FOCUS ENGINEERING WITH SPATIALLY VARIANT
POLARIZATION FOR NANOMETER
SCALE APPLICATIONS

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FOCUS ENGINEERING WITH SPATIALLY VARIANT POLARIZATION FOR
NANOMETER SCALE APPLICATIONS

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ABSTRACT

FOCUS ENGINEERING WITH SPATIALLY VARIANT POLARIZATION FOR NANOMETER SCALE APPLICATIONS

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Spatially variant polarization has stimulated continuous research interests due to its peculiar properties in focusing and surface plasmon excitation, providing broad applications in optical data storage, nano-fabrication, particle trapping, high resolution microscopy and metrology. In this dissertation, focus shaping, three dimensional (3D) state of polarization control, and plasmonic focusing with spatially variant polarization are investigated and demonstrated both theoretically and experimentally.

This research shows that 3D flattop focusing with extended depth of focus and optical bubble can be obtained using the combination of generalized cylindrical vector beams and diffractive optical element. Through combining the electric dipole radiation and the Richards-Wolf vectorial diffraction method, the input field at the pupil plane of a high numerical aperture objective lens for generating arbitrary three dimensionally polarization at the focal point with an optimal spot size can be found analytically by solving an inverse problem.
In addition to focusing with high numerical aperture lens, spatially variant polarization has great advantage in surface plasmon focusing. Optimal plasmonic focusing can be achieved through matching the polarization symmetry of a radially polarized illumination to axially symmetric dielectric/metal plasmonic lens structures. Three types of plasmonic lens have been studied in this research. Experimental realization of the nondiffracting evanescent Bessel beam generation via surface plamson resonance excitation on homogeneous metallic thin film with radially polarized beam illumination is first demonstrated. Then, plasmonic lens with annular rings under radial polarization illumination is studied. It is found that higher field enhancement factor can be achieved with increasing number of rings in the plasmonic lens. Finally, an apertureless near-field scanning optical microscope probe under radial polarization illumination is numerically studied with 3D finite element method model. The field distribution with a full-width-half-maximum as small as 10 nm and intensity enhancement of five orders of magnitude can be achieved with 632.8 nm optical excitation. Preliminary experimental results using Raman spectroscopy with the designed apertureless tip confirmed the field enhancement through comparing the near field and far field signals.
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<td>2D</td>
<td>Two Dimensional</td>
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<tr>
<td>3D</td>
<td>Three Dimensional</td>
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<tr>
<td>CV</td>
<td>Cylindrical Vector</td>
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</tr>
<tr>
<td>DOE</td>
<td>Diffractive Optical Element</td>
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</tr>
<tr>
<td>DOF</td>
<td>Depth of Focus</td>
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<td>FWHM</td>
<td>Full Width at Half Maximum</td>
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<tr>
<td>NA</td>
<td>Numerical Aperture</td>
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<tr>
<td>NSOM</td>
<td>Near Field Scanning Optical Microscope</td>
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<tr>
<td>SPE</td>
<td>Spiral Phase Element</td>
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CHAPTER 1

INTRODUCTION OF CYLINDRICAL VECTOR BEAMS

In general, the polarization of light can be classified into two categories. The first type has spatially homogeneous distribution in terms of the state of polarization for the light wavefront. This type of polarization, including linear, circular and elliptical polarization, is most familiar to the optical community. Contrarily, the second type is the so-called spatially variant polarization which has spatially inhomogeneous polarization distribution. Traditionally, the second type of polarization was considered as a nuisance or an aberration in the optical design and has not drawn much research attention. However, there is an increasing interest in the spatially inhomogeneous polarization recently, mostly driven by the advances made in micro-fabrication techniques and theoretical modeling techniques that were not previously available. One example of such spatially inhomogeneous polarization that has attracted much of the interest is the so-called cylindrical vector (CV) beams [1]. CV beams are solutions of Maxwell’s equations that obey cylindrical symmetry both in amplitude and polarization.
1.1 Generation of CV Beams

CV beams can be divided into radial polarization, azimuthal polarization and generalized cylindrical polarization, according to the actual polarization pattern (Fig. 1.1). One can use two cascaded half-wave plates to conveniently convert a radial polarization or azimuthal polarization into a generalized CV beam, or vise versa [2]. Recently, there is a strong interest in these beams due to their peculiar properties, especially when focused under high numerical aperture (NA) conditions. Many techniques to generate radially polarized beams or azimuthally polarized beams have been reported, including inserting specially designed elements into laser resonators [3, 4], such as conical elements [5-7], complex Brewster-type windows [8], polarization selective mirrors [9, 10], c-cut Nd:YVO₄ crystals [11], liquid crystal gels [12], or phase elements which allow for significant mode discrimination [13]. Other methods for obtaining CV beams include using computer-generated spatially variant dielectric or metal strip subwavelength grating [14], space-variant inhomogeneous media [15], or diffractive optical element (DOE) interferometer [16].

