Near-field study of surface plasmonic lenses for nanofocusing applications

A thesis submitted for the degree of
Doctor of Philosophy

by

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“Let the future tell the truth, and evaluate each one according to his work and accomplishments. The present is theirs; the future, for which I have really worked, is mine”

Nikola Tesla
Abstract

Optical imaging has been utilised as a unique method for investigating specific properties of materials. However, the optical resolution is limited by the Rayleigh diffraction limit. This limit for visible wavelengths in typical media is of the order of 100-200 nm. One of the strategies to improve optical resolution is to decrease the incident wavelength so as to decrease the diffraction limited spot size. For example, the optical data storage industry has employed the UV wavelengths of light to record and read data, commercially known as the Blu-ray discs, while the visible wavelength regime is mostly preferred for many other practical applications. Another method to improve resolution, which has been prevalent since 2003 is by tailoring surface plasmons (SPs). SPs can have a much shorter wavelength than the incident photon and also they can have a higher effective refractive index in the propagating medium.

The unique properties of SPs lend themselves to the design of a circularly symmetric three dimensional nanoscopic structure which can focus the energy of a plasmon to a very small volume, much beyond the diffraction limit. This dissertation subsequently presents the optimisation, fabrication, theoretical and experimental measurement of a circular ring plasmonic lens, composed of a single annular slit and an inner concentric groove, which can achieve nanoscale focusing in the far-field regime from the lens surface.

This thesis goes on to discuss a plasmonic lens design which can beat the
dispersion issues associated with multiple wavelength focusing. We report
for the first time the nanoscale focusing of dual-wavelength of light onto the
same far-field focal plane. We have investigated the excitation efficiency of
SPs of a dual-wavelength far-field plasmonic lens under different polarisation
illumination conditions and demonstrate that this lens can be operated at
linear and radial excitation polarisation states. Our characterisation results
of the far-field plasmonic lens using out-of-plane sensitive scanning near-field
optical microscopic probe indicate the significant role the excitation beam
profiles play on the SP manipulation of the plasmonic lens, and thereby, on
the focusing action.

We have found that in order to have a proper SP excitation and for an efficient
nanoscale focusing of a far-field plasmonic lens, radial polarisation is preferred
over other polarisation states, due to the dominant longitudinal polarisation
component in the focal region for the former. The experimental results
demonstrate a good agreement with our theoretical predictions. This lens
could be beneficial for a broad community for multi-wavelength applications.
The ease with which it can be fabricated using a focused ion beam milling
method would make it even more useful for the nanophotonic community.

To improve the resolution of a dual-wavelength far-field plasmonic lens,
we introduce the concept of incorporating the far-field plasmonic lens into
a stimulated emission depletion (STED) system. This thesis investigates
theoretical possibilities of such a system, and draws a comparison between
a standard STED system and a dual-wavelength far-field plasmonic lens
based STED system. Theoretically, the effective point spread function of the
proposed plasmonic lens based STED system can reach up to $\lambda/6$. This can be
proven useful in multi-color super-resolution nanolithography and ultra high
density optical data storage.
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Priyamvada Venugopalan
Declaration

I, Priyamvada Venugopalan, declare that this thesis:

“Near-field study of surface plasmonic lenses for nanofocusing applications”

is my own work and has not been submitted previously, in whole or in part, in respect of any other academic award.

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Chapter 1

Introduction

Global internet usage has increased exponentially in recent years, with users reaching to approximately 250 million by the end of 2015. This dramatic increase is attributed to the revolution in photonics which is the study of light and its interaction with matter. Telecommunications is one of the exciting industrial applications of photonics capable of transmitting signals/data across kilometres through optical fibres. This optical revolution is due to the advent of light technologies such as lasers, fibre optics and numerous other photonic devices. The awarding of the Nobel prize to the innovation in transmitting light through optical fibres was a way of acknowledging its helpful discovery to mankind.

With the ever growing demand for improving photonic industry, light has become a building block to make things cheaper, faster and smaller. This occurred earlier, in relation to Moore’s law [1], which governed the scaling down of transistors resulting in the growth of semiconductor industry towards the road of device miniaturisation. This industrial drive has pushed all fields of engineering, resulting in the commencement of photonic nanotechnology, which in turn resulted in thin, energy efficient and highly compact solar
cells, increased storage capacity in optical disks, new medical diagnostics with resolution at virus level, etc. But all this growth in reality is being limited by the diffraction of light. Nano-optics, an active and evolving field, is the modern solution to overcoming this barrier of growth which can mitigate the trade off between speed brought by photonic components and size of typical electronic systems. It is to be emphasised that nano-photonics is purely application oriented using the fundamentals of science and is strongly beneficial to society.

To make discoveries in nanoscale, the challenge being faced by both engineering and science is to find an efficient way to couple electromagnetic radiation at optical frequencies to the nanoscale. A more recently discovered phenomenon, surface plasmons (SPs), provides a range of ways to meet the above mentioned challenge, the details of which are mentioned in Section 1.1. Potential applications of SPs in the photonics community can be found in Chapter 2. Plasmonics is a technology aiming at engineering material surfaces to control SPs. It focuses at understanding optical phenomena on the nanometer scale, i.e. near or beyond the diffraction limit of light. Figure 1.1 shows a graph of operating speed of devices versus their critical dimensions for different technologies [2]. Plasmonics has the capacity in improving the processing speed of future integrated circuits.
This chapter serves as an introduction to the exciting field of plasmonics. It presents a brief overview of the properties of SPs and a note on why they are important in many nano-photonic applications. The chapter concludes with the objectives and organisation of this thesis.

1.1 The diffraction limit of light

In 1873, the German physicist Ernst Abbe [3] realised that the resolution of optical imaging instruments, including telescopes and microscopes, is fundamentally limited by the diffraction of light. The smallest size to which light can be concentrated with standard optical elements such as lenses and mirrors is limited by the wavelength of light. This is because a plane wave incident on a circular lens or mirror is not focused to a point. While, it forms an Airy disk in the focal plane with the radius of central spot to first null of

\[ d = 1.22 \frac{\lambda}{NA} \]  

where, \( d \) is the minimum resolvable distance, \( \lambda \) is the wavelength of light and \( NA \) is the numerical aperture of the focusing lens.

His finding indicated that ultimately the resolution of an imaging instrument is not constrained by the quality of the instrument, but by the wavelength of light used and the aperture of its optics. Thus, the resolution of an imaging system can be improved with larger aperture of the lens and smaller incident wavelength of light. Recently, however, several new and exciting approaches in imaging have emerged that can break this limit of diffraction under certain circumstances, for example, by making use of light-matter interactions at a metal surface. An interface between a metal and a dielectric supports SPs [4], which are evanescent electromagnetic waves bound to the interface that are
strongly coupled to coherent oscillations of free charges at the metal surface. They can manipulate optical energy at very small dimensions [5, 6]. In recent years, the study of SPs has attracted much attention [7, 8], spurred by the possible applications of strongly confined optical fields [9] and enabled by advances in nanofabrication technology [10].

1.2 Plasmonics

Plasmonics or study of surface plasmon photonics, a field with important potential technological applications in sensing, imaging, and information processing, has been the subject of extensive research in recent decades. SPs were first observed by R W Wood in 1902 as some unaccounted features in his metallic diffraction grating experiments [11]. Later, Garnet observed colours in metal glasses and in metallic films [12] and in 1908, Mie proposed the theory of light scattering by spherical particles [13]. Indeed, Wood’s anomaly was not completely understood until the time when Fano [14] figured it out as the excitation of SP waves in 1941. However, since the pioneering work of Ritchie in 1957 [15], SPs have been scientifically recognised in the field of photonics. In this section an overview of the properties of SPs are provided and stipulate the reasons for considering it for sub-wavelength optics.

SPs can be defined as the oscillations of free electron density against the positive ions in a metal, when the latter are exposed to electromagnetic radiation. There is a dense assembly of negatively charged free electrons inside a conductor and also an equally charged positive ion lattice. Positive ions have an infinitely large mass compared to these free electrons and therefore, according to the Jellium model, a positive constant background can replace these ions with the total charge density inside the conductor remaining zero [16]. The incident electric field shifts the free electrons collectively with respect
Figure 1.2 (a) Schematic illustration of SPs propagating along a metal-dielectric interface. They are transverse magnetic in nature (H is in the y direction). (b) The exponential dependence of the electromagnetic field intensity on the distance away from the interface. The field in this perpendicular direction is said to be evanescent, reflecting the non-radiative nature of SPs. $\delta_d$ and $\delta_m$ are the penetration depths into the dielectric and metal respectively. From Barnes et al. [8]. Figure has been removed from the electronic version of the thesis due to copyright requirements and can be found in the original article.

to the positive charge of the lattice ions, resulting in a charge separation. This produces an attractive force and they move to the positive region. At the same time, the Coulomb repulsion between the accumulated electrons produces another restoring force in the opposite direction. The resultant of these two forces produces the longitudinal oscillations among the free electrons and is known as plasma oscillations. SPs are the quanta of oscillations at the metal surface or at the interface of the metal and the dielectric material (Figure 1.2(a)). An SP wave is an electromagnetic mode propagating along the surface which exponentially decays into each medium from the interface [17] as in Figure 1.2(b) [6], that is, they are evanescent in nature. Due to the exponential decay, the field is maximally close to the surface which can be confined to much smaller dimensions than free space wavelengths, with propagation lengths up to hundreds of micrometres [18].

1.2.1 The surface plasmon dispersion relation

From the dispersion relation of SP modes, i.e., the relation between the frequency $\omega$ and the wave vector, $k_{SP}$, much information about SPs can be
deduced. For light in free space, the wavevector, $k_0$ is given as $\frac{2\pi}{\lambda_0}$, where $\lambda_0$ is the wavelength of the incident light which excites SPs. In a quantum scenario, the momentum of the associated photon is $\frac{\hbar k_0}{2\pi}$ and the dispersion relation of a photon is $k_0 = \frac{\omega}{c}$, where $c$ is the speed of light in vacuum. In a medium of relative permittivity, $\varepsilon_d$, the dispersion relation of the same photon changes to $k = n_d k_0 = \sqrt{\varepsilon_d} k_0$. By employing Maxwell’s equation under appropriate boundary conditions, the dispersion relation between the frequency and the wavevector of SP modes [17] can be deduced as

$$k_{SP} = \frac{\omega}{c} \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}}$$

(1.2)

where $\varepsilon_m$ and $\varepsilon_d$ are the relative permittivities of the metal and the dielectric respectively.

For supporting SPs at the interface of metal and dielectric, $\varepsilon_m$ plays an important role. For the charges, which forms SPs, at the metal surface to be sustained, the electric field normal to the interface ($E_z$) must change sign across the interface. The displacement field defined as $D_z = \varepsilon E_z$ has to be conserved and therefore $\varepsilon_m$ and $\varepsilon_d$ must be of opposite sign in order for the interface to support SPs [7]. Since dielectrics have a positive permittivity, this condition is fulfilled by metals whose permittivity is real and negative [19].

The SP dispersion can be further modified by using a simple model for a metal, i.e., the Drude model for a free electron gas [20], in which the relative permittivity is given by

$$\varepsilon_m(\omega) = 1 - \frac{\omega_p^2}{\omega^2 - i\Gamma \omega}$$

(1.3)

where $\omega_p$ is the plasma frequency and $\Gamma$ is the scattering rate.
Figure 1.3 Dispersion curve of a SP wave, $k_{SP}$ and $k$ are the SP and free-space wavevectors, respectively. The wavevector of light is always less than the wavevector of SP for the same frequency, $\omega$. From Zhang et al. [6]. Figure has been removed from the electronic version of the thesis due to copyright requirements and can be found in the original article.

Substituting this in equation 1.2, SP dispersion relation can be plotted as in Figure 1.3. The curve shows the dispersion relation of a plasmon lies to the right of the light line, which means that SPs have a larger wave vector than light waves of same frequency in the dielectric medium. Also, from equation 1.2, we can see that the magnitude of the plasmon wavevector, $k_{SP}$ is always more than that of the photon in the dielectric medium bounding the metal, $\sqrt{\varepsilon_d}k_0$. To excite SPs, the wavevectors of two modes should be equal. Hence, light from dielectric materials cannot excite SPs directly. For exciting SPs, the momentum and hence the wavevector of the exciting light in dielectric medium should be increased using a coupling medium such as prism [21], grating [22] or surface corrugations or defects [23]. When the energy as well as the momentum of both is matched, there will be strong coupling between the incident light and SPs.

1.2.2 Properties of surface plasmons

The wavelength and the propagation length of SPs can be found from the real and complex dispersion relation by taking the real and imaginary parts respectively (from the complex relative permittivity of the metal, $\varepsilon_m = \varepsilon_m + i\varepsilon'_m$, the complex SP wave vector can be deduced as $k_{SP} = k_{SP} + ik'_{SP}$). From
the real part of the SP wavevector, the SP wavelength can be given as

$$\lambda_{SP} = \lambda_0 \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}} \quad (1.4)$$

where, $\lambda_{SP}$ and $\lambda_0$ are the surface plasmon and the incident wavelengths respectively.

The fact that the SP wavelength is less than the photon wavelength indicates the bound nature of SP modes. Also, it shows that if one wanted to use periodic structure to modulate SP, then the dimension of those structure should be of the order of the wavelength involved. SPs propagate along the metal surface but gradually attenuate owing to losses arising from absorption in the metal. The propagation length of SPs is therefore limited by the imaginary part of the complex plasmon wave vector $k_{SP}$ due to the internal damping. Based on the SP dispersion relation in equation 1.2, the propagation length of SP, $\delta_{SP}$ is given by

$$\delta_{SP} = \frac{1}{k_{SP}} \lambda_0 \frac{(\varepsilon_m)^2}{2\pi\varepsilon_m} \quad (1.5)$$

where $\delta_{SP}$ is the distance after which the SP intensity decreases to $1/e$ of its starting value.

