 0.13$. Here $T_n$ is the transmitted power at the $(1,0)_{SPP}$ resonance for a given refractive index $n$.

![Figure 90: Simulated results for absolute transmission (obtained from the scattering parameter $S_{21}$) showing the shift in $(1,0)_{SPP}$ resonance vs. the change in the refractive index $n$ for (a) $P_x = 366$ nm (i.e. incident polarization angle $\alpha = 0°$) and for (b) $P_y = 417$ nm (i.e. incident polarization angle $\alpha = 90°$).](image)

The change in transmitted power vs $n$, can be explained in terms of standing waves formed along the holes that give rise to the Fabry-Pérot (FP) resonance at wavelengths corresponding to $(1,0)_{SPP}$ modes. Transmission through a hole array is influenced by the interaction between the FP resonance and the surface modes which in turn depends on
the film thickness, array periodicity and the diameter of holes, hence the Fabry-Pérot Evanescent Wave (FPEV) [225]. In the context of plasmonic hole arrays, this implies that the strength of the fundamental modes vs the refractive index is dictated by the offset between the $(1,0)_{\text{SPP}}$ wavelength and the wavelength of the FP resonance. The fundamental SPP mode at $\lambda_{(1,0)_{\text{glass}}} \approx 790 \text{ nm}$ for $n = 1.52$ with the highest transmission of 0.87, for example, is the result of coupling of the FP mode inside the cavity to the SPP mode on the film’s surface. A parametric sweep over the film thickness with $n = 1.52$ at $\lambda_0 = 790 \text{ nm}$ revealed that the first FP resonance occurs at thickness $h = 80 \text{ nm}$, Figure 91(a). Figure 91(b) confirms that most of the incident power is transmitted through the device with minimum reflection. Transmission less than unity, however, is attributed to the mismatch between the $\lambda_{(1,0)_{\text{glass}}}$ mode and the FP resonance.

**Figure 91:** (a) Absolute transmission vs. the film thickness $h$ for $\alpha = 90^\circ$ and $\lambda = 790 \text{ nm}$ (i.e. $\lambda_{(1,0)_{\text{glass}}}$ for Py). (b) Calculated $|E|^2$ for $h = 80 \text{ nm}$, $\alpha = 90^\circ$ and $\lambda = 790 \text{ nm}$.

The transmission and the polarization response of the array was also modelled for $n_1 = n_2 = n_3 = 1$ and 1.52 vs. the incident polarization angle in the range of $0^\circ \leq \alpha \leq 90^\circ$.

Figure 92(a)-(b) shows that the device can act as a tuneable polarization dependent colour filter, a similar effect to this was previously reported based on shape resonances of asymmetric nano-cross particles [226]. Figure 92(c)-(d) and shows that a high degree of CPL is transmitted for $40^\circ \leq \alpha \leq 60^\circ$ in both cases. The experimental data are depicted in Figure 93 and are in agreement with the simulations. Experimentally measured $S_3$ parameters revealed a high degree of CPL being transmitted in the range of $40^\circ \leq \alpha \leq 55^\circ$ for $n_1 = n_2 = n_3 = 1$ and $40^\circ \leq \alpha \leq 60^\circ$ for $n_1 = n_2 = n_3 = 1.5$. The difference in the predicted $(1,0)_{\text{SPP}}$ resonances in comparison to those obtained
experimentally may be attributed to various factors. The effect of temperature [182, 227-229] (rising during the measurements), surface roughness in the film [230-232], the use of refractive index data for bulk silver in modelling nanoscale features[209], fabrication artefacts discussed in section 7.1.1 and perhaps the presence of off-normal incident fields all impact the experimental results. In the case of $n = 1$, experimentally obtained transmissions of 0.38 (vs. the simulated peak transmissions of ~0.8) is due to the presence of the glass substrate beneath the air gap that results in internal reflections reducing the coupling between the incident light and the SPPs. The 100% transmission at $\lambda_{(0,1)} = 874$ nm observed for $n = 1.52$ is attributed to the perfect alignment between the FP resonance and the (0,1) mode. To the best of my knowledge, this is the first experimental demonstration of Fan’s predictions [180, 184, 185], that no matter how small the cylindrical hole, at some wavelength near-complete optical transmission occurs. This is significant given that it occurs for $d/\lambda_{(0,1)} = 0.2$. Experimentally measured $S_3$ parameters revealed a high degree of CPL being transmitted at $\lambda_{CPL}$. These are near-perfect states of CPL being transmitted.

Figure 92: Simulated results for incident polarizations $0^\circ \leq \alpha \leq 90^\circ$. Absolute transmission vs. the wavelength for (a) $n_1 = n_2 = n_3 = 1$ and (b) $n_1 = n_2 = n_3 = 1.52$. $S_3$ vs. the wavelength for incident polarizations $0^\circ \leq \alpha \leq 90^\circ$ for (c) $n_1 = n_2 = n_3 = 1$ and (d) $n_1 = n_2 = n_3 = 1.52$. 
Figure 93: Experimentally obtained results for incident polarization $0^\circ \leq \alpha \leq 90^\circ$. Absolute transmission vs. the wavelength for (a) $n_1 = n_2 = n_3 = 1$ and (b) $n_1 = n_2 = n_3 = 1.5$. $S_3$ vs. the wavelength for (c) $n_1 = n_2 = n_3 = 1$ and (d) $n_1 = n_2 = n_3 = 1.52$.

With such a simple planar design this device proves to be a viable solution for applications such as colour filters, sensors and spectroscopy [215, 217, 218, 220, 233-235] [214].

### 7.3. SPP-LSP Coupling

Cross-shaped apertures are classified as slot antennas, and as such, they possess a distinct LSPR, which is the function of the aperture’s shape, dimensions and constituent materials. Localized surface plasmons associated with shape resonances, however, are highly sensitive to the geometry and the dimension of the metallic nanostructures [10, 20], hence more prone to fabrication errors. Relative fabrication error is defined as $\Delta L/L$, where $L$ is the target dimension and $\Delta L$ is the intrinsic absolute error that depends on the fabrication technique, material, skill set and the health of the instrument. Although the smallest spot size achievable by an Electron Beam Lithography (EBL) or a Focused Ion Beam (FIB) can be as low as $\sim 5$ nm [145, 236], the fabrication error can
be tens of nanometers. Consider an array of cross-shaped apertures having arm-widths $W = 40$ nm and arm-lengths $L = 180$ nm, perforated in a 140 nm thick silver film. Such a structure has its resonance at $\lambda = 660$ nm \cite{111}. This is the same resonant wavelength corresponding to the (1,0) surface mode of a hole array with periodicity $P = 600$ nm with hole diameters $d = 300$ nm perforated in a 200 nm thick silver film \cite{237}. The latter results in a lower relative fabrication error of $\Delta L/600$ nm corresponding to the periodicity $P = 600$ nm vs. $\Delta L/180$ nm associated with the cross-shaped aperture with arm-lengths $L = 180$ nm. It is highly desirable to fabricate relatively larger nanostructures, i.e. periodic surfaces in this case, while maintaining the same operational wavelength.

By tailoring the aperture’s dimension and the periodicities of a rectangular array, it is possible for the aperture’s resonance to coincide with the centre wavelength of the two orthogonal SPP modes. The aim is to model a less-than-perfect scenario that includes a $\pm 10$ nm fabrication error associated with apertures and show that despite the aperture’s dimension being more prone to fabrication errors, it is still possible to produce a SPP induced Circularly Polarized Light (CPL) response. Our previous simulations \cite{238} showed that an isolated symmetric cross-shaped aperture with arm-lengths $170 \pm 5$ nm and arm-widths 40 nm perforated in a 100 nm silver film supported by a glass substrate has a peak in transmission at the target wavelength $\lambda_0 = 700$ nm. Here, an asymmetric array of “symmetric” crosses having arm-lengths $L = 165$ nm was modelled. Choosing the value of the lower bound for arm-lengths allows for parametric sweeps over periodicities without violating the periodic boundary condition. Arm-lengths are aligned with the $x$ and $y$ axes. The arm-widths $W = 40$ nm, film thickness $t = 100$ nm, and the target wavelength $\lambda_0 = 700$ nm were set as before. For a square array of such symmetric crosses, periodicity of $P = 325$ nm, maximizes the throughput. Optimum periodicities for $S_3 = 1$ at $\lambda_0 = 700$ nm were then found to be $P_x = 272$ nm and $P_y = 366$ nm, when illuminated with a normally incident plane wave at 45° polarization. Figure 94(a) depicts the simulated Stokes parameters for the device.
Figure 94: (a) Normalized Stokes parameters for a rectangular array of symmetric cross-apertures, 165 nm in arm-length and 40 nm in arm-width, perforated in a 100 nm thick silver film when illuminated by a normally incident plane wave at $\lambda = 700$ nm polarized at $45^\circ$. The periodicities of the array are $P_x = 272$ nm and $P_y = 366$ nm. (b) $S_3$ parameters for the rectangular array of symmetric cross-cavity apertures with various arm-lengths.

In order to study the impact of the $\pm 10$ nm fabrication error associated with apertures, hence the shift in LSPRs, the array was modelled with cross aperture having various arm-lengths ranging from 155 nm to 175 nm while retaining the same dimensions for the array. Figure 94(b) depicts the $S_3$ parameter vs. the incident polarization for each arm-length. It is apparent that despite the variation in arm-length, a high degree of CPL is possible, provided that the polarization of the incident light is adjusted accordingly. Yet a more remarkable implication is that the surface modes, (rather than the constituent resonant apertures), are dictating the polarization response of the array. Although LSPs are highly confined to the apertures, SPPs also exist in the vicinity of an apertures; see Figure 98(g) for discussions on SPPs in the vicinity of a cross-shaped aperture. One cannot avoid SPPs.

### 7.4. Conclusion

It has been shown that the periodicities of a rectangular array of holes perforated in metal can be optimized to produce two orthogonal SPP Bloch modes that propagate with a relative phase difference of $\pi/2$, resulting in transmitted light that is circularly polarized. It is also possible to exploit the resonant Wood’s anomalies in conjunction to resonant SPP Bloch modes to produce linearly polarized light when illuminated with an unpolarized light at normal incidence. The array that was originally supported on a glass substrate was successfully converted into a free-standing array that may act as a tuneable quarter waveplate, a tuneable colour filter and a refractive index based sensor.
with high EOT. The experimental results confirmed that the coupling between higher order SPP modes, and the fundamental mode is either insignificant or non-existence for the given incident intensity that the experiment was conducted. With its simple design due to reliance on SPPs rather than LSPs, fabrication of such devices is highly feasible. Devices such as that presented here may play a role in future optical communication and sensing systems as well as in novel displays.
8. Bullseye (Single Aperture Surrounded by Corrugations)

In this chapter, certain claims regarding the design criteria and characterizations of plasmonic bullseye (BE) structures are investigated. Does the matching of the grating’s periodicity to the wavelength of surface waves results in a strong collimated beam along the optical axis of the device? What are the requirements for such devices if they are set to enhance the radiative decay rate of a quantum emitter? These questions are answered by proposing and modelling a new bullseye structure, optimized based on maximization of the far-field intensity along the optical axis. Dimensions and the performance of the proposed BE are then compared to that reported by another author. The other question to be answered is the characterizations techniques. What is the best approach in characterizing such devices? How many focal points are there along the optical axis? What are the strengths and depths of focus at each point? How collimated is the output? Techniques proposed by other authors are debated and a technique based on 3D confocal microscopy is proposed and applied experimentally to the newly proposed device.

8.1. Introduction

Bullseye structures are relatively simple, i.e. a single subwavelength cavity (or aperture/hole) surrounded by concentric circular gratings. Centre of a BE structure may accommodate annular rings [239], a circular hole [240] or a flat disk with no opening [128]. The cavity (or the aperture) in a BE structure may host a quantum emitter, such as a nano-diamond with NV- vacancy, to enhance it radiative decay rate (RDR) and improve the collection efficiency [73, 96, 241, 242]. Their simple planar design also makes them ideal for integration with the exit surface of other sources such as a quantum cascade laser [93] or fibre optics [128] to collimate the output light. The role of the corrugation may vary depending on the application. I tend to agree with the definition provided by Genet and Ebessen [243] and I quote: “If the output surface surrounding the aperture is also corrugated, a surprisingly narrow beam can be
generated, having a divergence of less than a few degrees, which is far smaller than that of the single apertures discussed earlier. This is because the light emerging from the hole couples to the periodic structure of the exit surface and to the modes existing in the grooves, which in turn scatter the surface waves into freely propagating light. This then interferes with the light that has travelled directly through the hole generating the focused beam [243].”

The influence of the BE geometry on the transmission efficiency has previously been investigated for a given wavelength [244], where the authors showed that the optimum groove width for maximum transmission efficiency is approximately \(0.5 \times \lambda_{\text{SPP}}\), where \(\lambda_{\text{SPP}}\) is the wavelength of surface plasmon polaritons (SPP). The number of corrugations where the transmission efficiency approaches saturation was found to be 6-10. Furthermore, it has been claimed that the far-field radiation pattern of a BE structure depends primarily on the corrugations surrounding the hole rather than the aperture’s geometry. It was confirmed that the distance between the aperture and its nearest groove, i.e. the radius of the first corrugation, influences the coupling between the LSPs inside the cavity and the SPPs in the corrugations, thus affecting the transmission efficiency [245]. The directionality of the emission from a NV- inside a cylindrical nano-diamond cavity surrounded by concentric circular corrugations engraved in a silver film was investigated in [73], see Figure 95. Such a design operates in the reflection mode, where the incident and the collection surfaces are the same.