Figure 1.1 Radial polarization, azimuthal polarization and generalized CV polarization.

\[
\begin{align*}
\vec{E}(r,\phi) &= P(r)e_r \\
\vec{E}(r,\phi) &= P(r)e_{\phi} \\
\vec{E}(r,\phi) &= P[\cos\phi_0 e_r + \sin\phi_0 e_{\phi}] 
\end{align*}
\]

Radial polarization  Azimuthal polarization  Generalized cylindrical polarization
Radially polarized beam can be generated by superposition of two orthogonal linearly polarized Hermite Gaussian beams [17,18]. This technique uses the coherent summation of two orthogonally polarized TEM\textsubscript{01} and TEM\textsubscript{10}. The mathematical expression of a linearly polarized TEM\textsubscript{01} and TEM\textsubscript{10} beam is

\begin{align}
E_{01}(x, y) &= \frac{E_{0}}{2W} \exp\left[-\frac{(x^2 + y^2)}{W^2}\right]e_x, \\
E_{01}(x, y) &= \frac{E_{0}}{2W} \exp\left[-\frac{(x^2 + y^2)}{W^2}\right]e_y,
\end{align}

where \(W\) is the beam waist and \(e_x\) (\(e_y\)) is a unit vector along the \(x\) (\(y\)) axis, simple vector addition yields a total field given by

\begin{equation}
E_x(x, y) = E_{01}(x, y) + E_{01}(x, y) = \frac{E_{0}}{2W} \exp\left[-\frac{(x^2 + y^2)}{W^2}\right](xe_x + ye_y),
\end{equation}

This equation indicates that the polarization state is radial and the intensity distribution is also radially symmetric. The generated radially polarized beam is shown in Fig. 1.2. The other similar interferometric technique is the interference of two opposite hand circularly polarized Laguerre-Gaussian (LG) beams of opposite topological charges [17,19]. Right and left handed circularly polarized beams are also orthogonal polarizations that can be used to create a radially polarized beam. Both techniques are insensitive to radially symmetric aberrations on the input beam (e.g. focus and spherical aberration of all orders). A critical requirement for both methods is stable alignment.
Another powerful and flexible method to generate arbitrary vector beams is using spatial light modulator (SLM) [20-22]. The first SLM generates a pure phase pattern for the whole aperture. The second SLM and the $\lambda/4$ waveplates form a pure polarization rotator. The amount of rotation is controlled by the phase retardation of each pixel on the second SLM. Combining these two SLMs, arbitrary vector beams with relative phase can be obtained, including CV beams.

Several techniques to generate CV beams have been developed in our lab. In addition to using SLMs as mentioned above, we can obtain radially polarized beam with high stability and high efficiency through coupling a charge +1 vortex beam into a few-mode fiber [23], or via combining of a radial polarized and a spiral phase element (SPE). The setup is illustrated in Fig. 1.3. The combination of linear polarizer and a $\lambda/4$ waveplate is used to produce a circularly polarized beam as the input.
Figure 1.3 Generation of radially polarized beam with the combination of radial analyzer and SPE.

In a cylindrical coordinate system, a left hand circularly polarized beam with a planar wavefront can be expressed as

$$
E_{LHC} = P(r)(\vec{e}_r + j\vec{e}_\phi)/\sqrt{2} = P(r)[(\cos\varphi\vec{e}_r - \sin\varphi\vec{e}_\phi) + j(\sin\varphi\vec{e}_r + \cos\varphi\vec{e}_\phi)]/\sqrt{2}\\
= P(r)e^{\varphi_0}(\vec{e}_r + j\vec{e}_\phi)/\sqrt{2}, \quad (1.4)
$$

where $P(r)$ is the axially symmetric amplitude distribution of the beam [24]. Similarly, for right hand circularly polarization we have

$$
E_{RHC} = P(r)e^{-j\varphi}(\vec{e}_r - j\vec{e}_\phi)/\sqrt{2}, \quad (1.5)
$$

If we let a left hand circularly polarized beam pass a radial analyzer and a SPE with a charge of +1 then we can get

$$
E = [P(r)e^{-j\varphi}e_r/\sqrt{2}] \times e^+ j\varphi = P(r)e_r/\sqrt{2}, \quad (1.6)
$$

A pure radial polarization will be obtained. However, for many of the applications, such as those surface plasmon focusing applications that I will investigate in Chapter 5, the
radial polarizer can be spared if we are only interested in the phenomena caused by the radially polarized component.

![Figure 1.4 Polarization rotator consisting of two half-wave plates [2].](image)

Two cascaded half-wave plates can be used to conveniently convert a radial polarization or azimuthal polarization into a generalized CV beam, or vice versa [2]. Figure 1.4 shows the polarization rotator consisting of two half-wave plates. The Jones matrix of this polarization rotator can be shown as

\[
\begin{bmatrix}
\cos(2\Delta\phi) & -\sin(2\Delta\phi) \\
\sin(2\Delta\phi) & \cos(2\Delta\phi)
\end{bmatrix} = R(-2\Delta\phi),
\]

Unlike the rotation from a single half-wave plate, this rotation is independent of the initial polarization. The amount of rotation is determined by the angle \(\Delta\phi\) between the fast axes of the two half-wave plates. When \(\Delta\phi = \phi_o / 2\), a generalized CV beam can be obtained as shown in Fig. 1.1.