The propagation length is usually dependent on the dielectric constant of the metal and the incident wavelength. As one would expect, for a long propagation length, a small real part of the permittivity and a large imaginary part are required. Silver is the metal with the lowest of the losses in the visible spectrum [6, 24, 25]. As a result of the interaction between the surface charges and the electromagnetic field, the field perpendicular to the surface decays exponentially with distance from the surface. This is in contrast to the propagating nature of SPs. The field in this perpendicular direction is said to
be evanescent in nature and is due to the bound nature of SPs which confines power close to the surface. This penetration of the fields into the materials bounding the interface, can be calculated from the wavevector of SP. Consider a medium with relative permittivity, \( \varepsilon_i \). The total wavevector of light of free space wavevector, \( k_0 \) is given by \( \varepsilon_i k_0^2 \). The relation between the SP wavevector propagating in z-direction and the total wavevector is

\[
\varepsilon_i k_0^2 = k_{\text{SPP}}^2 + k_{Z,i}^2
\]  

(1.6)

As noted above, the SP wavevector is always greater than the total wavevector, i.e., \( k_{\text{SPP}}^2 > \varepsilon_i k_0^2 \), which indicates the z component of the wavevector as always imaginary, indicating the exponential decay of the field with distance into the two media. By substituting equation 1.2 in equation 1.6, the penetration depths into the dielectric and metal are calculated as \[7\],

\[
\delta_d = \frac{1}{k_0} \left| \frac{\varepsilon_m + \varepsilon_d}{\varepsilon_d^2} \right|^{1/2}
\]  

(1.7)

\[
\delta_m = \frac{1}{k_0} \left| \frac{\varepsilon_m + \varepsilon_d}{\varepsilon_m^2} \right|^{1/2}
\]  

(1.8)

In the dielectric medium above the metal, typically air or glass, the penetration depth, is of the order of half the wavelength of light involved, whereas the penetration depth or decay length into the metal, is determined by the skin depth. The penetration depth into the metal is an indication of the thickness of metal films required in many application involving surface plasmons \[26, 27\]. In fact, associated with the field localisation near the metal, there is as well field enhancement \[28, 29\] due to an increase in the local photonic mode density \[30\].
1.2.3 Applications of surface plasmons

Owing to the unique features of SPs mentioned above, they have received much attention from physicists, chemists, and biologists. Most importantly, the electric fields of SPs are tightly confined to the surface of the metallic structure, to sizes much smaller than the wavelength of light, thereby beating the diffraction limit of light and creating a huge enhancement of the electric field at the surface. This has lead to much progress in the lab-on-chip scenario because of its potential to create sub-wavelength optical devices enabling the miniaturisation of optical components to size dimensions comparable to their electron counter parts. These have an important role in meeting the need for miniaturisation of laser elements. SPs can be focused on very small spots to the range of nanometers and proposed as plasmon laser sources at the deep sub-wavelength scale [31]. Light can be confined and guided along nanometric sized two dimensional metallic structures, which can be operated as SP waveguides [32–34]. Also, optical and infrared antennas based on metal nanostructures can be employed for highly enhanced and confined fields [35].

Another potential application of SPs is surface-enhanced Raman scattering (SERS) which results in electromagnetic enhancement due to the excitation of SPs [36]. Due to the local field enhancement of SPs, metal nanostructures can be used for probes for scanning near-field optical microscopes (SNOM) realising super-resolution or manipulation of photons beyond the diffraction limit [37]. Another application of SPs is in optical data storage [38, 39] using metal nano rods realising five-dimensional optical recording [40]. With the development and improvement of nanotechnology, plasmonics have shown fascinating prospects in optical interconnects [41], field-enhanced spectroscopy [42–44], microscopy [45], sensors [46] and solar cells [47, 48]. Moreover, SPs can be used for nanofocusing applications which in turn can result in diffraction unlimited imaging, nanolithography etc. This thesis is mainly focused on this
spectacular application of SPs which will be discussed in Chapters 2, 4 and 5 in detail. The increasingly wide availability of high precision fabrication and characterisation methods for sub-micron metallic structures allows further advances in the field of plasmonics [8] achieving high performance, high efficiency and high stability in nano-photonic devices.

1.3 Thesis objective

The main agenda of this thesis is to achieve nanofocusing in a variety of plasmonic nanostructures. Our aim is to realise and investigate far-field nanofocusing using a concentric metallic ring structure or plasmonic lens, under different polarisation illumination conditions.

The incident beam is focused using a low NA objective lens before being incident on the back focal plane of the plasmonic lens. Understanding the focused field components of a low NA objective lens is thus important for subsequent controlling of the efficient excitation of SPs of a plasmonic lens. With a high NA objective, the dominant field components at the focus are longitudinal in nature and have been extensively explored; however their properties with a low NA lens have not been achieved. To experimentally understand this, a SNOM system which can directly map the field distribution at the focus of the objective lens is employed. Since the dominant field distributions are transverse in nature, the near-field focusing properties of these beams are thoroughly investigated using an in-plane polarisation sensitive SNOM probe. This probe collects only the transverse polarisation fields with high spatial resolution resulting in three dimensional mapping of the transverse fields at the focus.

Plasmonic waveguides have as well been extensively used as nanofocusing structures and have exhibited confinement of energy guiding SP modes.
However, the polarisation distributions along the nanowire (NW) have not been extensively studied, and this forms another objective of this thesis. We employ the in-plane polarisation sensitive probe to investigate the polarisation properties of the plasmonic modes along a silver NW which is placed on a dielectric substrate.

Another novelty of this work is the design of a plasmonic lens which can realise dual-wavelength focusing. Theoretical simulations are carried out initially and we then scrutinise experiments to realise a plasmonic lens which can focus two wavelength of light to the same far-field focal plane. Furthermore, we investigate methods for improving the resolution of a dual-wavelength far-field plasmonic lens and investigating how it can be used effectively for nano-imaging applications. We propose the idea of incorporating stimulated emission depletion (STED) imaging principle with a plasmonic lens and our theoretical calculations and preliminary experimental results are also discussed in this thesis.

1.4 Organisation of the thesis

In this thesis, theoretical calculations and experimental results are discussed with the aim of understanding and demonstrating the nanofocusing of light using SPs in a variety of nanostructures. The strategies of these are outlined below.

Chapter 2

This thesis begins with the background information required for this work. A comprehensive overview of existing technologies in this field is given in Chapter 2. The exciting and useful properties of SPs and their different coupling schemes are briefly reviewed in Section 2.2. The typical concepts of plasmonic
lens as well as the polarisation properties along a plasmonic waveguide are also reviewed in this section. In Section 2.3, principles and applications of SNOM are particularly emphasised, as they are extensively used in the whole thesis for the characterisation of various plasmonic structures we deal with. A brief review of the existing super-resolution microscopic technique, stimulated emission depletion (STED) microscopy, is given in Section 2.4.

Chapter 3

This chapter will discuss the in-plane polarisation fields at the focus of a low NA objective lens and along a plasmonic waveguide. In this chapter, the three-dimensional mapping of the intensity distributions is done by a specific SNOM probe. Its properties and applications are dealt with in Section 3.2. The direct measurement of evanescent fields, which are transverse in nature at the focus of the objective lens under linear, radial, azimuthal and azimuthal first-order vortex (FOV) polarised illuminations, are undertaken in Section 3.3. Using an azimuthally polarised FOV incident beam, we are able to realise super-resolved pure-transverse focal fields. The detailed investigation of such in-plane polarisation fields at the focus is extremely important in the successful completion of this thesis. In Section 3.4, transverse polarisation components of a plasmonic NW are experimentally characterised, both in the far-field and in the near-field. In all these sections, experimental outcomes are compared with our theoretical findings.

Chapter 4

In this chapter, we introduce a nanoscale focusing device which can focus dual-wavelength of light to the same far-field focal plane. The nanoscale focusing performance of a dual-wavelength far-field plasmonic lens is demonstrated for the first time and is thoroughly investigated in Chapter 4. The quest for this device, which can efficiently focus light to nanoscale, began with its design
and simulation by Zhang et al. [49]. This nanoscale focusing device will be the primary focus of this thesis, from the design optimisation to its fabrication as discussed in Section 4.2. The characterisation of this device is done using SNOM under different polarisation illumination conditions. The improved SP excitation efficiency of a radially polarised beam generates a much brighter and tighter focal spot in the far-field focal plane of a plasmonic lens. Our investigation of the transverse polarisation fields at the focus of a low NA lens (Chapter 3) helps better understanding of the focusing action of a far-field plasmonic lens with different polarisation illuminations.

Chapter 5

To further improve the resolution achieved by a dual-wavelength far-field plasmonic lens, we propose the idea of dual-wavelength far-field plasmonic lens based STED systems. The preliminary theoretical calculations towards this work are presented in this chapter. This work demonstrates a novel approach for a dual-wavelength plasmonic lens in imaging and sensing applications.

Chapter 6

In the conclusion chapter, we provide a summary and discussion on the achievements of this thesis. In addition, we discuss the possibility of these research findings and provides some perspectives for future research.
Chapter 2

Literature review

2.1 Introduction

The recent rapid advancement of research in the field of surface plasmons (SPs) is due to the realisation that SP modes in metallic nanostructures may lead to localisation of light signals far beyond the diffraction limit of light. This allows visible and infrared light to be concentrated in regions as small as few nanometers. This leads to the idea of nanofocusing using SP modes. This chapter begins with a brief review on the nanofocusing applications of SPs. This is then followed by brief reviews on the near-field characterisation methods of SPs. Finally we review a prominent super-resolution technology, stimulated emission depletion microscopy (STED) and its principle.

2.2 Surface plasmons for nanofocusing applications

The resolution of a conventional optical system is limited to $\lambda/2$ due to the “Abbe’s diffraction limit”. Abbe’s diffraction limit arises because evanescent
waves which carry sub-wavelength information decay exponentially, leaving only the propagating waves that carry low spatial frequency components to be collected at the image plane [50]. The key point in imaging with resolution beyond the diffraction limit lies on the collection and detection of both propagating and evanescent waves radiated by the object in the near-field. A monochromatic plane wave $U$ in free space with a refractive index $n$ inside a two-dimensional system can be written as

$$U = U_0 \exp\left[i(k_xx + \gamma_zz - n^2\pi ct/\lambda_0)\right]$$

(2.1)

where, $\lambda_0$ and $c$ are the wavelength and speed of light in vacuum respectively; $k_x$ and $\gamma$ are the transverse and longitudinal wave numbers respectively.

For a plane wave satisfying Maxwell’s equations, transverse and longitudinal wave numbers must satisfy, $k_x^2 + \gamma^2 = n^2k_0^2$, with $k_0 = \frac{2\pi}{\lambda_0}$. Propagating plane waves are characterised by transverse wave numbers such that $|k_x| \leq nk_0$, letting $\gamma$ be a purely real number; on the other hand, evanescent waves are characterised by $|k_x| \geq nk_0$, which sets $\gamma$ as a purely imaginary number, leading to a decay of the amplitude along the $z$ axis. Thus they disappear at a large distance and cannot be detected by a normal optical microscope. By having larger spatial frequencies than propagating waves, evanescent waves carry fine details of the object with a resolution beyond the diffraction limit [51]. Pendry discussed the possibility of restoring these evanescent waves using the amplifying property of a slab of metamaterial which has a negative refractive index [26]. Superlens [52, 53], which can transport and enhance a large bandwidth of evanescent waves from the bottom to the top side of metamaterial slab is one of the nanofocusing technologies. Excitation of SPs at both interfaces of an optical super lens is the mechanism for the enhancement of evanescent waves. Different coupling schemes for exciting SPs
Chapter 2

by momentum matching are given in Section 2.2.1. Plasmonic lens which can achieve nanofocusing of light is another of its kind and is discussed in Section 2.2.2. To achieve efficient on-chip nanofocusing, various plasmonic structures like metallic nano waveguides, tapered metallic structures etc. have been developed both theoretically and experimentally.

2.2.1 Coupling to surface plasmons

According to the SP dispersion relation in equation 1.2, the interaction between the incident electromagnetic field and surface charges results in an increase in the momentum of SPs compared to the incident wavevector. This give rise to a momentum mismatch between the two. As discussed in Section 1.2.1, only when the momentum of SPs and the incident light are matched and equal, a coupling happens. And it can be matched using different coupler configurations such as prism couplers, grating couplers, fibre and waveguide couplers.

2.2.1.1 Prism coupling

This configuration uses the phenomena of total internal reflection in a high index prism which is brought close to a metal interface. Considering light propagating in a dielectric and be incident on a metal interface at an angle $\theta_j$, a wave surface charges will be induced with a wavevector, $n_\text{dielectric} \sin(\theta_j)$, where $n$ is the refractive index of the dielectric. For all $\theta$, this wavevector is less than $n(\frac{\omega}{c})$. As described in Section 1.2.1, the wavevector of a SP is greater than $n(\frac{\omega}{c})$ which prevents SPs from being excited. Otto [54] introduced the concept of having a spacer layer with refractive index, $n_s$ such that, $n_p > n_s$ between the metal and the prism. Hence the component of the momentum along the interface between the media can be increased as,

$$k = n_p \sin(\theta_j) \left(\frac{\omega}{c}\right)$$ (2.2)
If the angle of incidence, $\theta$, at the prism/air interface is greater than critical angle, such that only evanescent field penetrates into the spacer layer, the electromagnetic field will be able to couple to the SPs at the metal interface if the increased momentum at the interface is equal to $k_{SP}$. This is an attenuated total reflection and is utilised in the Otto configuration as shown in Figure 2.1(a).

Another configuration is the Kretschmann configuration [21] which involves evaporating a thin metal film directly on to the prism surface. Here, the low index gap is provided by the metal itself. The evanescent waves pass through the metal film and excites SPs on the bottom surface of the metal. This gives rise to the tunnelling of evanescent fields through the metal film and as a result excites SPs on the bottom surface of the metal, i.e., in this case at the metal/air interface as shown in Figure 2.1(b).

### 2.2.1.2 Grating coupling

The mismatch between the in-plane momentum wavevectors of incident photon and SP can also be overcome by diffraction gratings patterned on the metal
Grating coupling to SPs using diffraction ($k_{ph}$: wave vector of the incident light, $m$: diffraction order).

When a beam of light is incident at an angle, $\theta$ on to a grating surface, different diffraction orders will be scattered from it, increasing or decreasing the component of its wavevector by integer multiples of grating wavevector, $k_g = \frac{2\pi}{\Lambda}$ (Figure 2.2). When a diffracted order has a wavevector greater than that of the incident radiation, it may couple to the SPs according to the coupling condition,

$$k_{SP} = nK_{ph}sin\theta \pm mk_g$$

where $K_{ph}$ is the wave vector of the incident light, $m$ is an integer and $n$ is the refractive index of the medium.

### 2.2.2 Plasmonic lens

Plasmonic lenses, consisting of metal and dielectric materials have been introduced for overcoming the diffraction barrier by employing evanescent waves focusing in the near-field region of an object, that carry the finest information from the latter. In this section, a literature review on the physics behind the principle of plasmonic lenses is given. The main focus is on typical concepts of plasmonic lens reported so far. Superlens and hyperlens are
examples of plasmonic lens on the basis of dielectric/Ag/dielectric sandwiched structures [55, 56]. However for a superlens, the range of practical applications are limited by the complicated fabrication methods, absence of working distance and amplification, along with its nature of one dimensional imaging.