![Figure 95: A cylindrical cavity in a metallic film surrounded by concentric circular corrugations to control the emission pattern from a dipole positioned at its centre [73].](image)

Although this design uses concentric circular corrugations to achieve collimated reflected beam, the enhancement to the RDR was reported to be poor, presumably due to the utilization of a non-resonant cavity. Yet, a more fundamental flaw in the design is the operation in reflection mode. In order to excite the single photon emitter inside the cavity, it must be positioned at the focal point of a confocal objective lens.
Point-illumination/point-collection using the same objective lens is an intrinsic feature of the confocal microscopy. Moving the objective lens away from the device into the far-field zone, (hence moving the emitter away from the focal point), reduces the incident power over the dipole drastically. Although the excitation of the dipole in the cavity may be achieved by a side-illumination technique via a separate objective lens, such configurations are more complex. In this design the choice of diamond superstrate is well justified as it prevents the emission of the nano-diamond cavity (at the centre of the BE) from total internal reflections. The inclusion of the diamond superstrate allows the emission to escape the nano-diamond and since diamond crystal has low absorption (due to its large band gap [246]) most of the emitted power may be collected from the superstrate. Other authors have also based their design on collecting the dipole’s emission from optically dense materials [240, 246-249]. Recent developments in nano-diamond fabrications have led to designs such as a thin diamond film (supported on a glass substrate) with BE grating etched on the surface surrounding a nitrogen vacancy to collimate its emission [242]. This device was reported to have a high collection efficiency when characterised with a confocal microscope in the reflection mode. The use of a homogenous diamond film with NV- near its surface was to avoid complications (such as total internal reflections) associated with designs where a nano-diamond is integrated with an external structure.

![Figure 96: BE grating etched on the surface surrounding a nitrogen vacancy [242]](image)

The far-field radiation pattern was obtained using back-focal-plane imaging techniques (using Bertrand lens) that eliminated the need to move the confocal objective away from the emitter, hence maintaining a constant power over the emitter at all time. Is this practical in real-life applications? One must consider both the in-situ and in-laboratory
conditions, where *in-laboratory* refers to the stage where a device is under investigation and *in-situ* referring to a stage where the device is integrated with its intended circuitry. When characterizing devices such as these (hence *in-laboratory* conditions), one needs to map its near- and far-field profiles. When using a Bertrand lens one has no idea on how far is the far-field. The ultimate goal behind subwavelength plasmonic devices here is miniaturization. Miniaturization of the whole confocal setup with Bertrand lens on the optical path so that the receiving antenna can benefit from the collimated beam in k-space is a very complex problem. It is only intuitive for such devices to be designed based on transmission geometry where *in-situ* and *in-laboratory* conditions are not too different. Figure 96 also shows that the field intensity in the air is lower in comparison to those inside the diamond film and the substrate. This may be due to the complex interaction of NV-emission with the patterned diamond film that possesses photonic crystal characteristics (i.e. band gap, wave-guiding … etc), and/or direct interaction of NV- with air/diamond and glass/diamond boundaries (which includes the total internal reflection and the quenching of the dipole’s radiation). It is obvious that the larger portion of the emitted power is unutilized, only trapped inside the diamond. Integrating nano-diamonds with a properly designed plasmonic structure in transmission mode may offer a solution to this problem.

### 8.2. Analysis and Design

Electromagnetic activities within the BE structure are complex. Dimensions of the cavity/aperture and the thickness of the film also have bearings on the strength of the electric field over the surface of the bullseye. The surrounding dielectric material, the distance of the first groove from the cavity/aperture, the depth, periodicity and the width of the corrugations all play a role in strengthening/suppressing scattered power and shaping the radiation pattern. In this section, a qualitative description of light-matter interactions in a plasmonic BE is provided and a BE design by another author is examined. Reliance on analytical solutions in modelling such a complex interaction in a seemingly simple structure is not fruitful. With the help of numerical solutions, however, one can optimize the design of the BE for a given wavelength that enhances the radiative decay rate (RDR) as well as producing highly directional light along its optical axis. Finally a novel technique in harnessing the power emitted by a NV- in a
diamond substrate is proposed and the performance of the BE is evaluated in such a setting. To date, the design of plasmonic BE structures has been primarily focused on utilizing a hole aperture and matching the period of its surrounding corrugations to the wavelength of the surface plasmon polaritons. Surface waves on a flat metallic film, however, are the results of evanescent electric fields normal to the surface boundaries. SPPs also carry electric fields that are parallel to the surface above and beneath a metal/dielectric interface. Components of the electric field that dominate the far-field along the optical axis, have been largely ignored in most BE designs so far. Here, it is first demonstrated that the normal and the parallel to the surface components of the electric field of light transmitted through a subwavelength resonant aperture exhibit higher amplitudes in comparison to those of a circular hole. By combining the resonant aperture with a BE structure and optimizing the corrugations to maximize the parallel components scattered into the far-field, it is shown that a highly directional beam reaching tens of microns away from the device is attainable. Furthermore, it is demonstrated that for a sufficiently thin metallic film, parallel components of the electric field from the substrate side, couple to the surface plasmon inside the corrugations resulting in further enhancements of the far-field intensity.

An investigation of the interaction between an aperture and a nearby groove via surface waves has been undertaken [250-252] and the coupling of waveguide modes to the fields inside the groove through a thin layer of metal has been demonstrated [253]. Here, a qualitative description of such interactions in 2D in terms of the $x$ and the $z$ components of the electric field are provided. Considering an $x$-polarized TM wave travelling in the $+z$ direction and normally incident on an optically thick “flat” metallic film (laid along the $x$-axis) perforated with a single subwavelength resonant aperture. The role of the aperture is to utilize some of the incident power to generate the electric field, $\vec{E}_a = E_{wa}\hat{n}_x + E_{za}\hat{n}_z$, which travels along the film’s surface. The interaction between $\vec{E}_u = E_{wu}\hat{n}_x + E_{zu}\hat{n}_z$ and the surface charge densities is limited only to surfaces that satisfy the condition $\hat{n} \cdot \vec{E}_u > 0$, where $\hat{n}$ is the vector normal to the surface. Corrugations surrounding an aperture, (Figure 97(a)), may be considered as a sequence of alternating vertical and horizontal surfaces. Upon arrival at a groove, $E_{zu}$ interacts with the surface charge density, $\sigma_z$, that leads to the accumulation of charges in the
corner of the groove, Figure 97(b). This gives rise to the surface charge density, $\sigma_x$, on the vertical surfaces and, subsequently, to an $x$-directed electric field, $E_{xc}$, inside the grooves. The interaction between the already established $E_{xc}$ and the newly arriving $E_{xa}$, (depending on their relative phase differences), either strengthens or weakens the surface charge density $\sigma_x$. The formation of a point source with a dipole moment $\vec{p}_i$ on the upper corner of a groove positioned at $(x_i, z_0)$, is the result of the surface charge oscillations $\sigma_z(x_i, z_0)$ and $\sigma_x(x_i, z)$, (Figure 97(b)). Properly spaced corrugations provide a mechanism to intercept the $E_{xa}$ with a correct phase. Spacing of the corrugations also controls the shape of the scattered light by the BE. Scattered field from the gratings, therefore, must make a constructive interference with that of the aperture along the optical axis.

Figure 97: (a) Components of a bullseye structure. (b) Formation of a hotspot with a dipole moment $\vec{p}_i$ on the upper corner of a groove at coordinates $(x_i, z_0)$, due to the surface charge oscillations $\sigma_z(x_i, z_0)$ and $\sigma_x(x, z)$. (c) Electric field components launched by a resonant cross-shaped aperture at $\lambda_0 = 700$ nm.

Contributions by the $E_{za}$ to these charge oscillations at $(x_i, z_0)$, however, are $180^\circ$ out of phase with those at $(x_i, z_0)$. This is simply due to the $E_{za}$ being an odd function of $x$, (compare the three components of the electric field along the film surface in Figure 97(c)). Consequently, any contribution by $E_{za}$ to $\vec{p}_i$ and $\vec{p}_{-i}$ (hence to their scattered fields) leads to a destructive interference along the optical axis. This is an adverse effect given that the $E_{za}$ is the strongest of the three components. In an optically thick BE structure, the corrugations should intercept the power carried by $E_{xa}$ that propagates along the surface to maximize $E_{xc}$ that propagates in the $z$-direction. For a BE structure
composed of \( N \) concentric corrugations, there are \( 2 \times N \) vertical surfaces that can potentially be utilized to maximize the \( x \)-component of the transmitted/scattered electric field. When drawing an analogy between the charge oscillations on the corner of a groove and a point dipole, a BE structure may be looked at as an antenna array that is composed of a number electric point dipoles, \( p_i \), positioned at \((x_i, z_0)\), where \( i \) is an integer. In such an antenna arrays, when \( x_i - x_{i-1} < \lambda_0 \), increasing the number of hotspots, strengthens the intensity in the central lobe \[132].

Garcia-Vidal et al. \[117\], proposed an analytical formalism to calculate the far-field radiation pattern of a slit flanked by surface corrugations using the Huyghen’s-principle \[95\]:

\[
H(r)_y = \frac{1}{\mu_0 c} \sum_{i=1}^{N} E_{xi} G(r, r_i) \tag{8.1}
\]

Here, \( H(r)_y \) is the total \( y \) component of the magnetic field calculated at position \( r \), from the superposition of electric fields at the indentations located at \( r_i \). Note that \( E_{xi} \) is related to the \( x \)-component of the electric field at the \( i^{th} \) indentation and \( G(r, r_i) \) is the scalar Green’s function, see equation (2) in \[95\] for full definition. Equation (8.1), however, does not reveal much about the interaction between the \( E_{xi} \) and its surroundings. The angular spectrum of a single dipole positioned near a planar interface is formulated by Novotny and Hecht \[8\]. The dipole moment of a point source may be described as \( \vec{p}_i = e^{-iq_i} p_{i0}(a\hat{n}_x + b\hat{n}_z) \), where \( \varphi_i, p_{i0} \) and \( (a_{ini} + b_{ini}) \) describe the phase, magnitude and the orientation of the \( i^{th} \) dipole moment respectively. The total transmitted/scattered electric field measured at an arbitrary location, \( r = \sqrt{x^2 + z^2} \), in the half-space above the BE surface is then described by the superposition of all the electric fields due to each dipole:

\[
\bar{E}(r)_m = \sum_{i=1}^{N} \omega^2 \mu_0 \mu_i \left[ \tilde{G}_0(r, r_i) + \tilde{G}_{ref}(r, r_i) \right] \vec{p}_i \tag{8.2}
\]

where \( r_i = \sqrt{x_i^2 + z_0^2} \) denotes the location of the \( i^{th} \) point source, \( \mu_0 \) is the permeability of free space and \( \mu_1 \) is the relative permeability of the medium within which the dipole’s radiation propagates. This is an extension to the equation (10.17) provided by Novotny
and Hecht [8]. The dyadic Green’s functions \( \tilde{G}_0(r,r_i) \) and \( \tilde{G}_{\text{ref}}(r,r_i) \) map the direct dipole radiation and its Fresnel reflection by the film into the half-space above the BE structure. For a full description of the dyadic Green’s functions \( \tilde{G}_0(r,r_i) \) and \( \tilde{G}_{\text{ref}}(r,r_i) \) see equations (10.6-10.21) and D1-D5 [8]. A consequence of equation (8.2) is the reflection from the planar interface contributing to the total radiated field. Considering a horizontally oriented dipole moment above a plane surface, it was shown that only 50% of the power is radiated into the upper half-space directly. The other 50% is incident on the surface below which is partly reflected back into the upper half-space. The remaining portion of the reflected power interacts with the dipole, see equation 10.48 [8]. Furthermore, Equation 10.32 [8] shows that only the parallel components of a dipole moment positioned near a planar surface, make contribution to the far-field along the \( z \) direction. For the reflection term not to vanish, however, the dipole must be positioned at a distance larger than \( \sim 10 \) nm above the metallic surface [8, 247]. High surface charge densities formed on the upper corners of a groove satisfy this condition with respect to the bottom surface, \( z_{\text{ref}} = z_0 - h \). To increase the reflective surface area, the width of periodic corrugations must occupy a larger portion of the BE structure as whole. To determine the optimum dimensions of a BE structure under such complex light-matter interactions, the use of equation (8.1) is not advisable since a previous knowledge about the amplitude and the phase of electric fields at each indentation is required. The same applies to equation (8.2) with respect to the magnitude and the phase of each dipole moment, not to mention the complexity involved regarding the interaction between dipoles and the film. The situation becomes even more convoluted when dealing with dimensions that are comparable with the skin depth. Therefore, the best approach here is to rely on numerical solutions, which have proven to be reasonably accurate all along this project.