1.2 Applications of CV Beams

Owing to their unique properties, CV beams have found wide applications in physics, chemistry and biology, such as particle guiding or trapping [25-29], scanning optical
microscopy [30], lithography [31], laser cutting of metals [32,33], particle acceleration [34,35], and single molecule imaging [36,37].

The propagation and focusing properties of spatially inhomogeneous polarization beams remain of continued interest. When focused with a high NA objective, radially polarized light leads to a strong longitudinal electric field component in the vicinity of the focus. The relative contribution of longitudinal component can be enhanced by using annular aperture. Theoretical and experimental investigations of focusing an incoming radially polarized beam into an ultrasmall spot have been reported [38-40]. The spot size can be much smaller than the diffraction limit of focused spatially homogeneously polarized beams. Besides an ultrasmall focus, it has been shown that focal distribution with extended depth of focus (DOF) can be achieved with inserting a simple DOE in the optical path under radial polarization illumination [38]. Furthermore, using a very simple two-half-wave-plate polarization rotator, one can conveniently convert radial polarization into a generalized CV polarization that can be used to generate focal field distribution with flattop transversal profile [41].

Optimal plasmonic focusing can be achieved through a combination of radially polarized illumination and axially symmetric plasmonic structures [42-46]. Surface plasmon is an electromagnetic wave due to the interaction of light and metallic structures. As a wave phenomenon, surface plasmon can be focused using appropriate excitation geometry and metallic structures. Surface plasmon can be excited by TM polarized light that has electrical field in the plane of incidence. When spatially homogeneous TM polarization illuminates an axially symmetric dielectric/metal plasmonic lens, the surface plasmon field will cancel out at the geometric center of the device and hence provide no
field enhancement or focusing effect. Contrarily, if a radially polarized beam is used instead, unlike linearly polarized beams, the entire beam is TM polarized with respect to the interface, enabling surface plasmon excitation from all directions and homogeneous plasmon focusing through constructive interference of these plasmon waves. Owing to the strong spatial confinement and high field enhancement, this type of optimal plamonic focusing has important applications in near field optical imaging and sensing.

1.3 Overview of Work in this Dissertation

This research aims at exploring the generation and applications of spatially variant polarization, especially CV beams. The topics to be covered in this dissertation include:

- Three dimensional (3D) focus shaping;
- 3D state of polarization control;
- Experimental generation of CV beams;
- Optimal plasmonic focusing with radially polarized beam;
- Experimental realization of evanescent Bessel beam;
- Experimental confirmation of optimal plamonic focusing with annular metallic rings;
- Tip enhance Raman spectroscopy with apertureless near field scanning optical microscope (NSOM) probe.

In Chapter 2, a 3D focus shaping technique using the combination of cylindrical polarization with binary DOE is proposed. The energy density pattern at the vicinity of the focus can be tailored in three dimensions by appropriately adjusting the parameters of the CV beam illumination, NA of the objective lens and the design of the binary DOE. Focus with extended DOF that has both transversal and longitudinal flattop profile is
obtained. An optical bubble that has a total dark volume surrounded by high field distributions is also shown. Potential applications of this focus shaping technique are discussed.

In addition to 3D focus shaping, a new approach enables full control over 3D state of polarization and field distribution in the vicinity of the focal point of a high NA lens is proposed. 3D state of polarization control has important applications in particle trapping and manipulation, single molecular imaging, tip enhanced Raman spectroscopy and high resolution microscopy. An electric dipole situated at the focus of the lens mimics the prescribed focused spot with desired linear polarization. Through solving the inverse problem of finding the field distribution at the lens pupil plane due to dipole radiation, the input field for generating the desired 3D linear polarization with optimal focal spot can be obtained. Arbitrary 3D elliptical polarization can also be achieved by introducing a second electric dipole polarized in the orthogonal plane with respect to the first dipole.

In Chapter 4, basic concepts and dispersion properties of surface plasmon excitation are briefly introduced. Shorter effective wavelength and strong local field enhancement due to surface plasmon excitation and focusing are demonstrated.