2.2.2.1 One-dimensional focusing of surface plasmons

Owing to the advent of the phenomena of extraordinary optical transmission through sub-wavelength metallic aperture array, there has been much interest in metallic sub-wavelength structures [57]. It has been observed that surface plasmons (SPs) excited in a corrugated metallic film can result in a highly directed beaming with a low divergence [58]. Based on this idea, new types of methods have been implemented which can bring modulation in light phase resulting in beam deflection and focusing if appropriately designed [59].

For achieving one dimensional focusing, two types of tuning methods for phase modulation can be employed: either depth tuned [60] or width tuned [61] structures. In a width tuning method, an incident light beam is manipulated by modulating light phase through a metallic film with arrayed nano slits, which have a varying width but constant depth. The sub-wavelength slits provide enough phase retardations in addition to the extra ordinary optical transmission of SPs. The experimental demonstration of planar lenses based on nanoscale slit arrays in a metallic film (Figure 2.3) is done by Verslegers et al. [62]. Based on the depth tuning method, efficient plasmonic focusing is achieved by three types of plasmonic slits (convex, concave and flat/constant groove depth). When a TM polarised plane wave impinges on the slit, it excites SPs which propagate along the surface and is diffracted to the far-field by the periodic grooves [63]. The focusing effect happens due to the constructive interference of all the diffracted beams at a certain point on the axis [64]. With this method, a tailored ultra compact lens is achieved, by tuning the
parameters of the groove such as its width, depth, period etc.

Nanometric structures with a set of split curved slit has been introduced that can produce SP vortices with arbitrary vortex topological charges [65]. The SP vortex refers to an optical vortex of plasmonic waves with a dark spot and phase singularity at its center and is of high significance due to the strong optical angular momentum in the evanescent region. Such plasmonic vortex lens have applications in nanoscale microscopy, optical data storage, and quantum computing as well as particle manipulation. It has as well been demonstrated that two dimensional arrays of plasmonic nanoholes can function as broadband microlenses where patches of nanoholes can focus single wavelength of light across the entire visible spectrum [66]. This broadband focusing ability opens applications in multicolor stereo imaging, broadband light collection, and multichannel optical communication.

**Plasmonic waveguides**

Plasmonic waveguide is another key one-dimensional element for the plasmonic nanofocusing applications which can localise guided light signals beyond the
Figure 2.4 Guided modes: dielectric fibres versus metal NWs. Typical field structures, localisation and wavelengths of the fundamental modes guided by (a) dielectric fibres and (b) cylindrical metal NWs for different core diameters. $\lambda_0$ and $\lambda_{0SP}$ are the mode wavelengths for infinite diameter fibres or metal NWs, respectively. The dashed horizontal lines show the localisation of the mode at the $1/e$ level of the field. From Gramotnev et al. Figure has been removed from the electronic version of the thesis due to copyright requirements and can be found in the original article. [72]. Figure has been removed from the electronic version of the thesis due to copyright requirements and can be found in the original article.

diffraction limit for electromagnetic waves [67, 68]. This results in concentrating visible and infrared light to regions as small as a few nanometers with relatively low loss for Au and Ag nanowires (NWs) [69]. The electromagnetic modes guided by dielectric fibres cannot result in a sub-wavelength localisation. As the diameter of the dielectric fibre is decreased, the number of guided modes along the fibre is reduced, leaving only the fundamental mode which penetrates into the surrounding medium. The wavelength of this mode decreases and becomes equal to that of the surrounding medium. And as a result, the mode size decreases when the fibre diameter decreases and then increases to infinity. This feature indicates the diffraction limit of light in dielectric waveguides/optical fibre. This forms the major drawback in achieving miniaturised optical devices and interconnects. While, the phenomenon happening in a metallic waveguide is the opposite. As the diameter of a metal NW decreases, the fundamental mode experiences an increase in localisation. And as a result the diameter of the guided mode can be decreased to just a few nanometers achieving sub-wavelength localisation [70, 71]. Comparison between the guided modes in dielectric fibres and metal NWs are shown in Figure 2.4.
Different nanostructures have been investigated as waveguides. Chemically synthesised crystalline Ag NWs can support propagating SPs with lower losses than lithographically defined NW waveguides, and can be easily manipulated to construct complex optical devices, which makes them ideal candidates for proof-of-principle studies of plasmonic circuits. Finite length Ag NWs are intriguing plasmonic structures due to their strong polarisibility compared with the spherical shaped structures and a low loss among the metallic NWs [73]. These kinds of anisotropic extended Ag nanostructures with sub-wavelength dimensions can support propagating SPs along the NW with a lateral confinement in the other two directions and with a micro-scale propagation length thereby acting as sub-wavelength plasmonic waveguides [74]. In addition, controlling the order and the polarisation properties of the plasmonic modes in free-standing Ag NWs has enabled a wide range of applications such as beam splitters, polarisation rotators, bio-sensors and so on [75–78]. Several groups have investigated SP modes along Ag NWs supported on a silica or silicon substrate, but mostly with a certain gap between the NW and the substrate [79]. In contrast to well-studied free-standing NWs, waveguiding properties of substrate-supported NWs have not been adequately investigated.

2.2.2.2 Two-dimensional focusing of surface plasmons

Circular grating based metallic structures for generation and focusing of SP waves by directional excitation were reported by Zhang et al. in 2005 [49] resulting in a complete harvesting of electromagnetic energy from SPs. Zhang’s plasmonic lens consists of a single annular sub wavelength ring milled into thin metallic substrate, width of it being smaller than half the wavelength of incident light. This elliptical or circular ring can excite as well as focus the electromagnetic energy of SPs to one single focal spot. The wavelength of SPs
depend on the wavelength of the incident photon and the dielectric functions of the metal and the surrounding medium. The direction of the wave vector of excited SPs will be normal to the slit and consequently, the energy will be directed towards the center of the plasmonic lens. This will result in a high intensity focusing at the centre of the structure based on the constructive interference of excited SPs. Since then, many plasmonic structures [61, 80] have been introduced to break the diffraction barrier of light and to achieve efficient nanofocusing.

A novel structure called plasmonic micro-zone plate or plasmonic lens with chirped slits is proposed [81, 82]. PMZP is an asymmetric structure with variant periods fabricated on a thin Ag film Figure 2.5. The focusing action is by the interference between the cavity mode-coupled SP waves and the diffraction wavelets passing through the rings. The final intensity at the focus point is obtained by iteration of each zone focusing and interference with each other and it can be expressed as [83]

\[
I = \alpha \sum C I_{i0} I_{SP} \frac{4r_i}{\kappa_{SP}} exp\left(-\frac{r_i}{l_{SP}}\right)
\]

(2.4)

where \(I_{i0}\) is the intensity of diffractive wavelet at the \(i^{th}\) zone, \(I_{SP}\) is the intensity of SPs passing through the \(i^{th}\) slit, \(r_i\) is the radius of each zone, \(i\) is the number of zones, \(l_{SP}\) is the propagation length for the SP wave, \(\alpha\) is the interference factor and \(C\) is the coupling efficiency of slits.

A hybrid Au-Ag sub-wavelength metallic zone plate like structure was proposed for preventing oxidation and sulfuration of Ag film along with achieving superfocusing [83]. Focusing action of such a structure is characterised using FDTD numerical calculations. It shows that thickness of Au and Ag films
Figure 2.5  Schematic of the plasmonic microzone plate for super-focusing. From Fu et al. [81]. Figure has been removed from the electronic version of the thesis due to copyright requirements and can be found in the original article.

has significant effect due to their immense contribution to superfocusing and transmission. This hybrid structure has potential applications in data storage, nanolithography, bio-imaging etc. However, these ring-based structures have stronger sidelobes. A circular hole based plasmonic lens was reported so as to suppress side lobes [84]. With a reduced periodicity of the nanoholes, the focusing action is obtained by the propagation waves, due to SP wave coupling and the diffraction wavelets from the pinholes. The proposed design has the advantages of micron scale propagation length, longer depth of focus and easier fabrication techniques. An elliptical nanohole based plasmonic lens [85] is put forth to further improve the focusing performance of the circular hole based plasmonic lens. A thin film of Ag deposited on a glass substrate is perforated by elliptical pinholes with different sizes, distributed in different rings with variant periods. This structure could realise a long focal length in free space with a depth of focus of around 13 μm under linearly polarised plane wave illumination.

The nanofocusing performance of a circular ring based plasmonic lens was experimentally demonstrated using radial polarised illumination by Lerman et al. [86, 87]. A sharp focal spot corresponding to zero order Bessel function has been observed compared to the two separated lobes using linear polarised illumination. Using scanning near-field optical microscopy (SNOM), the FWHM of the focal spot is measured to be 0.38 λ₀± 0.04 nm compared to the theoretical value of 0.36 λ₀.
It is known that, as a TM polarised (transverse-magnetic or magnetic field perpendicular to the plane of incidence) monochromatic plane wave is incident on the circular ring, it causes the excitation of SPs which propagate along the surface of the metal film and undergoes constructive interference. A tightly focused radially polarised light is TM polarised with respect to this boundary, thus providing cylindrical symmetric focusing. It is theoretically shown that, by illuminating a plasmonic lens with radially polarised light the out of plane electric field component interferes constructively at the optical axis of the plasmonic lens, thus forming a sharp focal spot compared to the two separated lobes achieved by linear polarisation. This can be explained by considering the in-plane \( (E_R) \) and out-of-plane \( (E_Z) \) field components as shown in Figure 2.6. When the structure is illuminated with radially polarised light, the \( E_R \) field components diminishes at the focus due to the destructive interference. This is attributed to the SP waves originating in antiphase at two opposite points on the circumference of the structure, due to the radial distribution. However, since the longitudinal field components of SPs always point in z-direction, they undergo constructive interference producing a sharp focal spot. In case of a linear incident polarisation, the \( E_R \) field components undergo constructive interference while the dominant \( E_Z \) component destructively interfere, producing two focal lobes. This results demonstrate the advantage of using radially polarised light for nanofocusing applications involving SPs.
A plasmonic leak free lens is proposed consisting of two asymmetric concentric ring slits which can enhance the field intensity at the center of the structure while suppressing the field leaking in the counter-focus direction [88]. A spiral plasmonic lens has been experimentally demonstrated by Chen et al. [89] which can differently focus light with different handedness resulting in miniature circular polarisation analyser (Figure 2.7). A circular plasmonic lens has been employed to detect orbital angular momentum of light [90] through division-of-amplitude interference.

The main limitation of a conventional optical lens is that it suffers from the diffraction limit because of its inability to collect evanescent waves in the far-field region. Plasmonic lens is a solution to this problem. The concept of negative refractive index (satisfied by metals in specific frequency) and the enhancement of evanescent waves are fulfilled by metal based plasmonic lenses. The ability to achieve nanofocusing beyond the diffraction limit opens up many applications as reported [91, 92]. Plasmonic lens structures can be also used as beam splitters [93] and for beam shaping [94]. With the growing innovation in the design and fabrication [95] of plasmonic lens, more extensive applications can be found in the very near future.
2.2.2.3 Three-dimensional focusing of surface plasmons

Plasmonic waveguides, as explained in sub-section 2.2.2.1, can focus surface plasmons one dimensionally. By gradually reducing the thickness of such waveguide geometries along the direction of propagation, SP modes they support are predicted to become more strongly confined and concentrated to very small dimensions [70]. Waveguides with tapering ends have widely been used for three-dimensional nanofocusing applications [72, 96, 97]. SPs propagating toward the tip of a tapered plasmonic waveguide are slowed down and asymptotically stopped when they tend to the tip, never actually reaching it. This phenomenon causes accumulation of energy and giant local fields at the tip as shown in Figure 2.8[45]. Three-dimensional nanofocusing of SPs with patterned gold and silver pyramids have been demonstrated that can generate well-defined plasmonic hot spots even with linearly polarised illumination unlike tapered waveguides [98]. These structures may have clear applications in optical trapping, high density optical data storage etc.

A three-dimensional plasmonic dimple structure have been demonstrated for achieving nanofocusing on the vertical edge of a chip based on focusing double sided plasmons using a tapered metal-insulator-metal structure [99, 100]. Three-dimensional nanofocusing was also realised in a tapered hollow metal waveguide with an elliptical cross section that can squeeze near-infrared radiation down to ~100 nm along the minor axis of the ellipse [101]. The rapid adiabatic nanofocusing promises to find various applications in nano-optics and nanotechnology where greatly enhanced local optical fields are required, in particular, for probing, spectroscopy and detection.
Figure 2.8  A typical distribution of $|E|^2$ along a tapered section of a metal film or a tapered metal ridge. The top inset shows a typical cross-sectional distribution of the field near the tip of a metal film on a dielectric substrate. The bottom inset shows an expanded view of the area near the structural tip. From Gramotnev et. al. Figure has been removed from the electronic version of the thesis due to copyright requirements and can be found in the original article. [101].

2.3 Characterisation of the surface plasmon modes

Now the concern is how to investigate experimentally plasmonic properties/modes which exist very close to the surface under study. A conventional far-field microscope cannot be employed in such a scenario, since to measure sub-wavelength features a device with sub-wavelength resolution is required. Microscopes with high temporal and spatial resolution are required which provide us with eyes for the nanoworld. And therefore, a sub-wavelength probe which can be brought close to the near-field, along the surface which the SP propagates, and can collect the near-field information to the far-field is a necessity. New types of high resolution microscopes have been developed for a better imaging of nanoscopic features; the electron microscope in transmission or reflection (TEM or REM), the atomic force or scanning tunnel microscope (AFM or STM) [102]. These high resolution microscopes are scanning probe microscopes [103], which indicate that the property of a sample is measured by
a probe which is scanned over the sample with nanometer precision [104]. The first scanning probe technique which was employed for studying SP properties was STM [105, 106] which relies on the collection of far-field signals scattered due to the local interaction with an STM tip. Later on, a dielectric SiN probe of an AFM was employed [107]. But these probes prevent direct measurements of the local SP field on a surface due to perturbations induced by the metal/silicon tip [108]. Although, STM and AFM can achieve high spatial resolution and can provide information on the atomic composition of objects at the nanometer scale, optical properties were yet not available at the nanometer scale. This can only be achieved by investigating the near-field signal of the surface under study.