It is obvious that \( \sigma(x_i,z) \) is a function of \( E_a \), however, the strength of the \( E_a \) depends on the distance travelled, decaying rapidly as it propagates away from the aperture along the surface. Utilizing a resonant aperture strengthens the \( E_a \) over the surface of the BE. To demonstrate this, a hole and a cross-shaped aperture were modelled in 3D. Our previous simulations [238] considered a symmetric cross-shaped aperture with arm-lengths \( 170 \pm 5 \) nm and arm-widths \( 40 \) nm perforated in a \( 100 \) nm silver film, laid
in z = 0 plane, supported by a glass substrate with a refractive index $n = 1.52$. The refractive index data for silver were taken from Palik [179]. These simulations showed a peak in transmission at the target wavelength $\lambda_0 = 700$ nm, corresponding to the emission wavelength of nitrogen vacancies in nano-diamonds at room temperature [254, 255]. The simulation is reproduced using an $x$-polarized TM wave at $\lambda_0 = 700$ nm, normally incident on the glass/silver interface. Full Maxwell equations were solved numerically using the finite element method (FEM) in COMSOL 4.3b. Simulations were repeated for a circular hole with a radius $R = 61.5$ nm. The radius of the hole was chosen so that it corresponds to the same surface area (or volume) as the cross. This will highlight the impact of the shape resonance associated with the cross-shape aperture on the surface fields. Electric field strengths at the silver/air interface calculated over the film surface produced by the cross and the hole apertures are depicted in Figure 98. The surface surrounding the cross, Figure 98(a)-(c), exhibits higher field amplitudes compared to that of the hole, Figure 98(d)-(f). Intensity ratios, 
\[ R_{x,y,z} = \frac{\int |E_{x,y,z}|^2_{\text{cross}} \, dV}{\int |E_{x,y,z}|^2_{\text{hole}} \, dV}, \]
calculated for each component over the film surface at the air/silver interface were found to be $R_x = 12$, $R_y = 27$ and $R_z = 32$. Here $|E|^2_{\text{cross}}$ and $|E|^2_{\text{hole}}$ are the field intensities associated with the cross and the circular hole respectively. Our models also showed strong localization of the electric fields in the vicinity of the aperture with their amplitude decaying rapidly with the distance, see Figure 98(a)–(c).
Figure 98: Amplitudes of the $x$, $y$, and $z$ components of the electric field obtained from 3D models, calculated over the exit surface surrounding (a)-(c) the cross aperture and (d)-(f) the circular aperture. (g) $z$ component of the electric field calculated in the vicinity of the cross aperture: (red) amplitude, $|E_z(x)|$ (grey and blue) $E_z(x,t)$ at two arbitrary times $t_1$ and $t_2$. (h) 3D calculations of the time averaged radial component of the Poynting vector vs. the angular direction in the $x$-$z$ plane for (red) circular aperture and (blue) cross aperture.

Furthermore, Figure 98(g), suggests that the separation between the LSPs and SPPs occur at $x \approx 200$ nm (see lines in brown and blue). Here $x = 0$ marks the centre of the aperture. Calculations of the time averaged radial component of the Poynting vectors $<S>_{\text{hole}}$ and $<S>_{\text{cross}}$ vs. the angular direction in the $x$-$z$ plane for the circular hole and the cross apertures, Figure 98(h), showed the power emanating from the cross (besides being stronger) is more directional in comparison to the hole. Enhancement to the overall throughput, $R = 11.3$, was calculated using $R = P_{\text{cross}}/P_{\text{hole}} = \frac{\int <S>_{\text{cross}} \cdot ds}{\int <S>_{\text{hole}} \cdot ds}$, where total powers, $P_{\text{cross}}$ and $P_{\text{hole}}$, were calculated by integrating the time averaged radial component of the Poynting vectors over the surface of a hemisphere, 2.3 $\mu$m in radius, encompassing the apertures.

Full 3D modelling of an aperture in the presence of corrugations is computationally expensive, particularly when parametric sweeps are performed over the geometry. To reduce the number of degrees of freedom, I have modelled the BE in 2D that exhibits
strong resemblance to the 3D model as far as the radiation patterns are concerned. The radiation pattern of a 50 nm wide slit perforated in a 95 nm thick silver film on a glass substrate modelled in 2D, closely matches that of a 3D model discussed above. Corrugations were incorporated into the 2D model with depth \( h = 50 \text{ nm} \) and number of grooves \( N = 7 \). The width of the periodic corrugations, was set to be a function of the periodicity, \( p \), such that \( w_p = p - 86 \text{ nm} \). This ensures that reflective surface at the bottom of the corrugations widen as the period increases. The 86 nm wide indentation was chosen to be 3.5 times the skin depth, to prevent the mixing of the charge oscillations formed on the adjacent vertical surfaces while allowing for the widest possible grooves. Note that COMSOL calculates the skin depth as \( S = 24.6 \text{ nm} \) at \( \lambda_0 = 700 \text{ nm} \). The width of the first corrugation was selected so that the edge of the disk surrounding the aperture coincides with the strongest peak associated with the SPP wave, see Figure 98(g). The device was illuminated from the glass/silver side by a TM wave at normal incident travelling in the \( +z \) direction and parametric sweeps were performed over \( p \). The transmitted far-fields were calculated over a closed arc encompassing the aperture and the corrugations. Integrating the \( x \)-component of the far-field intensity and plotting it vs. the period, revealed multiple maxima, see Figure 99(a). Although there are additional maxima beyond the range of data calculated here, to keep the device compact, a periodicity of \( p = 493 \text{ nm} \) was chosen. In the case of \( t_c \sim S \), see Figure 97(b), a new effect comes into play, namely the coupling of the \( x \)-components of the electric field from the substrate, \( E_{sg} \), to the LSPs inside the grooves. The increasing trend in the continuum with respect to \( p \), seen in Figure 99(a), is attributed to the widening of corrugations with increasing \( p \), that results in more of the incident power to leak through the film. This power leakage partially couples to the vertical surfaces and partially is transmitted without any interaction. While the use of resonant aperture enhances the \( E_a \), coupling of the \( E_{sg} \) to LSPs inside the grooves presents an opportunity to further enhance \( E_{xc} \) directly. Amplitudes of the \( x \) component of the electric polarization, \( |P_x| \), inside the film and the total electric field, \( |E_x| \), in the surrounding dielectrics in the \( x-z \) plane calculated for \( h = 50 \text{ nm} \), Figure 99(c) and (inset), also confirms the formation of hotspots and the coupling between the \( E_{sg} \) to the LSPs inside the corrugations.
Figure 99: (a) The \( x \)-component of the far-field intensity integrated over an arc encompassing the aperture and the corrugations vs. the period, normalized to the maximum intensity. (b) Calculated \(|P_x|\) inside the film and \(|E_x|\) in the surrounding dielectrics for \( h = 50 \text{ nm} \) and (inset) close-up of the corrugations showing \( E_x \) penetrating the film. (red = 1 and blue = 0)

Nominated configuration, C6, was selected with \( r_1 = 621 \text{ nm} \), \( w_1 = 422 \text{ nm} \), \( p = 493 \text{ nm} \), \( w_p = 407 \text{ nm} \) and \( N = 7 \). Configuration reported by Yi et al. [256] was also modelled at \( \lambda_0 = 660 \text{ nm} \) for comparison. Table 3 lists the dimensions associated with each device.

<table>
<thead>
<tr>
<th>Device</th>
<th>( r_1 ) (nm)</th>
<th>( w_1 ) (nm)</th>
<th>( p ) (nm)</th>
<th>( w_p ) (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C6</td>
<td>621</td>
<td>422</td>
<td>493</td>
<td>407</td>
</tr>
<tr>
<td>( d = 50 \text{ nm} ) ( h = 50 \text{ nm} ) ( t = 95 \text{ nm} )</td>
<td></td>
<td></td>
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<tr>
<td>Cref</td>
<td>620</td>
<td>200</td>
<td>620</td>
<td>200</td>
</tr>
<tr>
<td>( t = 300 \text{ nm} ) ( d = 300 \text{ nm} ) ( h = 80 \text{ nm} )</td>
<td></td>
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</tbody>
</table>

8.2.1. Excited with a normally incident light

Xiao Ming Goh [257] has investigated the integration of a resonant cross-shaped aperture with concentric circular corrugations previously. To compare the performance of the C6 configuration in the presence of a resonant cross aperture vs. a circular hole, corrugations were incorporated into the 3D models. The number of corrugations, however, were limited to four, (instead of \( N = 7 \)), to reduce the computational resources
needed. For each device, simulations were first carried out with a Gaussian incident beam 
\[ E_x = \frac{1}{\sqrt{2\pi}} e^{-\frac{x^2 + y^2}{2\alpha^2}}, \]
where \( \alpha \) is the standard deviation that sets the beam width.

Notice that the standard deviation is eliminated from the denominator of the multiplier, i.e. first term in the RHS, to keep the incident power over the aperture constant while varying the width of the incident beam. With \( \alpha = 199 \) nm, contributions resulting from the \( E_{xx} \) to LSP coupling are minimized, Figure 100(a)-(g). This ensures that the interaction between the corrugations and \( E_a \) is more observable, hence comparable. Simulations were then repeated with an incident beam covering the whole device to include the \( E_{xx} \) to LSP coupling, Figure 100(h). Notice the higher amplitudes of the electric field components on the surfaces surrounding the cross, Figure 100(a)-(c), compared to those of the circular hole, Figure 100(d)-(f). The cross-shaped aperture and the disk surrounding it constitute a “resonant system” that brings about further enhancement in the transmitted power and improvement to the directionality. Radiation pattern of the BE with the cross shows 5 times the intensity of the BE with the circular hole, Figure 100(g). Comparing the radiation patterns of the cross in isolation, Figure 98(h) (line in blue), and in C6 settings, Figure 100(g) (lines in blue), transmitted power along the optical axis is enhanced by a factor of \( \sim 10 \), accompanied with a significant reduction in angular divergence. Radiation patterns shown in Figure 100(g) were calculated along a semi-circle, 2.3 \( \mu \)m in radius measured from the centre of the BE.

Figure 100: Electric field components on the surfaces surrounding (a)-(c) the resonant cross aperture and (d)-(f) the circular hole obtained from 3D models. Calculations of the time averaged radial component of the Poynting vector vs. the angular direction, calculated in the \( x-z \) plane at the boundaries of a hemisphere encompassing the device for (g) \( \alpha = 199 \) nm.
When illuminated with a normally incident light, it is not possible to isolate the contribution of the leaked power that couples to the vertical surfaces from that transmitted directly into the air half-space. A comparison between C6 and C_{ref}, therefore, must be carried out using an optically thick film. The film thickness in the C6 configuration therefore was set to \( t = 300 \) nm. The aperture width of the C6 was also set to \( d = 318 \) nm in order to maintain the same aspect ratio \( d/\lambda_0 \) specified for C_{ref}. Figure 101 shows the radiation patterns of (a) C6 and (b) C_{ref}, with both film thicknesses set to \( t = 300 \) nm and the same \( d/\lambda_0 \) ratios. The radiation pattern associated with C_{ref} showed stronger side lobes in comparison to C6, where the radiation pattern is well confined to the central lobe. Although the radiation pattern of C_{ref} showed a longer reach along the central lobe, its intensity drops rapidly beyond 3 \( \mu \)m from the surface (in comparison to 8 \( \mu \)m observed in C6).

Figure 101: \( |E|^2 \times 62.5 \text{ (V/m)}^2 \) of (a) C6 and (b) C_{ref}, with both film thicknesses set to 300 nm and the same \( d/\lambda_0 \) ratios.

How do the C6 and C_{ref} configurations interact with the power emitted by a dipole placed inside the holes? Can these structures enhance the RDR of a single photon emitter while retaining their beaming qualities? What is the most efficient way of integrating a nano-diamond with the aperture? These questions are answered in the next section.
8.2.2. C6 with a nano-diamond inside the aperture

An isolated slit having a width $d = 50$ nm with a nano-diamond at its centre was modelled in 2D. The centre of the nano-diamond was configured with a dipole having its moment oriented along the $x$-axis. Total power scattered into the air was calculated at $\lambda_0 = 700$ nm for various nano-diamond diameters in the range of $40$ nm $\leq D \leq 50$ nm. Maximum power was obtained for $D = 50$ nm when the nano-diamond makes contact with the inner surfaces of the cavity. Various positions along the slit’s axis, i.e. $z$-axis, revealed that the power ratio $P_{\text{air}}/P_{\text{sub}}$ is maximized when nano-diamond is positioned at $z = 25$ nm where the nano-diamond makes contact with the substrate. Here $P_{\text{air}}$ and $P_{\text{sub}}$ are the total powers emitted into the air and the substrate respectively and the position $z$ is measured from the centre of the nano-diamond. For convenience, lets label this source system as “50/25”, signifying a 50 nm wide slit (perforated in a silver film) with a nano-diamond that is 50 nm in diameter, positioned at $z = 25$ nm at the centre of the slit. Here $z = 0$ marks the silver/glass interface.