Plasmonic focusing with metallic nanostructures is an effective approach to confine optical field into nanometric size, which breaks the diffraction limit of light. In Chapter 5, I explored three different metallic structures for nanofocusing of surface plasmons with radial polarization illumination. When radially polarized beam is launched into these plasmonic structures, the entire beam is TM polarized with respect to the dielectric/metal interface, providing an efficient way to generate highly focused surface plasmon through constructive interference and creating an enormously enhanced local field. An evanescent
nondiffracting Bessel beam can be obtained with a planar metallic/dielectric interface. Bessel beams are electromagnetic fields that do not suffer from diffractive spreading. It is also demonstrated that radial polarization can also be used to improve the excitation and focusing for an annular metallic ring plasmonic lens. From numerical simulations, the plasmonic focusing is clearly observed for single circular slit carved into metal thin film as well as plasmonic lens structures consists of multiple concentric rings (Bull’s eye). In the case of multiple concentric rings structures, if the location and the rings are chosen to satisfy the circular Bragg condition for plasmonic wavelength, it is found that as more rings are added to the plasmonic lens structure, the peak intensity of the plasmonic center gets stronger while the focal spot maintains approximately the same size. Plasmonic lens with conical shape under radial polarization illumination are numerically studied with a finite-element-method model. The field distribution with a full-width-half-maximum (FWHM) as small as 10 nm and intensity enhancement of five orders of magnitude can be achieved with 633 nm optical excitation.

In Chapter 6, radially polarized beam with high stability and efficiency is experimentally generated through coupling a +1 charge vortex beam into a few-mode fiber. I will report the first experimental confirmation of the evanescent Bessel beam generation via surface plasmon resonance excitation with radially polarized beam. The excitation of surface plasmon is confirmed by the observation of a narrow dark ring at the back focal plane. Two-dimensional (2D) intensity distributions at different distances from the sample surface are mapped by a collection mode NSOM to verify the nondiffracting and decaying nature of the evanescent Bessel beam. I also conducted the first experiments to verify the optimal focusing effect with the Bull’s eye plasmonic lens.
NSOM images reveal that the plasmonic waves indeed are generated from all directions and focus towards the center. It is found that as more rings are added to the plasmonic lens structure, the peak intensity of the plasmonic center gets stronger while the focal spot remains approximately the same shape. Enormous field enhancement and strongly confined plasmonic focusing is obtained at the end of an apertureless NSOM fiber probe tip. The enhancement is confirmed by exciting a Raman signal from silicon with the tip. Both near field and far field signals are collected. Strong enhancement at near filed is observed, enabling a significant reduce of the integration time compared with the previous cases with aperture probes.

1.4 Novelty of the Research

In this work, a 3D focus shaping technique using the combination of cylindrical polarization with binary DOE is proposed. Focus with extended DOF that has both transversal and longitudinal flattop profile is obtained. Optical bubble that has a total dark volume surrounded by high field distributions is also shown.

Arbitrary 3D state of polarization and field distribution control has been demonstrated for the first time. Different methods for 3D state of polarization control have been reported by other groups, but neither of them can control the field distribution simultaneously, which made their approaches difficult to be applied into areas such as tip enhanced Raman spectroscopy or 3D particle trapping and manipulation. I proposed a novel approach to control both the polarization and intensity distributions simultaneously by solving the input field in an inverse way.
This work demonstrates the first experimental confirmation of the evanescent Bessel beam generation via surface plasmon resonance excitation with radially polarized beam. The nondiffracting and decaying nature of the beam is verified with an aperture NSOM.

Optimal plasmonic focusing is experimentally demonstrated for a single circular slit carved into metal thin film as well as plasmonic lens structures consists of multiple concentric rings. Experimental results confirm that the field enhancement can be improved with more annular rings, while the spot size remains constant. Optimal plasmonic focusing using radially polarized illumination offers tremendous advantages in terms of the plasmonic focus shape, peak intensity and spatial localization.

Significantly enhanced localized electromagnetic field with a FWHM of 10 nm is obtained at the end of an apertureless NSOM probe tip with radially polarized excitation. More than 1000 times enhancement has been verified experimentally through collecting the Raman spectrum signal of silicon sample.
CHAPTER 2

3D FOCUS SHAPING

The capability of shaping the 3D focal intensity distribution and controlling the state of polarization within the focal volume is very important in high resolution microscopy, optical data storage, particle trapping and manipulation. Recently, there is an increasing interest in the focal spot control with radially polarized or azimuthally polarized beam and high NA lens. Focusing property of a generalized CV beams has not been fully explored. In this chapter, a 3D focus shaping technique using the combination of CV polarization with binary DOE is proposed. The high NA focusing properties of generalized CV beams are analyzed using the Richards and Wolf vectorial diffraction method. A generalized CV beam can be decomposed into radially polarized and azimuthally polarized components. The energy density pattern at the vicinity of the focus can be tailored in three dimensions by appropriately adjusting the parameters of the rotation angle of CV beam illumination, NA of the objective lens and the design of the binary DOE. Focus with extended DOF that has both transversal and longitudinal flattop profile is obtained. Optical bubble that has a total dark volume surrounded by high field distributions is also shown. The advantage of using generalized CV beams in laser cutting and machining is demonstrated. The application of optical bubble in stimulated emission depletion (STED) technique is discussed.
2.1 Proposed Setup and Modeling

The polarization pattern of a generalized CV beam is illustrated in Fig. 2.1. Each point of the beam has a polarization rotated by $\phi_o$ from its radial direction. For a radial polarization, $\phi_o=0$; for an azimuthal polarization, $\phi_o=\pi/2$. The electric field of a generalized CV beam can be decomposed into the combination of radial polarization and azimuthal polarization as

$$\vec{E}(r,\phi) = L(r)[\cos\phi_o \vec{e}_r + \sin\phi_o \vec{e}_\phi] ,$$

(2.1)

where $\vec{e}_r$ is the unit vector in the radial direction and $\vec{e}_\phi$ is the unit vector in the azimuthal direction. $L(r)$ is the pupil function denoting the relative amplitude of the field that only depends on radial position.