**Scanning near-field optical microscopy**

The concept of SNOM was first proposed by an Irish scientist E. Synge in 1928 [109]. He described an experimental scheme in which the diffraction limit could be broken using a sub-wavelength aperture which has to be in the proximity of the surface as in Figure 2.9(a). The aperture acts as nano-source which re-emits the electric field confined close to the surface. The first experimental proof of the validity of SNOM concept was given by Ash and Nichols in 1972 using microwaves [110]. Lewis et al. [111] and Pohl et al. [112] made the first near-field microscopes which can work in visible light. There are two different SNOM configuration: apertureless SNOM [113] and aperture SNOM [114], to detect the highly decaying evanescent waves. In apertureless SNOM, the probe is a nanometric metallic hole or a sharp metallic tip which is used as a scatter source. It is placed very close to the sample surface where it converts evanescent waves in the near-field into propagating waves in the far-field, by scattering. One of the main challenges in apertureless SNOM is the
strong background signal since a large area of the sample around the tip is also illuminated.

In aperture SNOM, the probe is a tapered optical fibre, the end of which is a small sub-wavelength aperture. In 1991, Betzig et al. [114] introduced the SNOM probe made from a single mode optical fibre, one end of which is tapered to a tip size of 50 nm and was coated with aluminium. The tip-sample distance is kept within a few nanometers and is controlled using shear-force microscopy by a distance regulation method [115]. In this way, the fibre tip is laterally vibrated, amplitude of which is constantly monitored. As the tip-sample distance decreases, the amplitude of the vibration dampens which sends out necessary signal to the control system so that the tip is pulled back from the surface and maintains the constant distance. During scanning, the fibre tip has to be maintained at a constant and small distance of the order of nanometers from the surface of the sample. And signal is collected only from a very small surface of the sample under the aperture. As the fibre probe is scanned across the sample, the signal is recorded point by point using a suitable photodetector. Thus, SNOM gives images with a resolution beyond the diffraction limit.

A typical resolution of an apertureless SNOM is about 5-30 nm, while the resolution of an aperture SNOM is 50-150 nm [116]. The reduced resolution of the latter is due to the fact that as the aperture diameter is reduced, the number of photons collected are also reduced which in turn reduce the signal to noise ratio. This results in a conflicting limit on the resolution improvement that can be achieved by the further reduction of the aperture diameter [103].

There are three main SNOM operation modes [103], two of which are shown in Figure 2.10.
Figure 2.9  (a) Synge’s representation of SNOM by an opaque screen with a sub-wavelength hole brought in the near-field zone. (b) Actual SNOM configuration: feedbacked nanoscale probe to study the sample in the near-field zone. From Lereu et al. [116]. Figure has been removed from the electronic version of the thesis due to copyright requirements and can be found in the original article.

Figure 2.10  Basic aperture SNOM scheme. (a) Collection mode (b) Illumination mode [103].
Figure 2.11 Schematic of a SNOM experimental set-up for studying SPs [117].

**Collection mode:** The sample is illuminated from the far-field. The evanescent waves generated in such a way at the sample surface are converted by the aperture into propagating waves, and conducted through the fibre to a suitable detector.

**Illumination mode:** Light is coupled into the flat end of the fibre and transmitted to the tapered end where it is squeezed towards the aperture. The sub-wavelength aperture converts the propagating waves into evanescent waves. These waves interact with fine features of the sample which convert evanescent waves into propagating wave to be detected in the far-field.

**Illumination-collection:** Aperture is used both as the light source and as the collector. This mode use a beam splitter which can separate light directed to the sample and light coming back from the sample.

For photonics/plasmonics applications the aperture probe in collection mode
is generally preferred. Since the first demonstration of SNOM imaging of evanescent fields of waveguide modes [118], SNOM has been used in a number of plasmonic mode studies. The scheme of a SNOM set up for studying SPs is shown in Figure 2.11. There has been a significant progress in the image formation in the collection SNOM with the advancement in SNOM instrumentation [119].

2.4 Stimulated emission depletion microscopy

2.4.1 Background

The introduction of confocal laser scanning microscopy which enabled real 3D imaging [120] was an important milestone in the field of microscopic imaging. The diffraction limit of light was the major obstacle of this traditional microscopic method. This resolution limit has been overcome by a number of diffraction unlimited optical imaging techniques, which can be categorised in two different groups [121]; stochastic and non-stochastic super-resolution microscopy. In the stochastic approach, the individual molecules of fluorescence emitters are randomly switched in space and is exploited to determine their position. To achieve this, several molecular switching methods like PALM (photoactivation localisation microscopy) [122], STORM (stochastic optical reconstruction microscopy) [123], dSTORM (direct stochastic optical reconstruction microscopy) [124] etc. have been developed. While, the non-stochastic techniques are based on point spread function (PSF) engineering. By this, the coordinates in the sample are targeted to actively define the areas in the sample where the fluorescence emitters must be on or off. SIM (structured illumination) [125], RESOLFT (reversible saturable
optical fluorescence transitions) [126], GSD (ground state depletion) [127], STED (stimulated emission depletion) [128] etc. are based on this technique.

2.4.2 The stimulated emission depletion principle

The basic idea behind STED is the selective deactivation of the fluorescence emitters. The concept of STED microscopy was first proposed in 1994 and experimentally demonstrated in 1999 [129, 130]. Today, it is a prominent and versatile form of super-resolution light microscopy. STED microscopic technique uses two overlapping laser beams; one is the excitation laser which excites and the second is the depletion laser which depletes the excitation of the sample. Fluorescence occurs when a dye is excited to a higher energy state by the excitation beam and spontaneously relaxes to the ground state once it finishes its life time (Figure 2.12). Due to vibrational relaxations, fluorescence emission is always red-shifted. A second laser beam coinciding with the emission wavelength of the fluorophore (the depletion laser) when allowed to be incident on the same position, it depopulates the excited state. This process is known as stimulated emission, which is the basic concept in STED microscopy. In the region, where both beams are present, stimulated emission (i.e. no fluorescence) occurs while in the region where only the excitation beam exists, spontaneous emission (i.e. fluorescence) is observed. Thus, the effective molecular excitation is confined to the doughnut center. The higher the intensity of the depletion beam, the narrower the spot becomes from which the fluorescence may originate.

In order to use this concept in microscopy, the PSF of the depletion laser beam should have a zero intensity in the center and non-zero in the periphery. The spatial structure of the depletion beam can be engineered by modifying the properties of the pupil plane of the objective lens, common example being
Figure 2.12  (a) Jablonski diagram showing the energy states of a fluorescent molecule. (b) Intensity profiles of the excitation beam in red, depletion beam in green and the resulting fluorescence in yellow [128].
diffractive optical elements, DOEs. This is used for lateral confinement in 2D which is generated by a helical phase ramp in combination with a circular polarisation of the depletion laser beam [131]. The lateral resolution of this DOE is typically between 30 and 80 nm. However, values down to 6 nm have been reported [132]. The wavelength of the depletion beam is such that it falls in the tail of the emission spectrum of the dye molecule without overlapping the absorption spectrum to avoid re-absorption. Using this method, a dramatic increase in lateral resolution is observed.

A number of concepts have been established for optically realising the driving of molecular transitions required in STED microscopy. The first implementation was based on pulsed lasers, where the effective stimulated emission probability can be optimised by proper selection of delays between excitation and depletion pulses and the depletion pulse length [133]. The need for expensive lasers and sophisticated pulse preparation methods, hampered the wider use of this concept. Thus an implementation based on continuous wave (CW) lasers was developed [134]. The ease of availability of CW laser sources has widened the usability of such systems. The viability of CW illumination greatly expands the range of STED microscopy with a demonstrated nanoscale resolving power both in the focal plane and along the optical axis.

The technique of STED microscopy is still evolving and has already become a useful tool for standard use. STED microscopy has found immense practical applications in numerous topics in cell biology, but also in material sciences that enables scientists to extend their study into the nanometer range [135, 136]. With multi-color super-resolution imaging, even the interactions between biological and engineered nanostructures can be studied in detail [137].
2.5 Chapter conclusions

In this chapter, we gave an overview of aspects of the current state of research in the fascinating nanofocusing applications of SPs. Plasmonic nanofocusing structures have demonstrated strong local field enhancement and confinement which has the potential for the design of new generations of sensors, detectors and nano-imaging techniques. A detailed investigation of different kind of nanostructures and their wide range of applications was given in this chapter. In the following chapters of this thesis we shall theoretically and experimentally investigate plasmonic lens structure which can achieve nanoscale focusing. A brief history and review of SNOM using tapered optical fibre as a probe has as well been discussed. A review on the current super-resolution technology of STED has also been given.
Chapter 3

Characterisation of in-plane polarisation fields using scanning near-field optical microscopy

3.1 Introduction

The primary aim of this thesis is to understand the focusing performance of a far-field plasmonic lens under different illumination polarisation conditions, which in detail is explained in the following chapter. And for this, it is necessary to understand the polarisation distributions at the focus of an low numerical aperture (NA) objective lens which focus the incident beam to the back focal plane of the plasmonic lens. This in turn will help in investigating the effect on surface plasmonic exciation of a plasmonic lens by different in-plane polarisation components. The focusing performance of a dual-wavelength plasmonic lens for different incident polarisations can then
be studied in detail from the knowledge of the distribution of the in-plane fields at the focus of an objective lens. This becomes an inevitable factor for the completion of this thesis. In this chapter, we determine the properties of an in-plane polarisation sensitive scanning near-field microscopic (SNOM) probe and its ability to detect polarisation fields at the focus of a low NA objective lens, which weakly focus the incident beam to the back aperture of the plasmonic lens. We will also determine the ability of the probe to characterise the plasmonic modes of a silver nanowire (Ag NW). Here we aim to address the following questions:

1. Can an elliptical shaped SNOM probe show polarisation sensitivity?

2. Can it resolve the field components of linear, radial, azimuthal and azimuthal vortex beams at the focus of a low NA objective lens?

3. Can it resolve near-field polarisation properties of a metallic NW?

As discussed in Chapter 1, the evanescent field components, accompanied with the highest spatial frequencies from an object, carries its finest information and are exponentially decaying in nature. A study of this field can result in a deeper understanding of the object’s characteristics possessing many advantages. In order to gain a physical insight into the property of this field at the focus of an objective lens, a detailed near-field characterisation mechanism is needed. SNOM, based on the detection of evanescent waves in the near-field zone, can have a resolution beyond the diffraction limit by bringing the probe at a distance around 20 nm, much smaller than the wavelength of light. The most common practical implementation of collecting the near-field signal is by employing a tapered optical fibre with a metallic coating and with a hole at the apex enabling a spatial resolution of 50-100 nm. This probe forms the most important part of a near-field microscope. To revamp the actual near-field distribution of the object under study, a thorough understanding of the
optical properties of the fibre probe is required.

The aim of this chapter is to present our contribution to the most important discussion in near-field optics, that is, the light-probe coupling providing the experimental details on how the shape of the probe aperture (elliptical or circular) give a differential information on the polarisation state of near-field light. Section 3.2 of the chapter covers the in-plane polarisation sensitive probe which can map transverse electric field components in the near-field regime of a structure. The three dimensional intensity distributions of the in-plane polarisation components at the focus of an objective lens for linear, radial, azimuthal and azimuthal first-order vortex (FOV) incident polarisations are studied in Section 3.3. Section 3.4 discusses the experimental investigation of the polarisation components of the propagating plasmonic mode of the Ag NW waveguide on a glass substrate. Section 3.5 delivers the chapter conclusions.

3.2 In-plane polarisation sensitive probe

For many applications involving SNOM, gaining information and controlling of the polarisation state of light in the near-field regime is important [138–140]. Previous studies show that typical SNOM probes with sub-wavelength apertures have higher sensitivity towards the electromagnetic components transverse or parallel to the plane of the aperture compared to longitudinal field components [49, 141]. Whereas scattering tips [142] and tip with apertures of approximately full wavelength diameter [143] have higher sensitivity towards longitudinal field components.

It has been shown in a previous report that the polarisation-sensitivity of the tip is related to the shape of the aperture [144]. An elliptical shaped probe has a proven sensitivity towards the in-plane field components ($E_x$ and $E_y$) and while a circular shaped probe has a high collection efficiency towards the
out of plane electric field components ($E_z$). Such a preferential sensitivity of the fibre probe controls the intensity of the in-plane and the out-of-plane electric field components in the collected signal since the field components of the optical near-field couple to the orthogonal polarisations in the optical fibre differently. In our measurements, we used commercially available SNOM probe (Nufern fibre probe, NT-MDT) with an Aluminium coating of a thickness of 50 nm. The aperture of the probe has a diameter around 50-100 nm which yields a resolution around 100 nm [145]. The transmitted signal from the probe is collected by a single photon photomultiplier tube (PMT). By scanning in a transverse plane above the lens surface, SNOM measures the evanescent waves in the near-field regime. A closer view of the shape of the probe is obtained from the scanning electron microscopic (SEM) image (Figure 3.1(a)). The measured aspect ratio of the aperture of the probe is $\sim 2:1$, indicating an elliptical shape. For comparison, SEM image of a circular (out-of-plane polarisation sensitive) probe is also taken (Figure 3.1(b)).
3.3 Characterisation of the in-plane polarisation components of the focal fields

The relative strength of the longitudinal ($E_z$) and transverse ($E_x$ and $E_y$) electric field components at the focus of linear, radial and azimuthal polarised beams are different when focused by objective lenses of different numerical aperture. In our experiments, we use a low NA objective lens to focus the incident beam and therefore the dominant electric field polarisation are transverse in nature [146, 147]. Pure-transverse focal fields are highly desired for energy efficient microscopic applications in harmonic generation [148, 149], single molecule detections [150, 151], coherence tomography [152] and multidimensional optical data storage [153–156]. In this section, we investigate the dominant in-plane (transverse) electric field components at the focus of an objective lens (NA = 0.7) using an in-plane polarisation sensitive SNOM probe.

3.3.1 Linearly polarised beam

We employ an in-plane polarisation sensitive SNOM probe for the characterisation of the focal spot of an 0.7 NA objective lens under linearly polarised plane wave illumination and for quantifying its directional polarisation sensitivity. A schematic diagram of the experimental set up is shown in Figure 3.2. The incident laser beam at a wavelength of 632.8 nm (He-Ne laser) is expanded by a beam expander to completely match the back aperture of the objective lens. A 0.7 NA objective focuses the incident beam to a cover glass which is mounted on a sample stage. A SNOM head with the fibre probe is placed vertical and perpendicular to the two dimensional sample scanning stage. A charge coupling device (CCD) camera is used to align the fibre probe in the
vicinity of the highly localised focal spot of the objective. A calibration of the SNOM is done each time before the experiment, by running a calibration file provided by the manufacturer.