Positioning a 50 nm wide nano-diamond at the centre of $C_{\text{ref}}$ (i.e. the centre of the hole that is 300 nm in diameter) did not result in a strong coupling between the dipoles radiation and the hole, Figure 102(a). Performance of $C_{\text{ref}}$ in the presence of the 50/25 system was also inadequate, see Figure 102(b). C6 configuration, on the other hand, exhibits a highly directional light with strongest intensity along the central lobe.

![Figure 102: $|E|^2/1.3\times10^{17}$ (V/m)$^2$ for (a) $C_{\text{ref}}$ with 50 nm in diameter nan-diamond positioned inside the aperture, (b) $C_{\text{ref}}$ in the presence of the 50/25 system and (c) C6 $C_{\text{ref}}$ in the presence of the 50/25 system.](image)

To make a fair comparison between C6 and $C_{\text{ref}}$, both devices were excited with the 50/25 source system. Optimum thicknesses for C6 and $C_{\text{ref}}$ in this case were found to be $t = \{365$ and $350\}$ nm respectively, corresponding to the second Fabry-Pérot resonance of each slit in the presence their respective corrugations [84]. Therefore, it is safe to say
that the comparison between the two would highlight the impact of BE geometry in the presence of a resonant slit and in the absence of any leakage. The enhancement to the RDR, \( RDRE = \frac{P_{\text{air}}}{0.5 \times P_{\text{nano-diamond}}} \), for \( C_{6_{r=365\text{nm}}} \) and \( C_{\text{ref}_{r=350\text{nm}}} \) were found to be 30 and 34 respectively. Here \( P_{\text{nano-diamond}} \) is the radiated power of a 50 nm nano-diamond in vacuum and \( P_{\text{air}} \) is the power radiated by the same nano-diamond into the air half-space when integrated with the device. The factor of 0.5 in the denominator is due to the \( P_{\text{nano-diamond}} \) being calculated along the arc length of a full circle, whereas \( P_{\text{air}} \) was calculated over a semicircle in the air half-space. Radiation patterns produced by \( C_{6_{r=365\text{nm}}} \) and \( C_{\text{ref}_{r=350\text{nm}}} \) are depicted in Figure 103(a) and (b) respectively and the \( x \)-component of the electric field at the silver/air interfaces of both devices are shown in Figure 103(c).

![Figure 103](image)

Figure 103: \(|E|^2/1.3 \times 10^{17}(\text{V/m})^2\) by (a) \( C_{6_{r=365\text{nm}}} \) and (b) \( C_{\text{ref}_{r=350\text{nm}}} \). (c) The \( x \)-component of the electric field at the silver/air interfaces of both devices.

Radiated power of \( C_{\text{ref}} \) becomes extremely weak along the central lobe beyond 2 microns from the surface. \( C_{6} \) on the other hand showed that most of the radiation is directed towards the central lobe extending \( \sim 9 \) microns away from the surface.
C6 configuration was also modelled with $t = 120 \text{ nm}$ and $h = 70 \text{ nm}$ in the presence of the 50/25 source system where the emission was found to be at its maximum. Figure 104(a) shows the electric field intensity scattered by the device. Directional gain of the antenna $|E(\theta)|^2 / |E_0(\theta)|^2$, Figure 104(b), shows that the field intensity along the optical axis of the device is 125 times that of a free standing nano-diamond in vacuum. Here $|E(\theta)|^2$ and $|E_0(\theta)|^2$ are the electric field intensities of the nano-diamond in the C6 settings and in vacuum respectively as a function of angle, $\theta$, from the optical axis. Placing a nano-diamond inside the 3D cross-shaped aperture, however, required further investigations to identify its optimum dimensions. 3D simulations revealed that a cross with arm-lengths $L = 140 \text{ nm}$ and arm-widths $W = 30 \text{ nm}$ perforated in a 120 nm silver film has its resonance at $\lambda_0 = 700 \text{ nm}$ when excited with a nano-diamond (50 nm in diameter) positioned at $z = 25 \text{ nm}$ at its centre. Figure 104(c) shows that the electric field intensity in the 3D model agreeing with the 2D model.

Figure 104: (a) $|E|^2/1.3\times10^{17} \text{ (V/m)}^2$ scattered by C6$_{t=120\text{nm}}$ when excited by the 50/25 source system with $t = 120 \text{ nm}$ and $h = 70 \text{ nm}$. (b) Simulated directional gain of the antenna $|E(\theta)|^2 / |E_0(\theta)|^2$ as a function of angle, $\theta$, from the optical axis. (c) 3D simulation of the $|E|^2/1\times10^{31}\text{ (V/m)}^2$ scattered by C6$_{t=120\text{nm}}$ with the cross arm-lengths $L = 140 \text{ nm}$ and arm-widths $W = 30 \text{ nm}$. Film thickness, the corrugations, nano-diamond diameter and its position inside the aperture were set according to the 2D model.
Further enhancement to the decay rate, \((\text{RDRE} = 39\) for \(C6_t=120\text{nm}\) compared to 30 for \(C6_t=365\text{nm}\)), however, is partly due to the difference in powers emanating from the slit at two different thicknesses. To quantify the impact of the leakage alone, the loss/gain factor due to the grooves were calculated as \((1-P_{\text{air-hole}}/P_{\text{air-C6}})\times100\%\) and was found to be -5% and +6% at thicknesses \(t = \{365, 120\} \text{ nm}\) respectively. Here \(P_{\text{air-hole}}\) is the power emitted into the air by the source system in the absence of any groove. This is a total of 11% gain due the \(x\)-component of the electric field that propagates along the silver/glass interface that partially couples into the corrugations via the leakage, see Figure 105. This is a remarkable result given that the power ratio \(P_{\text{sub-hole}}/P_{\text{total-hole}} = 0.22\) indicates that an isolated 50/25 source system (with thickness \(t = 120 \text{ nm}\)) scatters only 22% of the total power into the substrate with only \(P_{\text{sub-surface-hole}}/P_{\text{total-hole}} = 18\%\) available on silver/glass surface.

![Figure 105: x-component of the electric field calculated for C6 with at an arbitrary time for (a) \(t = 365 \text{ nm}\) and (b) \(t = 120 \text{ nm}\). (Red =1, blue = -1, green=0).](image)

### 8.2.2.1. Experimental trials

During the course of this project, nano-diamonds that exhibit anti-bunching properties were identified and characterized. The idea behind the pick-and-place approach, see Appendix B, was to identify and characterize a single photon emitter prior to its integration with any plasmonic device followed by a post-integration characterization to detect any changes in the RDR/life-time. Positioning a nano-diamond inside a plasmonic aperture/cavity using the pick-and-place technique, however, proved to be a challenge. Attractive forces between the tip of the micromanipulator and the nano-diamond (perhaps Van Der Waals) created an undesirable scenario where the nano-diamond became permanently attached to the tip. It was also argued that the pick-and-place approach may also change the orientation of the NV- with respect to that in which it was characterized, hence voiding any measurements of its life-
time/ant-bunching made prior to the integration. Furthermore, forcing the nano-diamond into the cavity, if achievable, may have some undesirable side effects such as distorting the apertures geometry, hence shifting its resonance away from the target wavelength. In the following section, a novel technique is proposed for harnessing the radiation of a dipole that may help avoiding such complications.

8.2.3. Surface Plasmon-Coupled-Enhanced Transmission in diamond substrates with a NV near the surface

Note that the main goal of this chapter is not the optimization of the BE structure, but rather a new approach of integrating a single photon emitter with the device. Therefore the simulations are carried out using the C6 configuration with the original $t = 100$ nm and $h = 50$ nm, unless specified otherwise. Previously I mentioned the SPCE and I quote:

(“Theoretical and experimental studies of surface plasmon-coupled emission have shown that fluorophores within about 10 nm of the metal are quenched. Hence surface plasmon-coupled emission occurs for fluorophores in a region 10 to 50 nm above the metal” [247]).

The quenching effect, however, is misunderstood mainly due to the theoretical studies and the experimentally reported data being limited to the calculated/measured “reflected” power of a dipole near a flat metallic surface [258-261]. In fact, this “quenching” effect in the region less than \(~10\) nm away from the metallic surface is nothing but a strong coupling of the dipole’s radiation to the surface waves that propagate at the supercritical angle along the metal/dielectric interface (results not shown here). Based on my numerical analysis of a dipole near a planar metallic surface, the most accurate account of such an interaction is given by Novotny and Hecht [8]. Foreseeing a future design, the device proposed in this report was developed so that a single crystal diamond membrane with nitrogen vacancy implants near the surface [262] is used as a substrate. The C6 configuration was simulated in 2D with a diamond substrate where the incident field was replaced by the radiation of a dipole positioned at $(0, z)$, with its moment oriented in the x-direction. Here $z = 0$ marks the diamond/silver interface. The power ratio $P_{\text{air-C6}}/P_{\text{sub-C6}}$ vs. the $z$ is shown in Figure 106(a). The
maximum power ratio of $P_{\text{air-C6}}/P_{\text{sub-C6}} \approx 4$ is achieved for $z \approx -10$ nm, for which the $P_{\text{air-C6}}/P_{\text{air-noFilm}} = 9$ is the enhancement due to the presence of the C6 device, where $P_{\text{air-noFilm}}$ is the total power scattered into the air in the absence of C6. And if one defines the RDRE with respect to the radiation of a free standing nano-diamond in vacuum, $P_{\text{nano-diamond}}$, the enhancement rises to $RDRE = P_{\text{air-C6}}/0.5 \times P_{\text{nano-diamond}} = 40$. Beaming profiles for various $z$ values are depicted in Figure 106(b)-(g). In the range of $0 > z > -35$ nm, most of the power emanating from the dipole is coupled into the corrugations, via the slit, producing transmitted beams that are collimated. For $z < -35$ nm, the coupling strength decreases and ultimately at $z = -200$ nm, fields from the dipole decouple from the surface and the diamond/silver interface becomes reflective, Figure 106(h).

Figure 106: (a) Power ratio vs. the $z$ calculated as $P_{\text{air-C6}}/P_{\text{sub-C6}}$ where $P_{\text{air-C6}}$ and $P_{\text{sub-C6}}$ are the total power scattered into the air and the substrate respectively. (b)-(g) $|E|^2/1.3 \times 10^{17}$ (V/m)$^2$ vs. the dipole distance from the surface. (h) weakly coupled regime occur between the surface modes and the dipole’s emission occurs at $z \leq -200$ nm, where the film becomes reflective.

Further simulations were carried out for few different scenarios. A diamond substrate in the absence of any silver film with a dipole positioned at $z = -10$ nm, Figure 107(a), resulted in a power ratio of $P_{\text{air-noFilm}}/P_{\text{sub-noFilm}} = 0.2$. This is a clear indication that in the absence of any plasmonic structure, the larger portion of the emitted power is scattered inside the diamond. Introducing a 100 nm thick silver film on top of the diamond membrane, Figure 107(b), reduced the scattered power into the substrate by only 20% while the transmitted power became negligible. If a 50 nm wide slit perforates the flat 100 nm thick silver just above the dipole, Figure 107(c), power ratios $P_{\text{air-Ag100nm+hole}}/P_{\text{sub-Ag100nm+hole}} = 6.5$, $P_{\text{sub-Ag100nm+hole}}/P_{\text{sub-noFilm}} = 0.29$ and $P_{\text{air-Ag100nm+hole}}/P_{\text{sub-noFilm}} = 0.29$.
Ag100nm+hole/P_{air-noFilm} = 9.5 are obtained, however, the radiation pattern in this case is dispersive. So it seems that by positioning the dipole inside the substrate just 10 nm (or less) below the aperture, most of the dipole’s radiation is scattered into the superstrate via the dipole-cavity coupling. A separate model with the C6 configuration having a film thickness \( t = 200 \text{ nm} \) while retaining all other parameters, resulted in \( t - h = 150 \text{ nm} \) being much larger than the skin depth of 25 nm. Although the radiation pattern of “C6_{Ag200nm}” is similar to that of the C6, see Figure 107(d), the power ratio \( P_{air-C6-Ag200nm}/P_{sub-C6-Ag200nm} \approx 0.5 \), (compared to \( P_{air-C6}/P_{sub-C6} \approx 4 \)) is obtained. Reduction in the power ratio is due to the increased film thickness that shifts the Fabry-Pérot resonance [84] (see Figure 19 in section 1.3.2) which also reduces the coupling via the leakage. The process of quantifying each effect is not repeated here. The comparison between the C6_{Ag200nm} and C6 is an indication of the sensitivity of the absorbed power by the substrate with respect to the film thickness. Note the higher field accumulation along the diamond/silver interface in Figure 107(d). Another scenario is when the corrugations are milled through the film, i.e. \( t - h = 0 \), Figure 107(e). In this case the power ratio \( P_{air-C6-h100nm}/P_{sub-C6-h100nm} = 0.92 \) (compared to 4 for \( h = 50 \text{ nm} \)) is a clear indication that the film underneath the corrugations also participates in harnessing the fields from the silver/diamond interface. Strong field distributions along the film surface observed in Figure 106(b) is an indication of an underutilized power.