Figure 2.2 shows the proposed setup for the three dimensional focus shaping. The incident light is a generalized CV beam described by Equ. (2.1) with a planar wavefront over the input pupil. An aplanatic high NA objective produces a spherical wave converging to the focus of the lens. A binary DOE with three concentric regions (shown in Fig. 2.3) is placed in front of the lens to modulate the wavefront of the generalized CV
beams. Due to the symmetry and normal illumination, the phase modulation is identical for the radial and azimuthal polarization components. The NAs that correspond to the outer edge of the three regions are $NA_1$, $NA_2$ and $NA$ respectively, where the local transmittances are $T_1=1$, $T_2=-1$ and $T_3=1$. Thus the field distribution at the focus is given by

$$E = E_1 - E_2 + E_3,$$

where $E_1$, $E_2$ and $E_3$ are the focal field contribution from each annular zone that can be calculated as follows.

\[E = E_1 - E_2 + E_3, \quad (2.2)\]

Figure 2.2 Focusing of a generalized CV beam with DOE.

Figure 2.3 Binary DOE with three concentric regions.
The focusing property of a highly focused laser beam can be analyzed with the Richards and Wolf vectorial method [2, 47-49]. Due to the rotational symmetry of both the setup and illumination, the electric field distribution in the vicinity of the focal spot can be calculated in cylindrical coordinates. The focal field of a generalized CV contributed from each concentric region of the DOE can be written as

\[ \vec{E}(r, \phi, z) = E_x \hat{e}_z + E_r \hat{e}_r + E_\phi \hat{e}_\phi, \]  

(2.3)

where \( \hat{e}_z \) is the unit vector in the z direction, and [2]

\[ E_x(r, \varphi, z) = jA \cos \phi \int_{\theta_1}^{\theta_2} \sin^2 \theta \, P(\theta) \, L(\theta) \, J_0(kr \sin \theta) \, e^{jkz \cos \theta} \, d\theta, \]  

(2.4)

\[ E_r(r, \varphi, z) = A \cos \phi \int_{\theta_1}^{\theta_2} \sin \theta \cos \theta \, P(\theta) \, L(\theta) \, J_1(kr \sin \theta) \, e^{jkz \cos \theta} \, d\theta, \]  

(2.5)

\[ E_\phi(r, \varphi, z) = A \sin \phi \int_{\theta_1}^{\theta_2} \sin \theta \, P(\theta) \, L(\theta) \, J_1(kr \sin \theta) \, e^{jkz \cos \theta} \, d\theta, \]  

(2.6)

where \( \theta_1 \) and \( \theta_2 \) is the angular transition points determined by the inner and outer transition points of the corresponding concentric region of the binary DOE. For example, for the concentric ring with \( \pi \) phase shift (see Fig. 2.3), the integration limits are given by \( \theta_1 = \sin^{-1}(NA_1) \) and \( \theta_2 = \sin^{-1}(NA_2) \), respectively. \( P(\theta) \) is the pupil apodization function, \( k \) is the wave number and \( J_n(x) \) is the Bessel function of the first kind with order \( n \). \( L(\theta) \) is the relative amplitude of the field, which is assumed to be dependent on the radial position only. Due to the continuity requirement, the on-axis electric field of cylindrical polarization is zero. The actual distribution of this hollow center depends on how the cylindrical polarization is generated. For simplicity, the illumination for all the examples here is chosen to be a planar wavefront over the pupil, where
\[ L(\theta) = \begin{cases} 
1 & \text{if } \sin^{-1}(N_{ao}) \leq \theta \leq \sin^{-1}(NA) \\
0 & \text{otherwise} 
\end{cases}, \quad (2.7) \]

where NA\(_{ao}\)=0.2 for our simulations. This means that the center of the lens pupil aperture is blocked to simulate this dark hollow center. Since the focusing properties of CV beams are largely due to the high spatial frequency components, the effects on the field distribution by blocking the center are negligible.

### 2.2 3D Flattop Focusing with Extended DOF

Here, we want to obtain a focal spot with two properties: (a) flattop focusing both in longitudinal and transversal directions; (b) long depth of focus. First, the focal fields created by radial polarization and azimuthal polarization are mutually orthogonal and spatially separated. This property was exploited to create flattop transverse focus as reported in reference (2). If a generalized CV beam is used as illumination along with the binary DOE, one should be able to adjust the relative weighing of the radial and azimuthal components to create flattop profiles both in longitudinal and transversal directions. Second, in ref. (38), similar binary DOE was proposed to generate ultrasharp focus with extended DOF using radially polarized illumination. Essentially, the field contributed from the middle ring destructively interferes with the field contributed from the innermost and outermost ring creating a saddle in the focus and elongate the DOF. However, it should be possible to adjust the transition points of the middle ring to change the relative interference contribution and “fill” the saddle to create a flattop longitudinal field profile.