The focus of the low NA objective is directly mapped by the probe in a transverse plane parallel to the sample surface. The input polarisation of the beam is rotated by a half wave plate from $0^\circ$ to $180^\circ$ without changing the incident laser power on to the sample. During experiment, the distance between the sample surface and the fibre probe is controlled by a shear force feedback mechanism. For each incident polarisation angle, the probe is scanned across the focal spot of the objective lens. The collected intensity by the probe is transmitted along a 1 m long optical fibre which in turn is fed to a PMT. The intensity of the focal spot of an objective lens measured by an in-plane polarisation sensitive probe as a function of the incident

Figure 3.2 Scheme of the experimental set up for the mapping of the focused evanescent field using a SNOM probe.
Figure 3.3 The intensity of the focal spot of an objective lens by an in-plane polarisation sensitive probe as a function of the incident polarisation rotation angle. (b) and (c) Simulated intensity distributions for the two incident perpendicular polarisations of a linearly polarised beam. SNOM optical images of the focal spot of a linearly polarised beam when the preferred polarisation sensitive direction of the probe is (d) perpendicular and (e) parallel to the excitation direction. The scale bar is 600 nm.
polarisation angle is shown in Figure 3.3(a). The mapping of the focal spot produced by the objective lens, when the preferred polarisation direction of the probe is perpendicular (Figure 3.3(d)) and parallel (Figure 3.3(e)) to the excitation direction. Since the probe employed is unable to collect the longitudinal polarisation components, the polarisation components we measured were completely transverse in nature. The coupling efficiencies of different transverse polarisation components to the probe is obtained by mapping the focal spot of the objective lens for each incident polarisation angle. As in Figure 3.3, the probe shows a ~10 times higher collection efficiency along one polarisation direction than the perpendicular direction and thereby showing its preferred polarisation sensitivity [157]. Similarly, this behaviour was already pointed out in previously published theoretical [158] and experimental studies [144].

3.3.2 Radially polarised beam

Due to its inherent symmetric polarisation behaviour radial polarisation have gained particular attention. Numerous theoretical and experimental studies have been extensively carried out to investigate the focusing properties of a radially polarised beam [159–161]. When radially polarised light is focused by a low NA objective lens, the transverse component is dominant in nature [146, 147]. To investigate the coupling of these components to the plasmonic lens, understanding radial polarisation and the characteristic features of its transverse components at the focus of a low NA objective lens is a prerequisite. From the previous sections in this chapter, now it has become quite obvious that SNOM employing an in-plane polarisation sensitive probe is inevitable for this.

The first step to carry out the experiment is to generate a radial polarisation
beam. We used commercially available radial polarisation converter (RPC, ARCOPTIX) for the generation [162]. We use the same experimental set up as in Figure 3.2, except that a radial polarisation converter is inserted in the beam path after the linear polariser. After beam expansion and spatial filtering by the two lenses, the vertically polarised laser beam is passed through the radial converter, which consists of a polarisation converter, TN (twisted nematic) cell and a phase retarder (Figure 3.4(a)). They together convert the linear polarised beam to a radially polarised one. The polarisation of light coming out from the RPC is checked by using a linear polariser. Figure 3.4(b) shows the intensity distribution when a vertical polariser is kept after the radial converter. The two lobes separated in vertical directions confirms the radial polarisation. Figure 3.4(c) shows the doughnut shaped focus when the radially polarised beam is focused by an 0.7 NA objective lens. Figures 3.4(f) and (g) shows the intensity distributions of the focal fields of an tightly focused radially polarised beam with the polarisation sensitive axis of the SNOM probe along the x- and y- directions respectively. Comparison of the intensity distributions obtained from experiment shows good theoretical agreement (Figures 3.4(d) and (e)).

### 3.3.3 Azimuthally polarised beam

As one of its fundamental aspects, focused light under optical microscopy [163, 164] can selectively interact with anisotropic materials through its polarisation state, which provides the basic principle of polarisation-sensitive nanophotonic devices. In this context, the transversely polarised beam such as the linear and circular polarisation focused by an objective lens with a high NA is essential for a high microscopic resolution with a high energy density of transverse focal fields. However, the depolarisation effect leads to the degraded transverse
Figure 3.4  (a) Scheme of the radial polarisation converter. Image of the radially polarised beam (b) after passing through a vertical polariser and (c) focused by an 0.7 NA objective lens. (d) and (e) Simulated intensity patterns of focal fields of $E_x$ and $E_y$ of an radially polarised beam, respectively. (f) and (g) Normalised intensity patterns of focal fields of $E_x$ and $E_y$ of an radially polarised beam, respectively. The scale bar is 1.0 μm and the two headed arrows indicate the direction of probe sensitivity.
polarisation purity of linearly and circularly polarised beam with a 17% energy loss at tightly focusing condition by an objective with a high NA of 1.4 through the emergence of longitudinal field components [165–167].

Owing to its symmetry in a tangential polarisation distribution, the azimuthally polarised beam can lead to pure-transverse focal fields disregarding the NA of the objective lens [159, 160]. However, the de constructive interference between its transverse focal field components can only result in a wide doughnut-shaped focus [168], degrading the energy density of the focal fields [169]. Figures 3.5(a) and (b) shows the simulated intensity distributions of the focal fields of $E_x$ and $E_y$ of an azimuthally polarised beam. Figures 3.5(c) and (d) shows the intensity distributions of the focal fields of $E_x$ and $E_y$ of an azimuthally polarised beam, with the polarisation sensitive axis of the SNOM probe along the x- and y- directions respectively. A doughnut-shaped distribution with a hollow intensity at its center can be clearly evident [160].
3.3.4 Azimuthally polarised first-order vortex beam

In this section, the phase effect on an azimuthally polarised beam is studied. We present the experimental demonstration of a super-resolved pure-transverse focal fields through focusing an azimuthally polarised beam superimposed with the FOV phase. Generating sharper focal fields of modulated azimuthally polarised beams is a subject that received numerous theoretical investigations [170–172].

The scheme of creating constructive interference between the transverse focal fields through superimposing the FOV phase is illustrated in Figure 3.6(a). The focal fields of such an azimuthally polarised FOV beam can be given as,

\[ E(r, \Phi, z) = \frac{\Pi}{\lambda} (E_x - iE_y) \]  

(3.1)

\[ E_x = \int_0^\pi P(\theta) \sin(\theta) \left\{ -\exp[i2\Phi] J_2(kr\sin\theta) - J_0(kr\sin\theta) \right\} \exp(-ikz\cos\theta) d\theta \]  

(3.2)

\[ E_y = \int_0^\pi P(\theta) \sin(\theta) \left\{ -\exp[i2\Phi] J_2(kr\sin\theta) + J_0(kr\sin\theta) \right\} \exp(-ikz\cos\theta) d\theta \]  

(3.3)

where, \( P(\theta) \) is the sine condition of the apodisation function. Figure 3.6(a) shows the numerical calculation of the focal fields of an objective lens with a high \( \text{NA}=1.4 \), following the Equations 3.1, 3.2 and 3.3. Indeed, the FOV phase can create constructive interference and gain the analogous advantage of the radial polarisation, resulting in a tight confinement described by the zero-order Bessel function accompanied by a negligible side lobe with a second-order Bessel distribution. The longitudinal component is null due to the
Figure 3.6  (a) Schematic illustration of creating the constructive interference between the transverse focal fields through the superposition of the FOV phase. Intensity patterns of focal fields of an azimuthally polarised beam superimposed the FOV phase. The image size is 1 μm. (b) and (c) Normalised intensity distribution of $E_x$ and $E_y$ components of an azimuthally polarised beam superimposed with the FOV, respectively. The scale bar is 300 nm and the two headed arrows indicate the direction of probe sensitivity.

axial depolarisation-free nature associated with the tangential polarisation distribution of such a modulated azimuthally polarised beam.

To verify the optimised confinement of pure-transverse focal fields, the spatial distribution of the focal field of an azimuthally polarised beam superimposed with the FOV phase is mapped through SNOM. In the presence of the FOV phase, the hollow intensity at the center of the focal area is removed. The FOV phase modulation generates a solid focal spot with a remarkably smaller distribution. A tight lateral confinement in the $y$-direction for $E_x$ with a FWHM of 0.52 $\lambda$ at a moderate focusing condition by an objective with NA=0.7 can be clearly revealed, as shown in Figure 3.6(b). Although this advantage is obtained at the cost of an increased side lobes, the intensities are negligible with less than 15% of the main peak. By rotating the polarisation-
sensitive axis of the SNOM tip $90^\circ$, a similar confinement in the x-direction for $E_y$ can be found in Figure 3.6(c). In addition, the peak intensity of the solid focal spot is measured to be approximately 1.8 times stronger than that of the doughnut-shaped wide focus, which is consistent with the calculations following Equations 3.1, 3.2 and 3.3 as depicted in 3.6(a).

In conclusion, we have demonstrated super resolved pure-transverse focal fields with an enhanced focal energy density through focusing an azimuthally polarised FOV beam [173]. This feature has been achieved through breaking the rotational symmetry in phase of an azimuthally polarised beam to create constructive interference between its transverse focal field components. This was experimentally corroborated by the near-field mapping using an in-plane sensitive SNOM probe. The remarkably reduced focal area maintains from the moderate to tight focusing conditions, paving the way for a variety of high-resolution microscopic applications.

### 3.4 Characterisation of the in-plane polarisation components of a plasmonic nanowire

Ag NWs on a dielectric substrate have been widely studied [174], owing to the relatively larger propagation length, stronger energy confinement and enhanced evanescent field at the interface compared to free-standing NW, which opens up the possibility for novel and high performance miniaturised optical devices [175, 176]. The presence of the substrate can break up the symmetry and fundamentally change the plasmonic modes supported in the hybrid structure. Thus the NW waveguide on the substrate can turn into a single mode operation [177], which is crucial for practical optical waveguide
applications. Because of its sensitivity to the evanescent wave and high spatial resolution, SNOM is widely used to study the localised plasmonic modes in Ag NW [178, 179]. However, the near-field studies of Ag NWs are mainly focused on intensity distribution [180, 181]. In this section, we discuss on the experimental investigation of the polarisation properties of the plasmonic modes along a Ag NW waveguide on a glass substrate. Initially, we did a far-field characterisation using a CCD camera and we observed two orthogonal polarisation light components at the distal end of the NW. To have a detailed understanding of this two polarisation components, we compare it with the near-field imaging using a SNOM probe exhibiting an in-plane polarisation sensitivity [157].

3.4.1 Theoretical study on the polarisation state of the light emitting from a silver nanowire

The wave guiding properties of an Ag NW, with the same diameter as employed in our experiment, is investigated using a COMSOL Multiphysics finite element method. The free standing NW shows a symmetry mode profile while the dielectric substrate breaks the symmetry of the modes and confines the energy at the interface of the NW and the dielectric. The arrow lines in Figures 3.7(a) and (b) shows the direction of the dominant modes along the NW. For the NW embedded in air, the dominant modes have a symmetric distribution (Figure 3.7(a)) while for the NW supported on a substrate, the dominant modes are confined to one direction, towards the substrate-wire interface (Figure 3.7(b)).

To understand the polarisation of the plasmon modes excited along the NW supported on a substrate, with linearly polarised illumination, the field intensity distribution in a vertical plane, around 100 nm away from the distal end of the NW is measured using COMSOL Multiphysics module. From this,
the degree of circular polarisation, $C$ is calculated and thereby evaluating the SP field at the distal end of the wire. When the NW is illuminated by a wave propagating along the wire axis, strong longitudinal components are also induced which is needed to be considered in the background.

$$C = \frac{2 < E_y(t)E_z(t)sin(\delta_y-\delta_z)>}{< E_x^2(t) > + < E_y^2(t) > + < E_z^2(t) >} \quad (3.4)$$

where $(\delta_y-\delta_z)$ is the phase difference between the transverse and longitudinal components of electric field, $E_y(t)$ and $E_z(t)$; $<>$ denotes the time average. The variation of $C$ with the diameter of the NW is shown in Figure 3.7(c). The measured smaller value for $C$ for NWs supported on a substrate gives an insight to the stronger linear polarisation of the modes in this condition. For a NW of diameter 150 nm, $C$ value is measured as 0.06, from Figure 3.7(c).

### 3.4.2 Far-field characterisation of a silver nanowire

For both far-field and near-field characterisation of the plasmonic modes of a Ag NW, we use the same experimental set up as in Figure 3.2. Propagating plasmonic modes are excited at one end of an Ag NW by the focal optical irradiation $[182, 183]$, due to the symmetry breaking at the excitation end of the NW $[184]$. This excited plasmonic modes have a propagation length ($L_{SP}$) which is defined as the energy decay length for plasmons propagation along the wires; The intensity of the SP field at a point $x$ along the propagation direction is given by

$$I_{SP} = I_0e^{-x/L_{SP}} \quad (3.5)$$
Figure 3.7  Schematic illustration of the arrow line distribution for the dominant modes on the cross-section of the NW (a) no substrate, m=0 mode (b) with substrate, the confined fundamental mode. (c) Variation of Degree of polarisation (C) with the diameter of an Ag NW in a vertical plane, 100 nm away from the exit end of the NW (length of the wire is kept as 4µm). The low value of C for 150 nm diameter NW confirms the mode polarisation as linear.
The Ag NWs are commercially available with a diameter of ~150 nm (±10 nm) and a length of ~7 µm (±0.2 µm). Figure 3.10(a) shows an SEM image of the typical example of the NW. The incident laser beam at a wavelength of 632.8 nm (He-Ne laser) is focused by a high NA (NA=1.4) oil immersion objective, to one end facet of the Ag NW on a cover glass (R.I = 1.46) [185]. The excitation geometry is shown in Figure 3.8. The polarisation angle (θ, defined as the polarisation orientation with respect to the axis of the NW) of the incident beam is changed by rotating a half wave plate placed after the linear polariser in Figure 3.2. The incident laser beam is coupled to the NW as propagating SPs. The SPs at the distal end is scattered to the far-field and imaged by the same objective with a CCD camera. When the input polarisation is parallel to the orientation of the NW, the depolarised longitudinal field components induced by the tight focusing with a high NA objective, match well with the electric field of the single mode in the NW which is highly polarised along the longitudinal direction. This feature leads to the maximal coupling efficiency producing a bright spot at the distal end of the NW (Figure 3.10(b)). When the polarisation is perpendicular to the orientation of the NW, the longitudinal
field components do not match with the electric field of the mode leading to a minimal coupling efficiency and thereby producing a dark spot at the distal end of the wire (Figure 3.10(e)) [157]. This observation is consistent with a previous report [178]. The dependence of the scattering intensity at the distal end of the NW on the incident polarisation orientation can be seen in Figures 3.10(b) to (e).