Fabrication of ultra-thin diamond membranes in the shape of rings is now a possibility [263]. Filling the corrugations with such a high refractive index material assists in trapping the fields that travel along the exit surface. With a properly designed BE structure, this results in further enhancements to the LSPs inside the grooves which ultimately fortifies the coupling between the substrate and the corrugations via the leakage. Figure 107(f) depicts the intensity of the electric field when the substrate and the material filling the C6 corrugations are set to diamond. In this configuration, although the power ratio \( P_{air-C6}/P_{sub-C6} = 2.7 \) (compared to 4 in Figure 106(c)), the transmitted beam profile shows no distinct side lobes with most of the power being redirected towards the central lobe. The enhancement to the radiative decay rate in this case was found to be \( RDRE = P_{air-C6}/0.5 \times P_{nano-diamond} = 45 \) (compared to 40 in Figure 106(c)). With the diamond substrate and diamonds filling the grooves, the loss/gain factor, \( (1-P_{air-hole}/P_{air-C6}) \times 100\% \), at two thicknesses corresponding to the Fabry-Pérot
resonances of the slit in the presence of the grooves were calculated as $-21\%$ and $-4\%$ for $t = \{360, 110\}$ nm respectively, i.e. a total of $17\%$ gain (compared to $11\%$ in the previous section) purely due to the coupling of the $x$-components of the electric field via the leakage.

Figure 107: $|E|^2/1.3\times10^{17}$ (V/m)$^2$, for a diamond substrate with an NV- positioned 10 nm below the surface with (a) no silver film, (b) 100 nm thick silver film, (c) 100 nm thick silver film with a 50 nm wide slit, (d) C6 configuration with $t = 200$ nm, (e) C6 when corrugations are milled all the way through the film and (f) C6 having refractive index of the material filling the corrugations set to 2.41.

One can therefore design a BE structure with no aperture and purely based on the leakage. One possible design criteria may be based on the wave function:

$$\psi = \frac{1}{\sqrt{2}} \left( \psi_a e^{i(k_a x - \omega t)} + \psi_g e^{i(k_g x - \omega t)} \right)$$

(8.3)

where $\psi_a e^{i(k_a x - \omega t)}$ and $\psi_g e^{i(k_g x - \omega t)}$ are the surface waves travelling at the silver/air and the silver/diamond interfaces respectively. Here the probability amplitude of $1/\sqrt{2}$ implies a state with equal contributions by SPPs from both the substrate and the superstrate. In an optically thick silver film, SPP wavelengths at the air/silver and the diamond/silver interfaces were numerically calculated for $\lambda_0 = 700$ nm to be $\lambda_a = 2\pi/k_a = 667$ nm and $\lambda_g = 2\pi/k_g = 230$ nm, see Figure 118(c) and Figure 119(e) in section 10.1. To satisfy the requirement for equal contributions, (hence equal probabilities of $(1/\sqrt{2})^2$ for periodicity to match SPP wavelengths from both sides of the film) the periodicity may be set to $P = (\lambda_a + \lambda_g)/2$. While SPPs launched at the air/silver interface couple to the corrugations through direct propagation along the
surface, those at the diamond/silver interface may couple into the corrugations via the leakage. Setting the film thickness to $t = 100$ nm prevents mixing of the SPPs from the two sides. The width of the corrugations were narrowed to $w = 50$ nm to minimized the surface area they occupy. The interaction between surface waves at the two interfaces is then limited only to their coupling to the LSPs inside the grooves. Depth of the corrugations were set to $h = 70$ nm (hence $t_c = 30$ nm) using a trial/error approach until a strong collimated beam was obtained. Figure 108 shows the radiation patterns of one possible configuration, $C_x$, with no aperture. In summary the film thickness, periodicity, height and the width of the corrugations in $C_x$ were set to $t = 100$ nm, $P = 445$ nm, $h = 70$ nm and $w = 50$ nm respectively with the corrugations “centred” at $m \times P$, where $m$ is an integer. Eliminating the aperture and the first groove surrounding it simplifies the design to a large extent, making the device suitable for mass production fabrication techniques, such as the single step nanoimprinting lithography [264]. Note that in this chapter, 2D field plots in Figure 102, Figure 103, Figure 104, Figure 106, Figure 107 and Figure 108 were produced on the same scale to facilitate the comparison between devices.

![Figure 108](image)

Figure 108: $|E|^2/1.3 \times 10^{17}$ (V/m)$^2$ calculated for the $C_x$ configuration that has no aperture with periodicity matching the SPP wavelength at (a) silver/air interface $\lambda_a = 2\pi/k_a = 667$ nm, (b) silver/diamond interface $\lambda_d = 2\pi/k_d = 230$ nm and (c) $P = (\lambda_a + \lambda_d)/2$. Device was excited with a NV- positioned at $z = -10$ nm from the silver/diamond interface.

Fabricating the device on a diamond membrane with a NV positioned at $z = -10$ nm is a challenge and beyond the scope of this thesis. Perhaps, in the near future, when the relevant technology matures, such a device becomes realizable. Therefore, the BE devices (see Appendix A) were fabricated on the glass substrate and characterized by simply measuring their transmission when they were illuminated with an incandescent light filtered at $\lambda_0 = 700$ nm, incident on the substrate. In the next section fabrication and characterization of one such device is reported.
8.3. Fabrication and Characterization

A 2 nm thick germanium film followed by a 100 nm thick silver film were deposited on a glass substrate using IntlVac Nanochrome II electron beam evaporator. The germanium layer acts as an adhesion layer between the glass and the silver film and has little impact on the overall optical properties of the device. Corrugations for the C6 were engraved on the silver film according to the target dimensions discussed above with a depth of \( h = 50 \) nm, using a Focused Ion Beam (FIB) (Helios NanoLab 600). The aperture was milled through the film as a symmetric cross with target arm-lengths \( L = 175 \) nm and arm-widths \( W = 40 \) nm. It is worth mentioning that the preparation of bitmap files in our previous attempts to mill the cross-shaped apertures was based on a pixel size occupying a 10×10 nm\(^2\) area on the film surface, see section 5.3.1.1. With a Gaussian beam profile and an effective beam diameter of 7.5 nm, (corresponding to the 1.5 pA ion current), milling a perfect 10 nm × 10 nm square pixel on the film surface is impractical, see Figure 109(a). Here, a new technique was employed based on a 3.25 nm per pixel resolution, while keeping the incident ion beam as above. Such a technique leads to a beam overlap between the neighbouring pixels, Figure 109(b), that produce a cross-shaped aperture with abrupt geometries, hence improve the fabrication quality.

Fabricated device dimensions are listed in Table 4 measured from the relevant SEM images, Figure 110.
Figure 110: SEM images for the fabricated C6 configuration. (a) Top view. (b)-(c) close-up images for the aperture. (d) close up of the top view.

Table 4: Summary of the fabricated dimensions.

<table>
<thead>
<tr>
<th></th>
<th>$r_1$ (nm)</th>
<th>$w_1$ (nm)</th>
<th>$p$ (nm)</th>
<th>$w_p$ (nm)</th>
<th>$L_x$ (nm)</th>
<th>$L_y$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>FIB</td>
<td>634±12</td>
<td>425±3</td>
<td>506±13</td>
<td>413±6</td>
<td>185±2</td>
<td>177±2</td>
</tr>
</tbody>
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The film was mounted on a 3D piezoelectric translational stage within a confocal microscopy system. The glass/silver interface was illuminated with a collimated white light source (Supercontinuum Fianium SC-450-2), band-pass filtered at $\lambda = 700$ nm with a FWHM of 10 ± 2 nm (Thorlabs FB700-10). Transmitted intensities were collected via a LU PLAN Nikon 100× objective with a numerical aperture of NA = 0.95 and coupled into a multimode fibre with a core diameter of 50 µm (acting as a pinhole). The high axial resolution of the pinhole geometry allowed us to measure the intensity at the objective’s focal positions with a high accuracy (see Table 5). The light was detected using high sensitivity single photon counting modules (SPCM). The detector and the stage were controlled using a LabView program, which also collected data. Here, the surface of the film is taken to be parallel to the $x$-$y$ plane. A set of 2D images were obtained by raster scanning in the $x$-$y$ plane. The closest distance between the objective and the sample, $z = z_{ini}$, was set by an algorithm that maximized the photon counts as a function of $z$. The distance was increased in steps of $\Delta z = 0.1$ µm, between each scan up to 25 µm. The 2D images were stacked to form a high-resolution 3D volumetric map of the Fresnel zone. The process was then repeated with $\Delta z = 1$ µm to map the intensity over longer distances up to 55 µm. Although $\Delta z = 1$ µm results in a lower resolution in $z$, it reduces the time required for longer range measurements where the intensity
variation with respect to $z$ is relatively slow. Figure 111 shows a schematic of the scanning process.

![Figure 111: A set of 2D images obtained by raster scanning in the x-y plane at various distances from the device. 2D images were stacked to form a high-resolution 3D volumetric map of the transmitted intensity.](image)

8.4. Results and Discussion

Figure 112(a) shows the simulated result for $|E|^2$ along the optical axis. Depth of focuses, $\text{DOF}_1$ and $\text{DOF}_2$ are the lengths over which the intensity remains greater than half the local maxima at $z_1 = 5.8 \, \mu m$ and $z_2 = 27 \, \mu m$ respectively. Figure 112(b) depicts the $|E|^2$ in the $x$-$z$ plane. Normalized $|E|^2$ calculated along the $x$-axis at the two maxima, Figure 112(c)-(d), reveals the lateral beam profiles where $\delta_1$ and $\delta_2$ are the corresponding FWHM. Figure 113 depicts the results obtained experimentally. A 3D iso-surface obtained from the volumetric data corresponding to the $0.5 \times I_{\text{max}}$, where $I_{\text{max}}$ is the maximum photon counts, exhibits a rotational symmetry about the z-axis, see Figure 113(a). Normalized photon counts along the central lobe, Figure 113(b), and a 2D slice in the y-z plane obtained from the volumetric map, Figure 113(c), show good agreement with our simulations. The experimental results for $\delta_1$ and $\delta_2$ at their corresponding maxima were obtained from the normalized photon counts in the x-y
plane, \((I(x,y) - I_{\text{min}})/(I_{\text{max}} - I_{\text{min}})\), where \(I_{\text{min}}\) is the background radiation, see Figure 113(d)-(e). Table 5 summarizes the results.

**Figure 112**: Simulated results for the C6 configuration using a 2D model. (a) Maxima positions and their corresponding depth of focuses identified on \(|E|^2\) along the optical axis. (b) \(|E|^2\) in the x-z plane. (c)-(d) \(|E|^2\) calculated along the x-axis at their maxima.

**Figure 113**: Experimental results for the C6. (a) 3D iso-surface corresponding to the 0.5\(\times I_{\text{max}}\). (b) Normalized photon counts along the central lobe. (c) A 2D slice in the y-z plane obtained from the volumetric map. (d)-(e) Normalized photon counts in the x-y plane at \(z_1 = 5\ \mu m\) and \(z_2 = 26\ \mu m\).

Table 5: Locations of maxima and their corresponding full width half max and depth of focuses of the beam for the C6 configuration.
<table>
<thead>
<tr>
<th>Simulation</th>
<th>Experiment</th>
<th>Simulation</th>
<th>Experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td>$z_1$ (μm)</td>
<td>$z_2$ (μm)</td>
<td>5.8</td>
<td>5±0.5</td>
</tr>
<tr>
<td>$\delta_1$ (μm)</td>
<td>$\delta_2$ (μm)</td>
<td>27</td>
<td>26±0.5</td>
</tr>
<tr>
<td>0.84</td>
<td>0.9±1%</td>
<td>2.7</td>
<td>2.35±2.3%</td>
</tr>
<tr>
<td>DOF1 (μm)</td>
<td>DOF2 (μm)</td>
<td>N/A</td>
<td>55</td>
</tr>
<tr>
<td>6</td>
<td></td>
<td></td>
<td>32±0.5</td>
</tr>
</tbody>
</table>

Note that in Table 5, the resolution error in $z$ was calculated as $\Delta z/2$. The average FWHM and the relevant error, in percentage, were calculated using $\delta_i = 0.5 \times (\delta_{ix} + \delta_{iy})$ and $\Delta \delta_i = 50 \times (\delta_{ix} - \delta_{iy})/\delta_i$ respectively. The differences between the simulated and the experimental results are attributed to the fabrication artifacts such as imperfections in the milled cross/corrugations and the surface granularity, particularly inside the grooves. The use of experimentally obtained refractive index data for bulk silver of unknown surface roughness in simulating nanostructures that are modelled with perfectly smooth surfaces, also gives rise to discrepancies between the simulations and measurements[209].