From equations (2.3) to (2.6), the field distribution functions of radial and azimuthal components include Bessel function \( J_1(x) \), which is zero when \( x=0 \). Therefore, they do
not contribute to the on-axis field distribution. It is clear that the only non-zero contribution for on-axis field comes from the $z$-component. Thus, relative axial energy density distribution will not change with the rotation angle $\phi_o$ because only radial component of the generalized CV beams contributes to it. Therefore, during the first step, I set $\phi_o=0^\circ$ and change the transition points $NA_1$ and $NA_2$ of the binary DOE to find a flat longitudinal field distribution with long DOF. This is done by a linear direct search through manually adjusting the two transition points with 0.01 increments in the transition points. Once this is achieved, then I obtain flattop transversal field distribution through adjusting the rotation angle $\phi_o$ of CV beam.

For example, here I assume an aplanatic objective lens that satisfies the Sine condition is used, and the NA of the lens is chosen to be 0.8. Under the Sine condition, the pupil apodization function is $P(\theta) = \sqrt{\cos(\theta)}$ [50]. First, I adjust $NA_1$ and $NA_2$ to obtain long depth of focus and flattop focusing on the axis. I found the parameters for the binary DOE to be $NA_1=0.31$, $NA_2=0.5$. Then, I change the rotation angle of the CV beam to achieve flattop focusing in transversal direction. The rotation parameter for CV beam is found to be $\phi_o=38^\circ$. Figure 2.4(a) shows the calculated result of energy density distribution in the vicinity of the focus. The corresponding axial and transversal energy density distributions are shown in Fig. 2.4(b) and Fig. 2.4(c). For all of the calculations, the length unit is normalized to wavelength $\lambda$, and the maximum energy density is normalized to unity. It can be seen that flattop profiles along both the transversal and axial distribution have been obtained. The energy in the main lobe volume is about of the total energy. The DOF defined by FWHM of the $|E|^2$ along the axial direction is calculated to be $5\lambda$. For comparison, the axial field distribution for linearly polarized
distribution is also calculated and shown in Fig. 2.4(b). The DOF is calculated to be $2.4\lambda$. Clearly, an extended DOF is achieved with CV polarization. This extended DOF partly is due to the destructive interference from the middle ring of the DOE. In addition, the vector projection used in vectorial diffraction theory gives larger weighing to the higher spatial frequency components in the calculation of $E_z$, as seen in equation (2.4). This apodization effect also contributes to the longer DOF.

I also explored objective lenses under other pupil apodization functions. For an objective lens that obeys the Helmholtz condition where $P(\theta) = |\sqrt{\cos(\theta)}|^{-3}$ [50], with $\text{NA}=0.8$ and $\text{NA}_o=0.2$, the parameters for the binary DOE are found to be $\text{NA}_1=0.35$, $\text{NA}_2=0.55$, and the rotation angle for the CV beam is $\phi_o=39.5^\circ$. Figure 2.5 shows the calculated results of focal field distribution. Again, a 3D flattop focusing with extended DOF is obtained. In this case, the DOF is calculated to be $5.8\lambda$. This even longer DOF is due to the further increase of the higher spatial frequency weighing due to the apodization function. In comparison to an objective lens that obeys Sine condition, the objective lens under the Helmholtz condition is better for flattop focusing with extended DOF. This is because that the input energy at the outer portion of the DOE plays much more important role in the axial in energy density distribution. Under Helmholtz condition, this portion of the CV beams is strongly enhanced. Moreover, the main lobe energy in Fig. 2.5 is about 62.08% of the total energy, which means that more energy is focused into the main lobe if a Helmholtz condition objective lens is used. However, the sidelobe is slightly increased.

The high sidelobe is mainly due to the annular illumination and the diffractions from the abrupt changes of the binary DOE which can be somewhat eased by properly
choosing design parameters and using grayscale DOE design. One such example is shown in Fig. 2.6, where I increased NA to 0.815, instead of the NA of 0.8 used in Fig. 2.5, and the angle of cylindrical polarization $\phi_0$ is found to be $40^\circ$. A flattop focus with slightly lower sidelobe is obtained. It is also possible to reduce the sidelobe by using grayscale (amplitude modulation) DOE to change the local amplitude transmittance. However, both methods also decrease the DOF. A compromise between the DOF and sidelobe needs to be made. The design parameters should be appropriately chosen according to specific applications. Furthermore, focal spot with longer depth of focus can be obtained with binary DOE divided into more concentric regions.