To analyse the polarisation state at the distal end, a polarisation analyser is placed in front of the CCD. The dependence of the emission intensity at the distal end under three different incident polarisation orientations on the direction of the analyser is shown in Figure 3.9. In the current experimental geometry, only single mode can be supported in the NW. Thus, the ratio of the scattered electric fields in the two orthogonal directions is a characteristic of the single mode, which is independent on the input polarisation. At the distal end, the parallel electric field is constantly 2.5 times stronger than the perpendicular one, which might be attributed to the additional contribution from the depolarised transverse fields collected by the high NA objective. The output polarisation is sensitive to the shape of the distal end when high-order modes exist in the NW, as shown in Ref [182]. In contrast to the multimode case, the observed far-field polarisation in the distal end is dominantly parallel to the direction of the NW, which is a characteristic of the single mode. As the analyser in front of the CCD detector is rotated, different plasmonic mode distributions are observed as in Figures 3.10(f) to (i). The parallel (when the direction of the analyser is parallel to the axis of the NW) and perpendicular (when the analyser is perpendicular to the axis of the NW) polarisation observed in Figures 3.10(f) and (i), are the scattered electric fields of the NW and collected by the objective. The parallel electric fields are located in the center of the NW, while the perpendicular electric fields are along the edge of the NW. The different intensity distributions indicate that these field components originate from two different polarisation components.
Figure 3.9 Emission intensity from the distal end of the NW as a function of the analyser angle for different excitation polarisation orientations with respect to the axis of the NW.

with different spatial distributions [157].

Figure 3.10 SEM image of the NW (diameter ~150 nm (± 10 nm) and length ~7 µm (± 0.2 µm). (b), (c), (d) and (e) show the change in the intensity at the distal end of the wire as the incident polarisation is changed. (The scale bar is 2 µm and the double headed arrows indicate the excitation polarisation orientation). (f), (g), (h) and (i) show the different plasmonic mode distributions on the Ag NW when the analyser in front of the CCD detector is rotated. (The scale bar is 2 µm, the double headed arrows indicate the excitation polarisation orientation and the dashed arrows indicate the direction of analyser).
3.4.3 Near-field characterisation of a silver nanowire

For the further confirmation of the existence of the two polarisation components of the plasmonic modes, a near-field mapping of the propagating mode along the Ag NW is carried out. An in-plane polarisation sensitive SNOM probe is employed in the characterisation. For the near-field characterisation, the incident polarisation is aligned parallel to the axis of the NW along x-coordinate. The transverse mode imaging is obtained when the probe sensitive direction is rotated parallel and perpendicular to the axis of the NW (Figures 3.11(a) and (b)), respectively. Indeed, the images show two orthogonal polarisation components. The x-polarised component is located at the center of the NW, while the y-polarised component is at the edge of the NW. The high resolution of SNOM allows the existence of the periodic peaks due to the interference of the propagating plasmonic modes along the NW. The same periodicity between two polarisation components is revealed, indicating that they are two polarisation components of the same order of modes, which is consistent with the previous report [176].

For a comparison, we employed the finite element method to simulate the field distribution under the same excitation condition. The excitation geometry was depicted in Figure 3.8 with a Gaussian excitation beam. Optical constants of Ag at wavelength 632.8 nm are taken as refractive index, R.I = 0.1437 + i3.8082 [19]. In our case, only the first order mode is excited due to the symmetry breaking when the NW was on the glass substrate [176]. The electric field components of the plasmonic mode in x and y-directions are shown in Figures 3.11(c) and (d), respectively. The separation of the intensity peaks is measured as 350 nm, which is slightly larger than the simulation result of 250 nm. This discrepancy may be attributed to the Tien effect [186] considering the refractive-index gradient in the radial direction caused by the quick oxidation of the surface of the Ag NW in air. While, the qualitative agreement between
the simulation and experimental results confirms that individual polarisation components have been revealed distinctively by the in-plane sensitive probe.

In conclusion, we have experimentally investigated the polarisation components of the propagating plasmonic mode of the Ag NW waveguide on a glass substrate by the SNOM. The observation of the two orthogonal polarisation components of the plasmon modes along the axis of the NW is confirmed by the near-field mapping experiments by an in-plane polarisation sensitive probe [157].

3.5 Chapter conclusions

In conclusion, the near-field polarisation properties of SNOM fibre probes were characterised, demonstrating that an elliptical shaped probe have a preferential sensitivity towards different in-plane polarisation components. We investigated and resolved the in-plane field components of linear, radial, azimuthal and
azimuthal vortex beams at the focus of an objective lens (NA = 0.7). In-plane polarisation fields at the focus of a low NA objective lens forms an important factor for the excitation of our far-field plasmonic lens. And therefore, a comprehensive understanding of the former will help to investigate the focusing action of plasmonic lens for different incident polarisations. We demonstrated the super resolved pure-transverse focal fields with an enhanced focal energy density through focusing an azimuthally polarised FOV beam using a low NA objective lens. Our near-field mapping results on the orthogonal polarisation components of the hybrid NW structure can open up a new avenue for developing polarisation-sensitive applications [166] such as optical nano-polarisers.
Chapter 4

Focusing dual-wavelength surface plasmons to the same focal plane by a far-field plasmonic lens

4.1 Introduction to far-field plasmonic lenses

Plasmonic focusing devices utilising sub-wavelength light confinement and strong field enhancement of surface plasmons (SPs), can break the diffraction limit and can allow for manipulation of light on sub-wavelength scales, opening up intriguing applications in imaging [187], sensing [188] and integrated optical circuits [41]. Nanoscale focusing devices based on SPs have dimensions of the order of microns and can as well produce nanoscale focal spot with a local field enhancement in the focal region. A plasmonic lens [49] consisting of a single annular slit with sub-wavelength width has been reported to achieve high intensity focusing at the surface. The sub-wavelength slit can excite and
focus SPs. However, there was no physical mechanism to convert SPs into propagating waves in free space, thus restricting its focus to be in the near-field. Many plasmonic structures such as nano hole arrays [189], nano slits [62, 190] and annular ring structures [80, 191] have attracted intense research efforts, although their applications are limited to near-field operations. On the other hand, a plasmonic lens with an annular slit and a concentric groove has been shown via numerical simulation to achieve sub-wavelength focusing at the far-field [192]. Many structures demonstrating diffraction limited far-field focusing have been introduced, including flat metamaterial based lenses [193], but the specific dispersion property associated with the structure design limits their performance to usually single-wavelength SP operation. Once the geometry is given, these far-field plasmonic lenses can operate only at a specific wavelength of SPs [194]. Achieving far-field focusing of multiple wavelengths to the same focal plane is therefore elusive. The major aim of this thesis is to provide solutions to control the dispersion and to achieve far-field nanofocusing of multiple wavelength of light to the same focal plane.

In this chapter, we present a far-field plasmonic lens capable of nanofocusing of dual-wavelength of light to the same focal plane by optimising the concentric groove structures. Dual-wavelength focusing to the same focal plane is crucial for applications such as stimulated emission depletion based imaging and lithography. Numerical calculations in COMSOL theoretically demonstrate dual-wavelength nanofocusing by the same far-field plasmonic lens. Our experimental demonstration provides further convincing validation of this concept [195]. The chapter is organised as follows. Theory and design principles of a far-field plasmonic lens and a dual-wavelength far-field plasmonic lens is presented under Section 4.2. The optimisation of the plasmonic lens design by changing different structure parameters and its fabrication details are shown in this section. The experimental demonstration of the far-field focusing behaviour of a dual-wavelength plasmonic lens under
different polarisation illumination conditions viz; linear and radial is provided in Section 4.3. Mapping of the focal field distribution by a scanning near-field optical microscope (SNOM) reveals the super-resolved diffraction limited focal spot produced at the same far-field focal plane of the plasmonic lens structure under two different incident wavelengths. In Section 4.4, conclusions of the chapter are discussed.

4.2 Theory and design

4.2.1 Far-field plasmonic lens

Theoretical simulations on plasmonic lens are done using finite element method (COMSOL). Due to the rotational symmetry of both the illumination and the plasmonic lens structure, simulation can be performed using the axial symmetric module of COMSOL. This reduces the full 3D model into a quasi-2D problem in the r–z plane, saving a considerable amount of memory and computing time. Continuity boundary conditions are applied to the dielectric/metal and metal/air interfaces. The scattering condition is chosen for the outer boundary to simulate an open boundary. We optimised our structure with the incident polarisation as radial because of its centrosymmetric transverse-magnetic (TM) polarised illumination, which is modelled using the electric field equation 4.1,

\[ E = \frac{r}{\omega} \exp\left(\frac{-r^2}{\omega^2}\right) \]  \hspace{1cm} (4.1)

where, \( \omega \) is the beam waist and \( r \) is the domain of the calculation in the \( r \)-direction.

Figure 4.1 represents the focusing scheme of the proposed far-field plasmonic
Figure 4.1  Scheme of focusing by a far-field plasmonic lens.

lens structure consisting of an annular slit and a single concentric groove within the slit. The thickness of the gold coating on the substrate is chosen to be above the skin depth of the gold so as to avoid the direct transmission of light, the latter may affect the quality of focusing \cite{26, 27, 53}. The sub-wavelength annular slit, fabricated on a thin gold film, acts as a circular grating and couples the incident light into SPs, as discussed in Chapter 2. The counter propagating SPs, which are thereby excited, undergo interference producing a standing wave pattern with the annular slit as the boundary \cite{38}. The outer diameter of the annular slit structure, its depth and width are numerically optimised using COMSOL \cite{192}. As the slit diameter is increased, more energy can be converted to SPs due to the increase in the circumference of the structure. By taking into account the energy loss of SPs in the propagating process, we find an optimised value for the slit diameter, 5 µm. Inside the annular slit, a concentric groove is milled with a depth of 80 nm and a width of 100 nm.

The inside groove scatters SPs out of the interface and plays an important role in the scattering of SP waves as propagating waves at a certain angular spectrum distribution \cite{192}. The groove is placed at locations where the node of the standing wave pattern appears while keeping its depth and width constant. The focal length and thereby the focusing efficiency of the plasmonic
lens can be tuned by changing the annular groove radius as shown in Figure 4.2 with a fixed groove depth of 80 nm. This is because as the groove diameter is varied, the scattering angle is changed thereby changing the point where all the in-phase propagating waves undergo interference thereby changing the focal point. From this result, an optimised focusing condition can be obtained. The field scattered by the groove undergoes interference with the transmitted field from the annular slit, concentrating most of the energy in the far-field regime of the structure.

The incident wavelength is chosen to be 632.8 nm for the calculation. The complex permittivity of Au corresponding to this wavelength is $\varepsilon_{\text{Au}} = -9.7997 + i1.9649$ [196]. The corresponding wavelength of SPs to this incident wavelength is 599.0 nm obtained from Equation 1.4.

In order to attain a high SPs coupling efficiency, radially polarised light is chosen to be the illuminating source for the calculation [86, 87]. Three-dimensional nanoscale focusing in the far-field (1.2 $\mu$m from the lens surface) is realised by converging SPs scattered by the groove. When there is no groove,
Figure 4.3 (a) and (b) shows the near-field (without groove) and far-field (with groove) focusing by a plasmonic lens under radially polarised excitation. (white dashed lines show the focal plane and the white dotted lines show the scheme of the far-field plasmonic lens).

there is no scattering of SPs and no far-field focusing. Figures 4.3(a) and (b) shows the near-field and far-field focusing by a plasmonic lens, respectively. The overall transmitted energy of the structure is calculated to be about 13%.

4.2.2 Dual-wavelength far-field plasmonic lens

SPs are collective oscillations of free electron density at the surface of a metal. As discussed, to support SPs at the interface of the metal and the surrounding medium, the relative permittivities of the former and the latter must be opposite and these conditions are fulfilled by gold (Au) in visible and near-infrared regions as shown in Figure 4.4(a). Using equation 1.4, the SP wavelength for different incident wavelengths is obtained as illustrated in Figure 4.4(b). For the free space wavelength region from 600 nm to 1000 nm, the variation in Figure 4.4(b) has a flat behaviour. This window can be employed for multi-wavelength focusing.
Figure 4.4  (a) Imaginary part and real part of the dielectric function of gold. The data is computed using the Drude approximation.  (b) Variation of normalised surface plasmon wavelength with free space wavelength in visible and near-infrared regions.
Chapter 4

Figure 4.5 Schematic illustration of the principle of a dual-wavelength far-field plasmonic lens (Black single headed arrow lines indicate the scattering of surface waves by the concentric groove). The inset shows the principle of the dispersion control by matching the node position (shown as dashed square boxes) of the surface waves for the two incident wavelengths.

The principle of dual-wavelength focusing by the far-field plasmonic lens, consisting of an annular slit and a concentric groove is shown in Figure 4.5 and is numerically studied using COMSOL. As described in Section 4.2.1, the far-field focusing of one single wavelength by the plasmonic lens is attributed to the relative position of the groove structure and the nodes of the standing wave pattern produced from the constructive interference of SP waves. For the realisation of far-field focusing of two wavelengths and for the dispersion control, relative position of groove still plays an important factor [192, 197]. Initially we optimised the focusing action of the plasmonic lens with one wavelength, 632.8 nm ($\lambda_1$) and then chose the 2nd wavelength ($\lambda_2$) whose node appears at the same position as 632.8 nm. The groove is placed where the node positions of the two interference patterns of standing waves of SPs match (shown as the dashed square box in the inset of Figure 4.5), which scatters the dual-wavelength SPs to the same focal plane. The scattering of the highly confined surface waves into propagating waves is indicated by the black single headed arrow in Figure 4.5.
In the simulation, beams at the wavelength of 632.8 nm ($\lambda_1$) and 750.0 nm ($\lambda_2$), respectively, were chosen (the same notation will be used throughout this thesis). The complex permittivities of Au corresponding to $\lambda_1$ and $\lambda_2$ are taken as $-9.7997 + i1.9649$ and $-16.9170 + i1.9602$, respectively [196]. The corresponding wavelengths of SPs ($\lambda_{SP}$) to these incident wavelengths are 599.0 nm and 727.5 nm, respectively, obtained from equation 1.4. Thus, the evanescent standing waves generated at the exit interface of the metal and dielectrics have a variant periodicity as $\lambda_{SP}/2$, following the dispersion of SPs in equation 1.2. Figures 4.6(a) and (b) shows the simulated nanofocusing with the normalised energy distribution in the xz-plane for $\lambda_1$ and $\lambda_2$, clearly exhibiting focusing to the same far-field focal plane [195].
Figure 4.7 Fabrication sequence for the far-field plasmonic lens: (a) Fused silica substrate (b) Sputtering gold film of 200 nm thickness on to the substrate (c) Fabrication of concentric ring structures on to the gold using focused beam of ions (FIB method) (d) Two dimensional cross-section of a far-field plasmonic lens.