### 8.5. Conclusion

This investigation confirmed that both the parallel and the normal components of the electric field emanating from a resonant cross aperture possess higher amplitudes along the film surface compared to those of a circular hole. Combining this with the strong SPP formation in the vicinity of the aperture created a resonance system that not only exhibits higher transmitted power but also improved the directionality compared to a resonant cross aperture in isolation. The role of the corrugations in our design is to further enhance the parallel components of the electric field that dominate the far-field along the optical axis of the BE, by combining the strong field generated by the resonant system and the coupling of the incident field to the LSPs inside the grooves. The resonant condition observed for our choice of the periodicity, is attributed to the complex interaction between the incident field, LSP oscillations inside the grooves and
the field emanating from the resonant system that led to a constructive interference of the \( x \)-component of the electric field in the far-field zone. To enhance the radiative decay rate slot antennas are the most efficient type of cavities to host a quantum emitter. That however poses a challenge and must be resolved in the future. In our measurements, 3D confocal beam profiling proved to be advantageous in producing a high resolution volumetric map of the field intensities. Results obtained experimentally agrees with the simulation. The high intensity in the central lobe in the far-field observed in our device proves to be an improvement to the previously reported nanostructures designed for beaming transmitted light [126, 265, 266].
9. Polarizing Bullseye Structure

Is it possible for a bullseye structure with concentric circular surface gratings to achieve a circularly polarized and collimated transmitted beam? In this chapter I report on the experimental findings concerning a BE structure with concentric circular corrugations surrounding an asymmetric cross-shaped aperture. Controlling the polarization state of a single photon source plays an important role in the quantum information as it provides a mechanism to define the computational basis states [4-7]. Single photon sources that emit one photon at a time may play a significant role in the future communication systems and quantum computing [2, 3]. However, due to their low rate of photon emission, detection of such light sources in isolation remains a major obstacle to the technological advances that they may bring about otherwise. Plasmonic nano-antennas may provide a solution to this dilemma. The ability to control the state of polarization and the radiation pattern as well as increase the radiation rate of quantum emitters is highly desirable for next generation telecommunications and quantum computing. Nanometric apertures perforated in metallic films exhibit LSPRs that depend on the shape and size of the aperture and the surrounding dielectric environment. It was previously shown computationally and experimentally that a high degree of circular polarization is achievable by detuning the two orthogonal LSP modes in an array of asymmetric cross-cavities [111, 112].

Figure 114: Design proposed by Gorodetski et. al. [113] that utilizes elliptical corrugations in their Bullseye structure to produce the transmitted CPL.
The utilization of surface plasmon polaritons and their coupling to the LSP modes in an elliptical Bullseye structure was also shown to produce the same effect [113], see Figure 114. However, this design does not satisfy certain criteria. The elliptical corrugations do not produce collimated beam and since the corrugations are tied up to the Circularly Polarized Light (CPL) functionality, one cannot alter/shape the output radiation pattern without impacting the degree of CPL produced. Here, a new plasmonic Bullseye structure is proposed that consists of an asymmetric cross-shaped aperture antenna surrounded by concentric circular corrugations to control the state of the polarization, as well as collimating the transmitted light. The reason for using cross-shaped aperture is to delegate the CPL functionality to the cavity and the shaping of the output beam to the corrugations.

9.1. Design, Fabrication and Characterization

Previously, it was shown numerically that an asymmetric cross-aperture with arm-lengths \( L_x = 150 \text{ nm} \) and \( L_y = 220 \text{ nm} \), perforated in a 100 nm tick silver film produced a high degree of circularly polarized transmitted light at \( \lambda_0 = 700 \text{ nm} \). Experimental results, however, showed a maximum degree of CPL of \( S_3 < 0.3 \) at \( \lambda_0 = 720 \text{ nm} \) [238]. The poor performance of the device was attributed to the mismatch between the SPP Bloch mode associated with the corrugations and the centre wavelength of the two orthogonal LSP modes corresponding to two arms of the asymmetric cross. For this exercise, the C4 configuration (see Appendix A) was re-fabricated with an asymmetric cross at its centre, here referred to as C7. Figure 115 shows the fabricated cross has arm-lengths \( L_x = 162.5 \text{ nm} \) and \( L_y = 200 \text{ nm}, \) with deviations \( \Delta L_y = -20 \text{ nm} \) and \( \Delta L_x = 12.5 \text{ nm} \), from the target arm-lengths.
Figure 115: Assymetric cross shaped aperture with target arm-lengths $L_x = 150$ nm and $L_y = 220$ nm.

![Figure 115](image1.png)

Figure 116: Experimental results: (a) A 2D slice in the y-z plane obtained from the volumetric map, (b) Normalized photon counts in the x-y plane at $z = 35$ µm, (c) beam intensities along the x and the y axis at $z = 35$ µm shows a symmetric profile.

The beam profile associated with the fabricated C7 configuration suggests a rotational symmetry about the z-axis despite the asymmetrical cross-shaped aperture. Transmitted stokes parameters through the C7 device were calculated by employing the technique described in [223] using a Nikon (Eclipse Ti - U) inverted microscope, Nikon 40x NA=0.6 objective, Nikon C-C Achromatic condenser operating in collimator mode, two linear polarizers (ThorLabs LPVIS050-MP) and a quarter waveplate (AQWP05M-600). The incandescent light was linearly polarized and collimated before incidence on the substrate. Transmitted light was collected by the objective from the air side, over an area that included the aperture and the corrugations. The light was then passed through the quarter wave plate and the second polarizer before being analysed by Andor Shamrock 303i-A spectrometer. The transmitted $S_3$ parameter for various incident
polarizations were calculated, Figure 117. At 45° incident polarization, $S_3$ parameter is maximized to $\sim0.7$ at $\lambda_0 = 520$ nm and drops to $\sim0.65$ at $\lambda_0 = 700$ nm. Although this is an improvement to our previous work [238], deviations, $\Delta L_y = -20$ nm and $\Delta L_x = 12.5$ nm, from the target arm-lengths had a negative impact on the C7 performance.

![Figure 117: Transmitted $S_3$ parameter for various incident polarizations.](image)

Note that transmission intensities were measured but not included. The only meaningful transmission would have been that normalized to the incident intensity. That, however, was not possible since we are dealing with extremely weak intensities transmitted through a subwavelength hole (and leakage through the film). The measurements were naturally carried out with the maximum incident intensity. Measuring the incident intensity implied removal of the sample, the polarizer, quarter waveplate and the analyser before the spectrometer, leading to saturating or even damaging the spectrometer. Reducing the intensity of the incandescent incident light by any means (including neutral density disk) changes the shape of the spectrum. The $S_3$ parameter on the other hand is normalized to $S_0$ due to the intrinsic nature of the measurement technique, which does not require removal of any optical element from the path.

### 9.2. Conclusion

Transmission response of an asymmetric cross-shaped aperture was numerically modelled previously, showing a high degree of circularly polarized light transmitted through the aperture. A bullseye structure was fabricated with the aperture integrated at its centre. Experimental results confirmed that it is possible to control both the directionality and polarization of the transmitted light. The Stokes parameter $S_3 \leq 0.7$,
however, is an indication of a poor performance with respect to the CPL functionality. This was partially caused by deviations in fabricated arm-lengths from those obtained during the design. Another factor responsible for the poor performance is the design process. The asymmetric cross was modelled in 3D and in isolation. Degree of CPL produced by an isolated cross may or may not be the same when integrated in a BE structure. To obtain the design parameters accurately, the cross must be simulated in the BE settings with the full set of corrugations intended for fabrication. Unfortunately, this required a full 3D model with an overall computation domain that is too large for the solution to converge before running out of memory. Note that unlike the symmetric cross, set in a BE structure, that may be accurately simulated using a 2D model or a quarter of a 3D model, simulating an asymmetric cross and calculating the relevant Stokes parameters may only be carried out in a full 3D model.
10. Travelling SPPs with Non-travelling Interference Envelope

The motivation behind this chapter was to answer a simple question: “what is the impact of film thickness on travelling surface wave?” This chapter answers the question via a series of simulations concerning surface waves launched by a single aperture perforating a silver film with various thicknesses.

Plasmonic aperture antenna arrays are based on planar designs that make them attractive for their simplicity and the ease of fabrication. In arrays of metallic nano-particles, the interaction between scatterers is limited to the near/far-field. Unlike the nano-particles, the interaction between apertures includes the exchange of power via the travelling/propagating surface modes. The influence of the film thickness and the substrate’s refractive index on the surface mode at the superstrate is an important study step that may help clearing some of the misunderstandings surrounding their propagation mechanism. A single sub-wavelength slit perforating a metallic thin film is amongst the simplest nanostructure capable of launching Surface Plasmon Polaritons (SPP) on its surrounding surface. Here, the influence of the substrate and the film thickness on the SPPs launched at the superstrate by a single sub-wavelength slit in a metallic film is investigated. When the thickness of the film is comparable to its skin depth, SPP waves penetrate the film. The penetration of SPPs from both sides of the film and their coupling inside the metal is nothing new. In a most relevant report to date (to a certain extent) Wang et. al. [267] modelled a free standing optically thin silver film in vacuum, where authors try to explain their findings in terms of long range SPPs, SPP Wave Packets and Quasi Cylindrical Waves (QCW) … etc. Prior to that, Verhagen et. al. showed that guided waves in a metal-dielectric-metal waveguide can penetrate the thin metallic cladding hence shortening the wavelength of the SPPs at the silver/air interface [268].

When the refractive index of the substrate differs from that of the superstrate, however, the superposition of the two waves from both sides of the film leads to travelling SPP waves modulated by a well define non-travelling interference envelope at
metal/dielectric interfaces. For a sufficiently thin metallic layer, SPPs formed at the metal/superstrate also interfere with those formed at the metal/substrate within the metal leading to a non-travelling periodic electric polarization inside the film. Note that this report is not concerned with the SPP eigenmodes [269-271], but rather it is an investigation on surface wave interference under the forced vibration.

10.1. Simulations, Results and Discussion

Fundamental equations governing the surface plasmons polaritons are stated here for convenience:

\[ k_{SPP} = \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon'_m + \varepsilon_d}} k \equiv k'_{SPP} + ik''_{SPP} \] (10.1)

\[ k_m \approx \sqrt{\frac{\varepsilon'_m + \varepsilon_d}{\varepsilon'_m + \varepsilon_d}} \left[ 1 + i \frac{\varepsilon'_m}{2\varepsilon'_m} \right] k \equiv k'_m + ik''_m \] (10.2)

Consider a metallic thin film with its surface set parallel to the x-y plane, equation (10.1) describes the complex wave vector for the SPP waves propagating at the metal/dielectric interface along the x-y plane, whereas the wave vector for the SPP waves penetrating the metallic film in the z direction is given by equation (10.2). In both equations the real part of the wave vector represents propagation constant, whereas the imaginary part defines the decay lengths, \(1/k_{SPP}\) and \(1/k_m\), over which the SPP’s amplitude decreases by \(1/e\). Note that in equation (10.2), the permittivity, \(\varepsilon_d\), corresponds to the dielectric material from which the field penetrates the film.

A 2D model of a 100 nm thick silver film perforated with a 50 nm wide slit was simulated using Finite Element Method (FEM) in COMSOL 4.3b with the model boundaries terminated with PML to eliminate reflections. The refractive index of the glass substrate supporting the film was initially set to \(n_1 = 1.52\) and the refractive index data for silver was taken from Palik [179]. The film was illuminated with a normally incident TM wave on the glass/silver interface. Figure 118(a) depicts the distribution of the real part of surface charge densities, \(\sigma(x,t) = |\sigma(x)|e^{i(k_{SPP} + \omega_0 t)}\), at an arbitrary time \(t_0\), calculated at both the air/silver and glass/silver interfaces. Note that only the real-part of the time-harmonic quantities are plotted in this chapter. The amplitude, i.e. the
envelope, of the surface charge density at the air/silver interface was calculated using

\[ |\sigma(x)| = \sqrt{\sigma(x,t) \sigma(x,t)} \]

and is depicted in Figure 118(b). The corresponding fast Fourier transforms, \( f[\sigma(x, t_0)] \) and \( f[|\sigma(x)|] \) were also calculated, see Figure 118(c)-(d).

**Figure 118:** (a) Surface charge density, \( \sigma(x,t_0) \), at an arbitrary time \( t_0 \), calculated at the air/silver and glass/silver interfaces. (b) The envelope, \( |\sigma(x)| \), at the air/silver interface. The corresponding fast Fourier transforms of (c) the wave \( f[\sigma(x, t_0)] \) and (d) the envelope \( f[|\sigma(x)|] \).