(a)
Figure 2.4 Energy density distributions in the vicinity of focus with pupil apodization function under Sine condition. (a) 3-D distribution; (b) Axial distribution; (c) Transversal distribution.
Figure 2.5 Energy density distributions in the vicinity of focus with pupil apodization function under the Helmholtz condition. (a) 3-D distribution; (b) Axial distribution; (c) Transversal distribution.
Figure 2.6 Energy density distributions in the vicinity of focus with a larger NA of lens. (a) 3-D distribution; (b) Axial distribution; (c) Transversal distribution.
2.3 Generation of an Optical Bubble

As shown in the previous section, the feasibility of adjusting the destructive interference from the middle ring to “fill” the saddle to create a longitudinal flattop profile. Through adjusting the destructive interference from the middle ring, it is possible to further increase the destructive interference to carve into the focus and generate an optical bubble which has a total dark volume surrounded by high field distributions. In this case, I limit our search to radial polarization with $\phi_0=0^\circ$, since this may allow one to create an ultrasmall dark volume. The objective lens is assumed to obey the Sine condition. Using the same manual search methods, the parameter for the binary DOE are found to be $\text{NA}_1=0.2$, $\text{NA}_2=0.65$ with $\text{NA}=0.8$ and $\text{NA}_0=0.2$. Essentially, the DOE has two concentric rings, instead of three. Numerical simulation results for this design are shown in Fig. 2.7. From this figure, it can be seen that a bubble-like focal field with a totally dark center is obtained. The optical bubble size defined by FWHM of the $|E|^2$ is calculated to be $2.01\lambda$ along the axial direction and $0.90\lambda$ along the transversal direction. A smaller optical bubble can be obtained with higher NA lens, or 4Pi focusing.
Axial distribution

| |E|^2 vs. z (\(\lambda\))

(a)

(b)
2.4 Applications and Discussions

Flattop focusing with extended DOF may have important applications in laser cutting of metals, particle acceleration, materials processing and microlithography. Laser cutting is one of the fastest growing processes in industrial manufacturing. Laser machining tools offer significant advantages in productivity, precision, part quality, material utilization and flexibility. During laser machining process, laser energy from a resonator is focused on a material in order to remove the materials. Laser beams that can be focused into a flattop spot will allow faster, high quality laser cutting with lower operating costs.

The laser machining efficiency of metals strongly depends on the polarization. Spatially homogeneous polarizations have substantial disadvantages. For linear
polarization, the laser-metal interaction depends upon the polarization orientation. The shape of laser cutting is directly related to how the linearly polarized beam is oriented with respect to the direction in which the cut is traveling. For circular polarization, these parameters are time averaged, neither optimized for minimum losses nor for maximum absorption. It has been pointed out that inhomogeneous types of polarization are much better in laser cutting [32,33]. In the case of cutting metals with a large aspect ratio of sheet thickness to width, the laser cutting efficiency for a radially polarized beam is shown to be 1.5-2 times larger than for TM-polarized and circularly polarized beams.

In order to increase the laser cutting efficiency and velocity, sharper focusing of incoming light is necessary for high-speed cutting of sheet steel. If linear or circular polarized light is used in laser cutting, the radiation intensity decreases quickly along the focal axis, giving rise to a small ratio of cutting depth to cutting width. On the contrary, using CV beams with flattop focusing and a long DOF, one may significantly increase the ratio of cutting depth to cutting width. Because of the threshold character of material removing, laser cutting works only when high densities of absorbed power exceed the threshold value. It is possible to adjust the beam power to make sure such threshold condition met by the power level in the main lobe only. Thus, the relative high sidelobe may not be a problem.

The optical bubble may find important applications in particle trapping and fluorescence microscopy [51, 52]. For example, the resolution of fluorescence microscopy is mostly determined by the extent of the fluorescence spot. In order to improve the spatial resolution of fluorescence microscopy, STED technique has been developed [51]. In the STED technique, a depletion pulse following an excitation pulse is
focused into a donut shape around the focus of the excitation beam. In the region where the focal field intensity of the depletion beam is above certain threshold intensity, fluorescence is inhibited. An optical bubble that has a total dark volume surrounded by high field distributions in three dimensions may be applied in the STED microscopy to improve the spatial resolution of fluorescence microscopy.
CHAPTER 3

3D STATE OF POLARIZATION CONTROL WITH OPTIMAL FOCAL SPOT

In this chapter I propose and demonstrate a new approach that enables full control over the 3D state of polarization and field distribution near the focal point of a high NA objective lens. By combining the electric dipole radiation and the Richards-Wolf vectorial diffraction method, the input field at the pupil plane of a high NA objective lens for generating arbitrary three dimensionally oriented linear polarization at the focal point with an optimal spot size is found analytically by solving the inverse problem. Arbitrary 3D elliptical polarization can be obtained by introducing a second electric dipole oriented in the orthogonal plane with appropriate amplitude and phase differences. An experimental setup using liquid crystal spatial light modulators is proposed for realization. 3D polarization control within a tight focus has important applications in single molecule imaging, tip enhanced Raman spectroscopy, high resolution optical microscopy, particle trapping and particle manipulation.
3.1 Introduction