4.2.3 Fabrication method

The first consideration for the fabrication procedure of the plasmonic lens structure is to have a smooth thin layer of gold film (200 nm) on a fused silica substrate, which is important to avoid the scattering processes [34]. Before that a 5 nm thick Ti layer was sputtered to enhance the adhesion between quartz substrate and gold layer [198]. A relative smooth gold layer was deposited above it using sputtering process. For fabricating ring structures on the gold film, we employed focused ion beam (FIB) milling method. A focused beam of Gallium (Ga+) ions is used for material removal. Due to the transfer of energy from Ga ions to the target atoms, atoms gaining sufficient energy leave the surface. The strength of the ion current is based on the feature sizes, dwell time and number of passes. The beam current we employed was 37 pA with a voltage of 35 kV. While the dose for the outer ring was 30K \(\mu\text{C/cm}^2\) and for the inner ring, it was 2K \(\mu\text{C/cm}^2\). The fabrication sequence for the far-field plasmonic lens is shown in Figure 4.7(a) to (c). Two dimensional cross-section of a far-field plasmonic lens is shown in Figure 4.7(d). Scanning electron microscopic (SEM) image of the fabricated far-field plasmonic lens is
Figure 4.8  (a) SEM image of the far-field plasmonic lens. (b) Surface scan profile of the lens structure acquired from the topographic SNOM scan. (c) Cross-section of the far-field plasmonic lens along the dashed lines of Figure 4.8(b).

shown in Figure 4.8(a). Surface scan profile of the lens structure obtained from the topographic SNOM scan and its cross-section are shown in Figures 4.8(b) and (c) respectively.

4.2.4 Enhancement in focusing efficiency by multiple ring far-field plasmonic lens

To address the issue of low-throughput of a single ring far-field plasmonic lens, multiple ring plasmonic lenses are proposed. For improving the enhancement of the field at the focus of the plasmonic lens, concentric rings are added to single ring structure. The separation between the rings is kept at half
the SP wavelength for the efficient constructive interference of propagating plasmons excited at each ring. As the radius of the outer ring is increased, the circumference also increases and thus more energy is converted to SPs. We optimised the structure with 11 numbers of rings (Figure 4.9(a)) due to the limited propagation length of the plasmons [80]. SNOM near-field optical image of a 5-ring far-field plasmonic lens is shown in 4.9(b), under a radial polarised illumination at an incident wavelength, $\lambda_1$. The focal field enhancement with the addition of annular rings is shown in Figure 4.9(c).

4.3 Experimental characterisation of a dual-wavelength far-field plasmonic lens using scanning near-field optical microscopy

The schematic illustration of excitation of a dual-wavelength far-field plasmonic lens by a weakly focused Gaussian beam by an objective lens of low numerical aperture (NA = 0.7) and the characterisation by a SNOM probe is shown in Figure 4.10. The transmitted signal from the probe is collected by a single photon photomultiplier tube (PMT). By scanning in a transverse plane above the lens surface, SNOM measures the evanescent waves in the near-field regime. In this section, the focusing action of the dual-wavelength far-field plasmonic lens is studied under different polarisation illumination conditions. The transverse and axial response of the focal field of a far-field plasmonic lens is characterised for linear and radial polarisation illuminations.

As discussed in Chapter 2, the dominant electric field components at the focus of a plasmonic lens is longitudinal ($E_z$). Therefore we employ an out-of-plane sensitive SNOM probe for the characterisation experiments. The sensitivity of
Figure 4.9  (a) SEM image of a 11-ring far-field plasmonic lens. (b) Near-field throughput image of a 5-ring far-field plasmonic lens obtained using SNOM, under a radially polarised illumination. (The non-circular symmetric field distribution in the figure is attributed to the sensitivity of the SNOM probe. Although the probe is $E_z$ sensitive, it has a higher sensitivity towards $E_x$ components than the $E_y$). (c) Variation of normalised electric field enhancement with number of concentric rings.
a probe is confirmed by employing it for mapping the focal spot of an objective lens under linearly polarised illumination. The two lobe pattern obtained as in Figure 4.11(b), confirms its sensitivity towards $E_z$ components. If the probe is in-plane polarisation sensitive, the focal spot will be a single bright one as shown in Figure 3.3.
4.3.1 Under linearly polarised illumination

A normal Gaussian excitation beam from a broad band laser at the wavelength of $\lambda_1$ and $\lambda_2$ is achieved by weakly focusing through an objective lens with numerical aperture (NA = 0.7) to the far-field plasmonic lens. The beam is collimated at the back aperture of the objective for a uniform excitation of SPs along the circumference of the structure. To measure the focal length of the far-field plasmonic lens, a z-scanning method of the fibre probe is employed. The probe is retrieved in regular intervals of 100 nm from the lens surface until it is 2 microns away from it. At each z-plane, a regular x-y scanning is performed. The axial response of the far-field plasmonic lens at $\lambda_2$ is measured and is compared with that at $\lambda_1$ and also with the focal length given by the numerical simulations. At the optimised groove radius of 1100 nm, corresponding to the node position of SP wavelength of 599.0 nm and 727.5 nm, the plasmonic lens can focus the dual-wavelength SPs to the same focal plane. From the SNOM characterisation experiments, the focal length of the far-field plasmonic lens at 750 nm excitation is experimentally measured to be $1.2 \mu m$ (dashed lines in Figure 4.12), which is identical for the illumination at the incident wavelength of 632.8 nm. This result is consistent with the simulation result obtained of $1.15 \mu m$ (dotted lines in Figure 4.6(a) and (b)).

When a linearly polarised beam is incident on the back aperture of the plasmonic lens only portions of the annular slit where a component of the incident electric field is perpendicular to the slit edge can excite SPs. This criterion is satisfied only at two opposite points on the circumference of the structure [38]. The $E_Z$ components in the focal plane become out of phase (Figure 4.13(a) and (b)) producing two focal lobes in the far-field, due to their destructive interference at the centre. At the exact focal plane the two-lobe intensity distributions can be obtained for the two incident wavelengths, $\lambda_1$
Figure 4.12  (a) Comparison of the axial response of the two incident wavelengths, $\lambda_1$ and $\lambda_2$ (the dotted and dashed lines indicates the focal length of the lens obtained numerically and experimentally).

and $\lambda_2$. At the focal plane of 1.2 $\mu$m with respect to the lens surface, the separation between the two lobes for $\lambda_1$ and $\lambda_2$ is measured to be $0.65 \pm 0.01 \lambda_1$ and $0.63 \pm 0.01 \lambda_2$ (Figures 4.13(e) and (f)) respectively. This is consistent with the numerical simulations (Figures 4.13(c) and (d)) [195].
Figure 4.13  Simulated phase distribution of the longitudinal components in the focal plane of a far-field plasmonic lens for (a) $\lambda_1$ and (b) $\lambda_2$. Simulated cross-sections of the normalised intensity distributions in the focal region of a far-field plasmonic lens for (c) $\lambda_1$ and (d) $\lambda_2$; Experimental cross-sections of the normalised intensity distributions in the focal region of a far-field plasmonic lens for (e) $\lambda_1$ and (f) $\lambda_2$; Polarisation orientation is indicated by arrow in the top left corner.
4.3.2 Under radially polarised illumination

For improving the SP excitation efficiency and thereby improving the resolution of the plasmonic lens, a radially polarised illumination is employed. A radial incident polarisation, which is always TM polarised with respect to the outer slit, causes the excitation of SPs all around the slit with their out-of-plane electric field components always in-phase when propagating towards the center [199, 200]. The groove scatters these components to the far-field, producing a much tighter focal spot at the center. This tighter focal spot is attributed to the $E_z$ components being in-phase in the focal plane (Figure 4.14(a) and (b)). A single focal spot with much improved resolution is observed numerically in the same focal plane for $\lambda_1$ and $\lambda_2$, respectively (Figures 4.14(c) and (d)). These features were corroborated by the experimental full width at half maximum (FWHM) of the lateral intensity distribution at the focal plane of 1.2 $\mu$m, measured as $0.45 \pm 0.01 \lambda_1$ and $0.45 \pm 0.01 \lambda_2$, for the two incident wavelengths respectively (Figures 4.14(e) and (f)) [195]. The generation of radially polarised beam is explained in Section 3.4.

We also studied experimentally the influence of the position of the groove structure at a single incident wavelength of 632.8 nm under radially polarised illumination. The groove is placed at locations where the node of the standing wave pattern appears while keeping its depth and width constant. A high intensity hot spot is observed in the optical axis of the lens, which moves further away from the lens surface as the groove diameter is increased, an effect that was predicted by our numerical calculations discussed in Section 4.2. Figures 4.15(a) to (d) show the numerical simulation results of the movement of the hot spot as a function of the groove diameter. Figures 4.15(e) to (h) show the SEM images of plasmonic lenses with the corresponding groove diameter, $d_1$ of 400 nm, 1000 nm, 1600 nm and 2200 nm, respectively. The efficiency of
Figure 4.14  Simulated phase distribution of the longitudinal components in the focal plane of a far-field plasmonic lens for (a) $\lambda_1$ and (b) $\lambda_2$. Simulated cross-sections of the normalised intensity distributions in the focal region of a far-field plasmonic lens for (c) $\lambda_1$ and (d) $\lambda_2$; Experimental cross-sections of the normalised intensity distributions in the focal region of a far-field plasmonic lens for (e) $\lambda_1$ and (f) $\lambda_2$; Polarisation orientation is indicated by arrow in the top left corner.
the scattering by the groove is drastically reduced when the groove is moved to the antinode position of the interference pattern of the standing waves. This observation is consistent with a previous report [192]. The simulation of the axial shifting of the focal spot as a function of the groove diameter was corroborated by the SNOM experiment, as shown in Figure 4.15(i). Simulation results of the movement of the focal spot away from the lens surface as well as the increase in intensity at the focal plane when the groove position is changed were confirmed by the experimental results. As the focal spot shifts away with respect to the exit surface of the plasmonic lens, the increase in the constructive interference along the axial direction leads to an elongated focal spot. Table 4.1 summarises the lateral FWHM measured from the SNOM images at the focal plane as a function of the groove diameter.

<table>
<thead>
<tr>
<th>Focal Length (in nm)</th>
<th>d1 = 400 nm</th>
<th>d1 = 1000 nm</th>
<th>d1 = 1600 nm</th>
<th>d1 = 2200 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Simulation</td>
<td>370</td>
<td>635</td>
<td>880</td>
<td>1150</td>
</tr>
<tr>
<td>Experiment</td>
<td>300 ± 50</td>
<td>600 ± 50</td>
<td>800 ± 50</td>
<td>1200 ± 50</td>
</tr>
<tr>
<td>FWHM of the focal spot</td>
<td>0.39 λ1</td>
<td>0.41 ± 0.01 λ1</td>
<td>0.41 ± 0.01 λ1</td>
<td>0.42 λ1</td>
</tr>
<tr>
<td></td>
<td>0.40 λ1</td>
<td>0.43 ± 0.01 λ1</td>
<td>0.45 λ1</td>
<td>0.45 ± 0.01 λ1</td>
</tr>
</tbody>
</table>

Table 4.1 Focusing performance of the far-field plasmonic lens with different groove diameters, λ1 = 632.8 nm.
Simulated electric energy distributions (a,b,c,d) and the corresponding SEM images of the far-field plasmonic lens (e,f,g,h) in the vicinity of the far-field plasmonic lens when the groove diameter $d_1$ is 400 nm, 1000 nm, 1600 nm and 2200 nm, respectively. The inclined dashed line in the top figures indicates the shift of the focal length. (i) Comparison between the axial distribution of the normalised transmission intensity of the far-field plasmonic lens obtained by experimental measurements (data points) and simulated results (solid lines). The peaks axially move to larger $z$-values as the groove diameter increases, indicating the lift of the focal position away with respect to the exit surface of the plasmonic lens or an increased focal length. The wavelength under study is $\lambda_1 = 632.8$ nm.
4.4 Chapter conclusions

In this chapter, the nanoscale focusing of two different wavelength of light to the same focal plane of 1.2 μm by a far-field plasmonic lens was investigated numerically and experimentally. This result was achieved by optimising the concentric groove structures for the dispersion control of dual-wavelength of SPs and hence a tunability in the focal length. The plasmonic lens can be operated at linearly and radially polarised illuminations. The very compact design of a dual-wavelength far-field plasmonic lens could potentially be employed in lab-on-chip applications in place of a normal achromatic high-NA objective lens. The focusing performance of the demonstrated plasmonic lens is superior to conventional achromatic lens, although the power throughput of the former is 13% which can be improved to 50% by employing 5-ring plasmonic lens. First, a better resolution of $\lambda/3$ can be achieved, whereas the diffraction-limited resolution of $\lambda/2$ can only be realised even by a high NA lens. In addition, the dominant longitudinally polarised fields in the focal region are highly desired for applications such as single molecules imaging, particle acceleration, second harmonic generation and so on. The demonstrated far-field plasmonic lens, with an improved resolution compared to its counter parts like microlens array [201], can find potential applications in stimulated emission depletion imaging systems [168] and ultra-high density optical data storage [156, 166].
Chapter 5

Dual-wavelength far-field plasmonic lenses for stimulated emission depletion systems

5.1 Introduction

The field of nano-optics is by no means limited to technological applications and instrument design, such is the case in nanofocusing performances of nano lenses. To further improve the resolution of a far-field plasmonic lens, we propose dual-wavelength far-field plasmonic lens based stimulated emission depletion (STED) system. In this chapter, we present the most practical application that arise from the discovery of dual-wavelength far-field plasmonic lens in super-resolution microscopy, or precisely in STED, which relies on a purely physical concept of fluorescence depletion as discussed in Chapter 2.

The chapter is organised as follows. Section 5.2 of this chapter focus on the theoretical point spread function (PSF) analysis on the intensity distribution at the focal plane of a dual-wavelength far-field plasmonic lens. Section
5.3 deals with the experimental implementation of dual-wavelength far-field plasmonic lens based STED system. Generation of a doughnut focal spot by an circularly polarised first-order vortex (FOV) beam incident on a plasmonic lens is demonstrated in this section. Selection of a proper dye molecule and the observation of depletion of its fluorescence by overlapping two Gaussian beams, one being the incident beam and the other depletion, is discussed here. Effective PSF obtained from our proposed STED system is estimated numerically in Section 5.4 with the concluding remarks mentioned in Section 5.5.