In Figure 118(b), the maximum accumulated charge density at the edge of the cavity, \( x_1 = 25 \) nm, is labelled \( C_{\text{max}} \). The decay length of the surface charge density, where the value of the \( C_{\text{max}} \) drop by \( 1/e \), was found to be \( \sim 10 \) nm from the edge (or 35 nm from the centre). At \( \lambda_0 = 700 \) nm, the decay length of an SPP along the silver/air interface is \( \sim 67 \mu m \) [8]. Activities near the slit, therefore, may not be considered as SPPs as they are highly localized. The inset of Figure 118(b), depict the \( |\sigma(x)| \) and \( \sigma(x,t) \) at \( t = t_0 + T/6 \) and \( t_0 + T/4 \). Here, \( T \) is the period and \( t_0 \) was set to a time when the surface charge density was at its maximum, \( C_{\text{max}} \), at \( x_1 \). The separation between the localized surface charges and the appearance of the harmonic wave occurs at \( t = t_0 + T/6 \) and \( x_2 \approx 75 \) nm, i.e. 50 nm away from the edge. In fact, the 10 nm decay length, closer to the \( 1/k_m \approx 25 \) nm obtained from equation (10.2), indicates that the surface charges in vicinity of the slit are due to the cavity modes, penetrating the metal and subsequently
decaying rapidly. This agrees to previous works [272, 273]. Furthermore, at \( t = t_0 + T/4 \) the surface charge density at \( x_1 \) drops to 0 and the peak at \( x_3 = 200 \) nm resembles that of a harmonic wave. The phase difference of \( 90^\circ \) between the oscillations at \( x_1 \) and \( x_3 \), resembles that of a forced vibration where the force leads the displacement by \( 90^\circ \) under resonance conditions [181]. However, the amplitude of the first peak at \( x_3 \) is \( 1.9 \times C_{0a} \), where \( C_{0a} \) is the DC component of \( |\sigma(x)| \), hence the average amplitude of the travelling SPP waves, Figure 118(d) and (a). By examining Figure 118(a) it was determined that the amplitude of the wave drops to \( C_{0a} \) at \( x_4 \approx w/2 + 2\times\lambda_{SPP} \), i.e. 2 wavelengths away from the edge of the slit. Although the surface charge density resembles that of a harmonic oscillation in the \( x_3 \leq x \leq x_4 \) range, its rapid decay and non-conformance to the \( 1/k_{SPP}^2 \), suggests a kind of transient state. To evaluate the \( \lambda_{SPP} \), fast the Fourier transform \( f[\sigma(x, t_0)] \) was calculated for both the silver/air and the silver/glass interfaces, Figure 118(c). The weighted average, \( K_{SPP} = \sum_{i=1}^{5} (K_{SPP}C_{SPP}) / C_{SPP} \), that included the centre mode and the four immediate neighbouring modes, i.e. two on each side of the maxima, provides a good estimate of SPP wavelengths on both surfaces. The SPP wavelengths were then calculated using \( \lambda_{SPP} = 1/K_{SPP} \), where \( K_{SPP} = \text{Re}(k_{SPP})/2\pi \). In summary, \( \lambda_a = 1/K_a = 667 \) nm and \( \lambda_g = 1/K_g = 427 \) nm are in agreement with \( \lambda_a = 682 \) nm and \( \lambda_g = 433 \) nm obtained analytically using equation (10.2). The difference between the simulated and the analytical values was attributed to the contribution of the electric fields associated with the QCW waves, \( E_{CW} = E - E_{SPP} [273] \), in the vicinity of the slit and is not the concern of this report. Fast Fourier transform of the envelope, \( f[|\sigma(x)|] \) in Figure 118(d), identified the DC components (or the amplitudes of the travelling SPP waves), \( C_{0a} \) and \( C_{0g} \) at both interfaces. The ratio \( C_{0a}/C_{0g} \) depends on the cavity’s dimension and the substrate’s refractive index.

Examining the \( |\sigma(x)| \), an additional second harmonic at both interfaces were observed. Although, the amplitude of the second harmonic modulation is weak, one must search for possible mechanisms responsible for oscillations at double the fundamental frequency. The boundary conditions were set to eliminate all reflections so it cannot be due to simulation artefacts, even more so that such second harmonics do not manifest themselves over the surface of a PEC that does not support SPPs. Frequency Domain in COMSOL solves the Maxwell’s equations for the spatial dependent time-harmonic
variable $E_x$, $E_y$ and $E_z$ simultaneously. All subsequent time-harmonic quantities are derived from these. One possible scenario that may lead to oscillation at double the fundamental frequency in \(|\sigma(x)|\), is that the normal-to-the-surface component of SPPs being modulated by the parallel-to-the-surface component at the interface via a relationship that involved multiplication. If indeed such faint second harmonic modulations are observable over an optically thick silver film in a real-life scenario, one may hypothesize on their origin. One possible explanation that could lead to such modulation may be attributed to normal-to-the-surface components of SPPs experiencing a perturbation caused by the parallel-to-the-surface components of SPPs that is analogous to the force $F \propto \sigma(x,t)E_x(z_0,t)$ on the surface. The electric force experienced by the SPP’s charge bundles at an arbitrary point along either interfaces, would then be proportional to $F \propto e^{-i2(k_x-k_g)\xi}$. Therefore, the force modulates the amplitude of the surface charge density wave over $T_0/2$, during which the SPP has travelled a total distance of $\lambda_g/2$. However, since COMSOL solve only for the spatial dependent of the fields, such second-harmonics are only manifested spatially in the solution. Note, however, that there won’t be any nonlinearities involved in the second harmonic mode as both the $\sigma$ and $E$ scale linearly with the incident power.

Regardless of the origin of such faint and uncontrollable modulation for optically thick films, it was envisaged that by reducing the film thickness, it would be possible for SPPs from the glass/silver interface penetrate the film and interfere with those at the air/silver interface. This would result in a series of minima/maxima in the charge density along the surface and inside the film with fixed loci that are $1/K_{beat}$ apart, where $K_{beat} = |K_a - K_g|$, hence by controlling the film thickness and the refractive index of the substrate, one could control the modulation strength and frequency of the envelope.

Keeping the superstrate and the substrate intact as before, two additional simulations, with $h = \{50, 25\}$ nm, were carried out in order to investigate the influence of the film thickness. In order to shift the $K_{a,beat}$ to overlap with the second harmonics, hence strengthening the second harmonic component of the envelope, the required value for the substrate’s refractive index at $\lambda_0 = 700$ nm was found to be 2.41 that corresponds to diamond [274]. With the recent advances in nano-diamond technology, use of diamond substrate is both feasible and practical [263]. Therefore, an additional simulation was also carried out with a 25 nm thick silver film supported on a diamond substrate. Figure
119 depict the calculated \( f[\sigma(x, t_0)] \) and \( f[|\sigma(x)|] \) from the simulation. Fast Fourier transforms of the propagating SPP waves, \( f[\sigma(x, t_0)] \), are shown in Figure 119(a), (c) and (e) with \( h = \{50, 25\} \) nm when the film is supported on a glass substrate and \( h = 25 \) nm with a diamond substrate respectively.

In all cases, \( K_a \) was found to be at the same position as it was for \( h = 100 \) nm. For \( h = \{50, 25\} \) nm on a glass substrate, \( K_g \) was also found to be at the exact location as it was for the 100 nm thick silver film. In the case of the diamond substrate, \( \lambda_g = 1/K_g = 230 \) nm, was found to be close the \( \lambda_g = 246 \) nm calculated using equation (10.1). In all cases, the appearance of an additional peak at the air/silver interface, positioned at \( K_g \) having an amplitude \( C_{\delta g} = C_g e^{-\frac{z}{\delta}} \), corresponded to the SPP waves that travel along the glass/silver interface penetrating the film and emerging at the air/silver interface. Fast

![Figure 119: \( f[\sigma(x, t_0)] \) and \( f[|\sigma(x)|] \) calculated for (a)-(b) \( h = 50 \) nm on glass substrate, (c)-(d) \( h = 25 \) nm on glass substrate and (e)-(f) \( h = 25 \) nm on diamond substrate. Note that subscript ‘g’ is used to label the substrate in general.](image)
Fourier transforms of the corresponding envelopes, \( f[|\sigma(x)|] \) in Figure 119(b), (d) and (f), show the anticipated modulating envelope with \( K_{\text{beat}} \equiv |K_a - K_g| \).

Figure 120 depicts the modulating envelopes, \( |\sigma(x)| \), calculated over the air/silver interface for \( h = \{100, 50, 25\} \) nm when the film is supported on a glass substrate and for \( h = 25 \) nm with a diamond substrate. The aperture was normally illuminated with a Gaussian beam, \( 15\times\lambda_0 \) in waist, from the substrate side. The surface of a perfect electric conductor that neither supports SPPs nor allows the penetration of the fields, produced only a smooth line, see Figure 120-(line in black). Values for the PEC line were calculated using \( \varepsilon_0 E_z \) to retain the C/m² unit. The inset in Figure 120 shows the travelling SPPs, \( \sigma(x,t) \), that are modulated by the envelope \( |\sigma(x)| \), calculated over the air/silver interface for the case \( h = 50 \) nm when excited with a plane wave from the glass substrate. The presence of the second harmonic and the beat interference in the envelope are marked.

![Figure 120: Surface charge densities, \( |\sigma(x)| \), over the air/silver surface for \( h = \{100, 50, 25\} \) on glass substrate, \( h = 25 \) nm on diamond substrate and for PEC.](image)

Another noticeable feature is the relation between the SPP’s decay length, \( 1/k_{\text{SPP}} \), along the air/silver interface and the strength of the interference envelope. Travelling SPP waves along the air/silver interface may be described as the superposition of two waves
\[
\sigma(x,t) = C_{0a}e^{i(k_a x - \omega t)} + C_{0g}e^{i(k_g x - \omega t)},
\]
where each component decay according to their
respective decay length $1/k_a$ and $1/k_g$. Analytical values for decay lengths were found to be $\{67, 17, 3.2\}$ μm for the air/silver, glass/silver and diamond/silver interfaces respectively. This explains the decay length of the envelope clearly. For example, in the case of the 25 nm silver film supported on a diamond substrate, the amplitude of the $C_{g} e^{i(k_g x - \omega t)}$ component drops to $1/e$ of its maximum at $x = 3.2$ μm, beyond which the only component that continues to propagate is $C_{a} e^{i(k_a x - \omega t)}$ due to its longer decay length of $\sim 67$ μm. And since the modulating envelope with $K_{a,\text{beat}}$ requires the presence of both components at the air/silver interface, the decay length of the envelope is dictated by the component having the shortest of the two decay lengths, which in this example is 3.2 μm associated with the $C_{g} e^{i(k_g x - \omega t)}$. Experimental measurements of such effects, however, may not be possible. In agreement with Wang et. al.\cite{267}, positioning any probe, such as an AFM tip, in the vicinity of the slit establishes standing wave oscillations between the tip and the slit, leading to a series of minima/maxima that convolve with those of the interference envelope. Having said that, measurements carried out by Verhagen et. al.\cite{268} cannot be flawed either.

With the film thinness comparable to the skin depth, charge bundles are no longer confined to the surface of the metal. The $x$-component of the travelling SPPs from the substrate and superstrate penetrate the film and interfere with one another. This leads to a periodic arrangement of the electric polarization density, $P_x(x)$ having a periodicity $1/K_{\text{beat}}$ (results for $P_x$ is not shown here). Snapshot of electric field $E_x$ passing through a periodic charge screen created by the envelope is depicted in Figure 121. Periodicities agree with the $1/K_{\text{beat}} \approx 1100$ nm and $1/K_{\text{beat}} \approx 340$ nm obtained from Figure 119(d) and (f) respectively.
Figure 121: Snapshot of electric field $E_x$ passing through a periodic charge screen (with periodicity $1/K_{\text{beat}}$) formed inside the 25 nm thick silver film for (a) glass and (b) diamond substrates. Note that $E_x$ was calculated at an arbitrary time with the maximum of its amplitude falling over the silver film, hence highlighting the periodic arrangement of the field inside the film.

Periodic field inside the film are due to the localization of the electric polarization density, $\mathbf{P}$ and vice versa. A series of simulations were carried out using a Gaussian beam to excite the 50 nm wide aperture perforated in a 25 nm silver film on diamond substrate. While maintain a constant incident power over the aperture, increasing the beam waist resulted in the formation of additional hotspots along the film with $1/K_{\text{beat}}$ periodicity (results not shown here). For the diamond substrate, diffraction patterns through a 25 nm silver film with single hole when excited with a normally incident beam (Gaussian in $x$) having a waist of $2\times\lambda_0$, is shown in Figure 122(a). When the maximum intensity of the Gaussian beam falls away from the centre of the slit (an arbitrary displacement of 680 nm in this case) the intensity of the transmitted beam exhibits a tilt towards the displacement, Figure 122(b). Such light-matter interaction is not observed in the transmitted beam through a 25 nm silver film on a diamond substrate with no aperture, see Figure 122(c).
Figure 122: $|E|^2 \times 10^2 \text{V/m}^2$ Diffraction patterns of a transmitted Gaussian beam through (a) 25 nm silver film perforated with a slit, supported on a diamond substrate. (b) same as (a) with the maximum intensity of the Gaussian beam displaced to $x = 680$ nm away from the centre of the slit. (c) In the absence of the slit.

10.2. Conclusion

In conclusion, it was shown that for a sufficiently thin silver film sandwiched between two different dielectrics, the mixing of the two SPPs (formed at the substrate and the superstrate) produce an interference envelope that not only modulates the travelling
SPPs but also shapes the electric polarization density inside the film. Such findings may play an important role in the future optical antenna designs. The following statement must not be taken as claims/conclusions related to the work in this chapter, however, the presence of charge bundles inside the film may imply changes to the electronic density of states, electron-electron collision (hence the mean free path), electron-lattice interaction (hence the electron’s effective mass) and consequently conductivity, due to the presence of an additional periodic potential that may compete with that of the positive ions. Such a problem is analogous to that of a superlattice in semiconductors [275, 276]. As for a future direction, a proper analytical/numerical solution must take into the consideration all possible interactions within the film, therefore, it may best be formulated in solid state/condensed matter physics. Previous work by Yan et. al. [277] or Herrera et. al. [278] is a good starting point.
**Finale**

In **chapter 4** it was shown theoretically that pair of metallic nano-rods or an asymmetric nano-cross can act as a wavelength demultiplexer.