5.2 Theoretical calculations

In a typical STED microscopic system, a Gaussian shaped excitation focal beam is overlapped with a doughnut-shaped focal spot with a zero intensity at the focal center for fluorescence depletion. Our proposal is to generate these two spots at the same focal plane of 1.2 μm by a dual-wavelength far-field plasmonic lens and incorporate them in a STED system with continuous wave beams [134]. It has been shown in Chapter 4 that a Gaussian focal spot can be produced by a dual-wavelength far-field plasmonic lens under a radially polarised illumination at \( \lambda_1 \). In this section, we theoretically show the generation of a doughnut focal spot by the same lens under a circularly polarised FOV illumination at \( \lambda_2 \). Scheme of the configuration for the same is shown in Figure 5.1.

The field calculations for the excitation beam (\( \lambda_1 \)) and its corresponding intensity distributions are shown in Figures 5.2(a) and (c), respectively. The field calculations for the depletion beam (\( \lambda_2 \)) and its corresponding intensity distributions are illustrated in Figures 5.2(b) and (d), respectively, under
5.3 Experimental implementation

5.3.1 Doughnut focal spot generation by a dual-wavelength far-field plasmonic lens under circularly polarised first-order vortex beam illumination

Scanning near-field optical microscope (SNOM) is used to validate and experimentally confirm the theoretical findings about the generation of doughnut shape focal spot by plasmonic lens. The orbital angular momentum of optical vortices has widely been studied [202, 203]. Excitation of SPs using cylindrical vector beams [204] or circularly polarised beams has been
experimentally demonstrated and investigated in many applications such as tightly focusing [205], spin-orbital angular momentum interaction [206], high resolution imaging [207], circular polarisation analysers [89], etc. It is possible to manipulate SPs by using phase modulated incident beams [208]. In this section, we discuss the excitation of the far-field plasmonic lens using a circularly polarised FOV beam and the subsequent generation of a doughnut focal spot.

The experimental set up for the characterisation of a plasmonic lens is the same as in Figure 4.10. Additional optical components are inserted in the beam path to generate circularly polarised FOV beam. A linearly polarised incident beam is converted to circular polarisation by a quarter wave plate oriented at 45 degree with respect to the polarisation direction of the beam. Subsequently, circularly polarised beam is allowed to pass through a vortex
phase plate with a topological charge, $l = 1$. This is focused by an objective lens with a numerical aperture (NA) of 0.7 to the back focal plane of the plasmonic lens. There is no direct propagating beam near the beam center owing to the central dark region of the incident optical vortex beam. The interference of the SP waves propagating towards the geometric centers of the vortex beam from all azimuthal directions results in a null of the longitudinal field components, thereby generating a doughnut-shaped distribution. The central dark spot is stable throughout the propagation away from the surface, generating a doughnut focal spot at 1.2 $\mu$m from the lens surface and is observed numerically for $\lambda_2$ (Figure 5.3(b)). To verify the central dark spot in the focal plane, the experimental full width at half maximum (FWHM) of the field distribution is measured by SNOM at the focal plane of 1.2 $\mu$m as 0.53 $\pm$ 0.01 $\lambda_2$ (Figure 5.3(c)). Figure 5.3(a) shows the spiral phase distribution of the vortex beam in the focal region of the far-field plasmonic lens for $\lambda_2$.

### 5.3.2 Fluorescence depletion

The key point in a plasmonic lens based STED system is to find the proper dye molecule which has a strong emission at 632.8 nm and a tail at 750 nm. This leads us to use the commercially available, Atto 647 dye. Absorption and emission spectrums of Atto 647 dye are shown in Figures 5.4(a) and (b), respectively. The absorption peak at 632.8 nm and the clear tail at 750 nm are indicated by arrows. For understanding the depletion process, we focused the two beams into an aqueous solution of the fluorophore Atto 647 dye using an oil immersion lens (NA = 1.4) and directed the fluorescence to a single photon photo multiplier tube (PMT). We placed the dye molecules on a cover slip and then spatially overlapped the excitation and depletion beams (both Gaussian). Intensity of the depletion beam is increased from 0 to 240 mW. The depletion
Figure 5.3  (a) Simulated phase distribution of the longitudinal components in the focal plane of a far-field plasmonic lens. (b) Simulated and (c) Experimental cross-sections of the normalised intensity distributions in the focal region of a far-field plasmonic lens for $\lambda_2$. 
Figure 5.4  (a) Absorption and (b) emission characteristics of Atto 647 dye suitable for STED imaging. Arrows indicate the absorption peak at 632.8 nm and the tail at 750 nm in (a) and (b) respectively.

curve has been obtained as in Figure 5.5. At a depletion power of 50 mW, the fluorescence is reduced by half.

5.4 Results predictions

There are two factors which determines the resolution of a STED system: STED laser intensity and the saturation intensity of the fluorophore. The latter is a function of the STED laser wavelength and the lifetimes of the ground and excited states of the fluorophore. The continuous wave (CW)
Figure 5.5 Fluorescence depletion of Atto 647 dye in STED microscopy as a function of the depletion laser power. Stimulated emission becomes the dominant process after increasing the depletion laser power more than a critical depletion threshold.

Power required to produce depletion is given by Ref [209],

$$P_S = \frac{A \cdot h c}{\lambda_{STED} \cdot \sigma \cdot \tau_{fl}}$$  \hspace{1cm} (5.1)

where \(c\), \(h\) and \(A\) denotes the speed of light, Planck’s constant and doughnut area, respectively; \(\sigma\), molecular cross-section for stimulated emission is \(3 \times 10^{-17} \text{cm}^2\); \(\tau_{fl}\), lifetime of the excited state is app. 3\(\text{ns}\). An oil immersion objective lens of \(NA = 1.4\) yields \(A \sim 0.5 \times 10^{-9} \text{cm}^2\).

The spot diameter can be further squeezed by applying a greater STED power, \(P\). The resolution limit of a STED system can be mathematically described as [210],

$$d = \frac{\lambda_{exc}}{(2NA(1 + P/P_S)^{1/2})}$$  \hspace{1cm} (5.2)
The change in resolution due to the introduction of a far-field plasmonic lens in a STED system is theoretically calculated. The doughnut area of a plasmonic lens at 750 nm excitation beam, is measured as $1.1 \times 10^{-9} \text{cm}^2$ (Figure 5.3(c)). From our depletion experiment (Figure 5.5), $P_s$ is estimated to be 50 mW. Since $P_s$ and $A$ have a linear relation as per equation 5.1, the saturation power required for a plasmonic lens based STED system to achieve depletion can be calculated and is estimated to be 2.2 times more than that required for a STED system with NA of 1.4.

The effective NA of the far-field plasmonic lens is calculated using the formula,

$$NA = \frac{0.6 \times \lambda_1}{\text{FWHM}}$$  \hspace{1cm} (5.3)

where, $\lambda_1$ is the excitation wavelength and FWHM of the focal spot of a far-field plasmonic lens is 290 nm (Figure 4.14(e)). From this, effective NA of a plasmonic lens is determined as 1.31. Substituting these values in equation 5.2, with $P = 400$ mW, the theoretical maximum resolution that can be attained with a plasmonic lens enabled STED system is $\lambda_{exc}/6$. Using this relation, intensity distribution of the PSFs at different depletion beam powers are shown in Figures 5.6(a) to (c). Figures 5.6(d) to (f) are cross-sections of the intensity distributions corresponding to Figures 5.6(a) to (c). The calculated FWHM values obtained by a far-field plasmonic lens, as a function of the depletion beam power, are compared in Figure 5.7 with those obtained for objective lenses of two different NA. The results obtained shows an improvement of resolution from $\lambda_{exc}/3$ for a dual-wavelength far-field plasmonic lens to $\lambda_{exc}/6$ for a plasmonic lens based STED system.
Figure 5.6 The calculated intensity distribution of the PSF in the focal region for the dual-wavelength plasmonic lens based STED system at the depletion beam power of (a) 10 mW, (b) 200 mW and (c) 400 mW. (d) to (f) are the cross-sections of the intensity distributions corresponding to (a) to (c).
Figure 5.7  Comparison of the calculated FWHM values as a function of the depletion beam power between a STED system, when operated with NA = 1.2 and NA = 0.95, with a plasmonic lens based STED system.
5.5 Chapter conclusions

In this chapter the effective resolution of a plasmonic lens based STED system was theoretically calculated. Generation of a doughnut focal spot at the same focal plane as the Gaussian focal spot with different incident polarisations, for the two chosen wavelengths was experimentally demonstrated using a dual-wavelength far-field plasmonic lens. This makes the proposed plasmonic lens an ideal choice for incorporating in STED system. We have theoretically shown that the effective PSF of our proposed system can reach as small as $\lambda_{exc}/6$. The improved performance of a plasmonic lens based STED system in terms of its focal area reduction can be foreseen as a general approach which can be implemented in multiple ways especially in biological applications requiring super-resolution.
Chapter 6

Conclusions and perspectives

6.1 Thesis conclusions

The Excitation beam profiles play a significant role in surface plasmon (SP) manipulation and therefore they affect the focusing action of a nanostructure having plasmons associated with it. The research presented in this thesis theoretically and experimentally investigates the far-field focusing performance of plasmonic lenses under different incident polarisation conditions. A major outcome of this research is our design of a simple circular ring far-field plasmonic lens which can focus dual-wavelength of SPs to the same far-field focal plane. In this thesis, we proposed the idea of incorporating a dual-wavelength far-field plasmonic lens in a stimulated emission depletion (STED) system. The important achievements of this thesis towards the development of the dual-wavelength plasmonic lens based STED system are outlined below:

1. Focal field components of an objective lens play an important role in determining the excitation efficiency of the plasmonic lens. The three dimensional intensity distributions of the dominant transverse field focal components of a low numerical aperture (NA) objective lens were
characterised using an in-plane polarisation sensitive scanning near-field microscopic (SNOM) probe.

2. The experimental demonstration of super-resolved pure-transverse focal fields through focusing an azimuthally polarised first-order vortex (FOV) beam could be useful for a variety of high-resolution microscopic applications. Super-resolved focal fields were observed with a 31% reduced focal area compared to that of a tightly focused circular polarisation determined by the full width at half maximum (FWHM). The results presented open the way up for potential applications requiring sub-wavelength resolution and pure-transverse fields such as high-density optical data storage and high-resolution microscopy.

3. We investigated theoretically and experimentally the polarisation properties of an Ag NW placed on a glass substrate. The investigation results proved the existence of two orthogonal polarisation components of the propagating plasmonic modes along the NW. Our results constitute a comprehensive polarisation study of the propagating plasmonic mode of an Ag NW on a dielectric substrate.

4. The major outcome of this thesis is a dual-wavelength far-field plasmonic lens which can realise nanoscale focusing of SPs at two different wavelengths, 632.8 nm (\(\lambda_1\)) and 750 nm (\(\lambda_2\)) to the same far-field focal plane. The far-field plasmonic lens, consisting of an annular slit and a concentric groove and capable of focusing dual-wavelength of light to the same focal plane, was characterised by a SNOM probe and was demonstrated to work under linearly and radially polarised illuminations. We used an out-of-plane sensitive SNOM probe for characterising the dominant \(E_Z\) components at the focal plane of our nanofocusing lens. A splitting of the focal spot was observed when it was illuminated by a linearly polarised wave. However, a sharp focal spot was obtained
with radial polarisation due to better SP coupling efficiency. The experimental FWHM of the lateral intensity distribution at the focal plane of 1.2 \( \mu \text{m} \) was measured as 0.45 \( \pm \) 0.01 \( \lambda_1 \) and 0.45 \( \pm \) 0.01 \( \lambda_2 \), for the two incident wavelengths respectively, under radial polarisation excitation. Focal length of this lens can be modulated by changing the groove diameter. This tunable far-field nanoscale focusing by the dual-wavelength plasmonic lens makes it an inevitable device in the nanophotonics community.

5. To further enhance the resolution of a dual-wavelength far-field plasmonic lens we have proposed and theoretically investigated a dual-wavelength plasmonic lens based STED system. We have theoretically demonstrated and experimentally verified the generation of a doughnut focal spot by a dual-wavelength far-field plasmonic lens under circularly polarised FOV excitation. For the conditions of our proposed system, Atto 647 dye was chosen as the appropriate fluorophore. Theoretical predictions were shown to have an improved effective point spread function (PSF) of \( \lambda/6 \) for our proposed system, which would be greatly beneficial for many super-resolution applications.

To conclude, this PhD thesis has focused on developing a nanoscale device which can focus dual-wavelength of light without suffering from achromatic aberration. Our work provides the first experimental demonstration of a plasmonic lens capable of far-field focusing of dual-wavelength of light to the same focal plane. This is omitted in current plasmonic focusing devices but is urgently demanded for a variety of research topics requiring far-field, sub-wavelength and broadband operations. Polarisation studies of far-field plasmonic lenses have been undertaken realising multiple ways of manipulating their focusing action. We have shown the ability of focal splitting and bright single spot generation at the focus of a dual-wavelength plasmonic lens just by
tuning the excitation beam polarisation. This idea can put forth the possibility of an optical controlled SP device which could be useful for manipulating the electronic states of a fluorescent sample at the focus and hence their associated emission properties. Our promising theoretical calculations on the dual-wavelength plasmonic lens based STED system provide valuable insight and an important basis for numerous practical applications.

6.2 Future outlook

In this section, we will briefly highlight future directions which directly stem from this work. Our intention is to give a glimpse of possible new research perspectives for forthcoming investigations.

With the successful theoretical demonstration of the dual-wavelength plasmonic lens based STED system, a promising extension of this research would be the experimental realisation of it. Three dimensional nanoscale focusing of SPs by a dual-wavelength far-field plasmonic lens has been demonstrated at 1.2 $\mu$m from the lens surface. This forms a major challenge for any experiments involving plasmonic lens, as a sophisticated method is required with precise control of this distance between the lens and the sample under consideration. Our experimental set up involving a nanometric precision stage to keep the sample at the focus of a plasmonic lens is currently under development.

A successful experimental realisation of a dual-wavelength plasmonic lens based STED system is expected to generate exciting advances in the fields of super-resolution imaging, ultra-high density optical data storage, multi-color super-resolution nano lithography, and optical trapping.
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