In **chapter 5** most beneficial technique for fabricating subwavelength apertures was identified to be focused ion beam lithography. But more importantly it was noticed that when fabricating arrays of aperture, periodicity always reflected the target design dimension regardless of the technique while the aperture dimensions where more prone to errors. This lead to a series of investigations on arrays of cylindrical holes that are relatively easy to fabricate.

In **chapter 6**, the conditions in which off-normally incident light could have any impact on a particular SPP mode, were identified. This alleviated some of the concerns about depolarization associated with arrays that rely on surface modes (considering those relying on shape resonances). Also the impact of the dielectric constant surrounding the arrays of holes showed that the highest resonant transmission through an array of holes occurs in a symmetric setting where the dielectric constant of the substrate, the superstrate and the hole are the same, hence the need for and array in a homogeneous environment.

In **chapter 7**, it was shown that a bi-periodic array of cylindrical holes fabricated on a glass substrate could act as a wave plate, functioning purely based on surface modes. It was also shown that it is possible to convert this bi-periodic array (that was originally supported on a glass substrate) into a free-standing array, hence realizing an array in a homogeneous environment. The device functioned as a tuneable colour filter, a tuneable quarter wave plate and a refractive index based sensor with high throughput. While other authors invest on shape resonances to improve throughputs, achieving a 100% transmission observed at near infra-red in our device, I would consider it as underrated and requires more attentions, given its ease of fabrication and potential applications.

In **chapter 8**, we put to test the conventional claims about dimensions of plasmonic bullseye structures, showing that matching the period of the corrugations to the SPP
wavelength is not the optimum condition to reshape the radiation pattern of a quantum emitter into a collimated beam. A new design was proposed as one possible alternative. This outperformed bullseye designed based on the conventional approach proposed by others previously. With the new design being so different to the conventional ones one must raise concerns about what is being circulated as popular belief. In was also shown (in the same chapter) that is possible to harness the emitted power from a near-surface NV- in a diamond substrate using the new design. This deserves further investigations. In fact, a more elaborated work is being finalized using other designs that also differ from the conventional ones.

In chapter 9, it was experimentally demonstrated that is it possible to alter the polarization state of the transmitted light through an asymmetric cross-shaped aperture surrounded by concentric circular corrugations that produces focused beam. The degree of circularly polarized light, however, was no more than 0.7. To optimize the dimensions in order to achieve a perfect circularly polarized light, numerical modelling must encompass the entire device. This requires an enormous computational power when (for example) parametric sweeps are involved, and it was not possible during this project. Nevertheless, when and if such resources become available, future attempts in remodelling and fabrications are highly recommended.

In chapter 10, influence of the film thickness and the refractive index of substrate on travelling SPPs was investigated using FEM. Formation of periodic electric potential inside the film raises even more questions and I believe electrodynamics cannot answer all of them. Future studies must involve the condensed matter physics in examining electronic interactions inside film when periodic electric potential is form. I suspect there is new physics involved.
Appendix A - Other BE Devices

Apart from the C6, other BE configurations, with a resonant cross-shaped aperture incorporated, were also modelled and fabricated. Table 6 summarizes the dimensions for the first five configurations not reported in the previous section.

Table 6: Summary of target dimensions for the BE configurations

<table>
<thead>
<tr>
<th>Device</th>
<th>r1 (nm)</th>
<th>w1 (nm)</th>
<th>p (nm)</th>
<th>wp (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1</td>
<td>336</td>
<td>107</td>
<td>714</td>
<td>107</td>
</tr>
<tr>
<td>t = 95 nm</td>
<td>d = 50 nm</td>
<td>h = 50 nm</td>
<td></td>
<td></td>
</tr>
<tr>
<td>C2</td>
<td>720</td>
<td>490</td>
<td>650</td>
<td>140</td>
</tr>
<tr>
<td>C3</td>
<td>1110</td>
<td>876</td>
<td>500</td>
<td>200</td>
</tr>
<tr>
<td>C4</td>
<td>1090</td>
<td>600</td>
<td>525</td>
<td>187</td>
</tr>
<tr>
<td>C5</td>
<td>636</td>
<td>415</td>
<td>486</td>
<td>400</td>
</tr>
</tbody>
</table>

Figure 123(a)-(b) depicts the simulated far-field intensities, $|E_{\text{far}}|^2$ and $|E_{\text{far}}|^2 / |E_{\text{SH}}|^2$ as a function of angle, $\theta$, from the optical axis for C1-C6 and C_ref configurations. Here, $|E_{\text{SH}}|^2$ is the power transmitted through a single hole, 300 nm in diameter, perforated in a 300 nm thick silver film. In all cases, the simulation domain, the incident power and the number of corrugations ($N = 7$) were kept constant. Therefore, Figure 123(a)-(b) may be interpreted as the antenna efficiency and the gain respectively. C1 and C2 configurations have relatively low efficiency compared to other devices. C3-C6 exhibit between 4 to 5 times higher efficiency in the far-field zone and 4 to 5 time higher gain in the central lobe compared to that of the C_ref.
Fabrication and characterization process are described in the previous section. Table 7 summarizes the fabricated dimensions for each device.

**Table 7: Summary of fabricated dimensions for BE configurations, C1-C5**

<table>
<thead>
<tr>
<th>device</th>
<th>$r_1$ (nm)</th>
<th>$w_1$ (nm)</th>
<th>$p$ (nm)</th>
<th>$w_p$ (nm)</th>
<th>$L_x$ (nm)</th>
<th>$L_y$ (nm)</th>
<th>$f=(a-b)/a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1</td>
<td>325±13</td>
<td>106±2</td>
<td>722±8</td>
<td>105±2</td>
<td>175±2</td>
<td>175±2</td>
<td>0.015</td>
</tr>
<tr>
<td>C2</td>
<td>706±13</td>
<td>496±6</td>
<td>669±19</td>
<td>140±3</td>
<td>177±2</td>
<td>175±2</td>
<td>0.021</td>
</tr>
<tr>
<td>C3</td>
<td>1090±10</td>
<td>870±5</td>
<td>515±15</td>
<td>202±2</td>
<td>165±2</td>
<td>177±2</td>
<td>0.017</td>
</tr>
<tr>
<td>C4</td>
<td>1080±10</td>
<td>590±10</td>
<td>511±14</td>
<td>188±1</td>
<td>175±2</td>
<td>177±2</td>
<td>0.023</td>
</tr>
<tr>
<td>C5</td>
<td>650±15</td>
<td>414±5</td>
<td>513±28</td>
<td>415±15</td>
<td>178±2</td>
<td>172±2</td>
<td>0.021</td>
</tr>
</tbody>
</table>

Simulated $|E|^2$ and the measured intensity $I$, ($\Delta z = 0.1$ $\mu$m), associated with C1 configuration are depicted in Figure 125(a)-(b) respectively. Both the $|E|^2$ and the $I$ were normalized with respect to their maximum values. Figure 125(c) shows the intensity, $(I(x,y)-I_{\text{min}})/(I_{\text{max}}-I_{\text{min}})$, at $z = 0$ plane where $I_{\text{min}}$ is the background radiation. Notice how the field is localized near the surface (as expected) and the extent of the intensity at the central lobe is less than $1$ $\mu$m.
Figure 124: SEM image for the fabricated C1.

Figure 125: (a) the simulated $|E|^2$ and (b) the measured intensity, $I$, with $\Delta z = 0.1 \mu m$ and (c) the intensity, $I(x,y) - I_{\text{min}}/(I_{\text{max}} - I_{\text{min}})$ at $z = 0$ associated with C1 configuration.

Figure 127(a) and (b) shows the simulated $|E|^2$ and the measured $I$, ($\Delta z = 1 \mu m$), in the $x = 0$ plane for the configuration (C2). Whereas (c), show the normalized intensity at $z = 0$, i.e. the device plane. Despite the high intensity surrounding the aperture, the beam diverges rapidly with increasing $z$, Figure 127(d). The divergence was calculated by measuring the angle between the two intercepting lines drawn along the dark bands of the beam profile and was estimated to be 26°. This is in good agreement with the simulation, see Figure 127(e).
Figure 126: SEM image for the fabricated C2.

Figure 127: (a) Simulated and (b) measured ($\Delta z = 1 \, \mu m$) radiation patterns. (c) Normalized intensity at $z = 0$ plane. (d) Measured ($\Delta z = 0.1 \, \mu m$), and (e) close-up of the simulated radiation pattern for C2 configuration. Divergence angle was estimated to be $26^\circ$.

Simulated far-field intensity for C3 configuration indicated a strong central lobe. The full field calculation of the intensity, $|E|^2$, along the optical axis and in the $x$-$z$ plane are depicted in Figure 129(a) and (b) respectively. The FWHM of the simulated beam profile associated with the two distinct maxima at $z = 7.7 \, \mu m$ and $z = 36 \, \mu m$ were found to be approximately $\delta_1 = 0.88 \, \mu m$ and $\delta_2 = 2.74 \, \mu m$, see Figure 129(c) and (d). Corresponding experimental results are depicted in Figure 130. Maximization algorithm set the $z_{\text{ini}}$ to $\sim 5 \, \mu m$ away from the film’s surface. The normalized photon count along the optical path, Figure 130 (a), is obtained using $(I(z) - I_{\text{min}})/(I_{\text{max}} - I_{\text{min}})$, where $I_{\text{min}}$ is the background radiation. Beam profiles in the $x$-$y$ plane, Figure 130(c)-(d), represent the normalized $I(x,y)$ values at their maxima locations, $z = \{7.6 \, \mu m, 36 \, \mu m\}$. Corresponding FWHM were calculated to be $\delta \approx \{1.2 \, \mu m, 2.88 \, \mu m\}$ respectively and are in good agreement with the simulations.
Configurations C4-C5, were treated accordingly and the results are summarized in Table 8.
Figure 131: SEM image for the fabricated C4.

Figure 132: C4, simulations.

Figure 133: C4, experimental.
Figure 134: SEM image for the fabricated C5.

Figure 135: C5, simulations.

Figure 136: C5, experimental

Table 8: Locations of maxima and their corresponding FWHM of the beam for C3-C7 configurations.

<table>
<thead>
<tr>
<th>$z_1 , \mu m$</th>
<th>$\delta_1 , \mu m$</th>
<th>$z_2 , \mu m$</th>
<th>$\delta_2 , \mu m$</th>
</tr>
</thead>
</table>

171
The C3 configuration exhibits a slight broadening of the beam. The differences between the experiment and the simulated FWHM, $(\delta_{\text{exp}} - \delta_{\text{sim}})_{1,2} = \{0.37 \mu\text{m}, 0.14 \mu\text{m}\}$, are attributed to the granularity of the inner most corrugation, the position and the geometry of the cross, with respect to the corrugations, that fails to form a perfect concentric arrangement. The aperture dimensions also influences the SPP-LSP coupling and consequently the transmitted power. This can be demonstrated by comparing the C3 and C4 configurations. In the case of C4 configuration, the ratio $|E_1|^2/|E_2|^2 = 1.2$ is obtained from the simulation, where $|E_1|^2$ and $|E_2|^2$ are the field intensities at $z_1$ and $z_2$ respectively, see Figure 132. This value is very close to the experimentally measured ratio $I_1/I_2 = 1.6$, where $I_1$ and $I_2$ are the photon counts at $z_1$ and $z_2$ respectively, see Figure 133. On the other hand, the same ratios for the C3 configuration reads $|E_1|^2/|E_2|^2 = 20$ and $I_1/I_2 = 4.3$. Clearly, in the case of C3, the discrepancy between the measured and the simulated intensity ratios is due to the defect in the geometry of the aperture, i.e. the cross aperture is not fully milled through the film, compare SEM images in Figure 128(b)-(c) and Figure 131(b)-(c). A comparison between the fabricated C5 and C6 configurations, that are very similar in geometry, reveals the importance of the geometrical symmetry of the corrugations and their influence in shaping the beam profile. The differences $\{6.9\%, 6.3\%\}$ in the FWHM observed for C5, compared to $\{1\%, 2.3\%\}$ in C6, are attributed mainly to the inconsistency in the milled corrugations and partly to the presence of a debris near the aperture in C5, compare SEM images Figure 134 and Figure 110. Presence of the debris also results in dampening of the resonance system. The power ratio $I_{C6}/I_{C5} = 1.2$ calculated along the central lobes at $z_2$ is attributed to the dampening by the debris.
Appendix B – Nano-diamond: Preparation, characterization and integration steps

Figure 137: Step 1- Markers are milled on the surface of a silicon substrate using Focused Ion beam.
Figure 138: Step 2- Droplets of a solution of containing nan-diamonds are placed over the markers and let dry.

Figure 139: Step 3- Using a confocal microscope, nano-diamonds with nitrogen vacancies that exhibit anti-bouncing (i.e. Single Photon Emission) properties are located and identified.
Figure 140: Step 4- Anti-bunching properties of nano-diamonds are measured and recorded at various pomp powers prior to the integration with plasmonic devices.

Figure 141: Step 5- The micromanipulator instrument in an SEM machine may be used for integrating a nano-diamond with a plasmonic device using a technique called “pick and place”. Yellow circle shows a nano-diamond attached to the tip of the micromanipulator’s probe.
References


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