Optical microscopy has been an indispensable tool in many scientific disciplines and industries owing to its nondestructive nature and the multi-dimensional information it provides. It utilizes a tightly focused optical field as a probe to interrogate the sample properties within the focal volume and generate the contrast for imaging. Hence, controlling the optical field properties within the focal volume plays a critical role in determining the function and performance of optical microscopy. For example, pupil plane apodization techniques have been developed to affect the spatial resolution of microscopy. The Gouy phase shift within the focal volume has been found to be important in determining the signal of coherent anti-Stokes Raman spectroscopy (CARS) [53] and third harmonic generation (THG) microscopy [54-56]. Spatial engineering of the focal intensity profile for a depletion pulse has been explored in the stimulation emission depletion (STED) microscopy to provide spatial resolution far beyond the diffraction limit [57,58].

Besides the intensity and phase distribution within the focal volume, polarization of the focal field is another important parameter that deserves attention. Many molecules and crystalline structures are anisotropic due to their specific spatial orientation, making polarization response a very sensitive contrast mechanism. For example, polarimetric imaging microscopy has been used to study the cellular organelle to infer nanoscale crystalline process as it occurs [59]. Magneto-optic (MO) Kerr imaging has been developed to study the domain structures and magnetization process of magnetic thin film [60]. A polarization dependent fluorescent pattern has been utilized for molecule orientation imaging [61]. Optical focus with prescribed linear polarization finds many
applications in tip enhanced optical near field imaging [62-64]. In principle, full control of the state of polarization in the focal plane could provide much richer information in optical microscopy and significantly expand its functionality. For example, laser beams with cylindrical polarization symmetry has been extensively studied recently to create strong longitudinal focal field component [1]. It has been shown that smaller focal spot can be generated with a highly focused radial polarization [39]. This optimal focusing can be explained by the concept of the time reversal of the electric dipole radiation. Vector point spread function (PSF) generation technique has also been demonstrated to generate linear, radial or azimuthal state of polarization in the focal volume [20]. Recently the generation of an optical “needle” that consists of almost pure longitudinal field and its applications in optical microscopy has been proposed and discussed [65].

However, so far the polarization control in the focal volume has been limited to a few special cases (x-, y-, z- or circularly polarized) in the plane that is transversal with respect to the optical axis. Control of the focal field with arbitrary 3D state of polarization has not been fully exploited. 3D state of polarization control at the focal point of a NA lens has recently been proposed using low-order azimuthal spatial harmonics of a linear polarization in the input field [66]. However, this method does not provide a full control over the field distribution in the vicinity of the focus. The state of polarization could quickly change and deviate from the desired state as the observation point moves away from the geometrical focus. The conversion efficiency from the input field into the desired state of polarization is low. In most of the situations, the desired state of polarization does not occur at the peak intensity of the focal field. Furthermore, due to the introduction of the spatial harmonics, the focal field is no-longer a diffraction limited
spot. These drawbacks severely limit the applicability of such method in practical optical microscopy.

Recent developments in nanofabrication and spatial light modulators (SLM) offer unprecedented level of light polarization engineering [14,20,67]. It becomes feasible to fully control the 3D polarization within the focal volume while maintaining the optimal diffraction limited spot. In this chapter, I propose a novel method that can generate arbitrary 3D state of polarization within an optimal focal spot. By combining the electric dipole radiation and Richards-Wolf vectorial diffraction method [47,48], the input field at the pupil plane of a high NA objective lens for generating arbitrary 3D oriented linear polarization at the focal point with an optimal spot size can be found analytically by solving the inverse problem. The corresponding input field to generate an arbitrary 3D elliptical state of polarization can be found using two electric dipoles situated at the focus and polarized at orthogonal planes with different phase and amplitude. An experimental setup using liquid crystal spatial light modulators (LC-SLMs) will also be proposed as a potential realization method.

3.2 Methods and Results

A 3D state of polarization control at the focal point of a high NA objective lens has been proposed using low-order azimuthal spatial harmonics to express the x-polarized input field [66]. However, this method does not provide a full control over the field distribution in the vicinity of the focus. The peak intensity region may be shifted out of the focal point where the state of polarization is supposed to be controlled. This is due to the fact that for x-polarized input beam, the conversion efficiencies from it into y-polarization and longitudinal polarization (z-polarization) after the refraction of objective lens are lower
than the conversion efficiency into itself. Thus, in order to achieve a higher ratio between y- and longitudinal polarization over x-polarization at the focus, the majority of the input power must be transferred to off axis x-polarization. One way to overcome this problem is to introduce an orthogonal polarization component to the input field to obtain a vector beam at the pupil plane. However, the spot size at the focus is still not optimized.

In order to be able to use 3D polarization control in practical optical microscopy, ideally we want to control the 3D state of polarization while maintaining the focal spot to be diffraction limited simultaneously. This requires us to systematically obtain the necessary input field at the pupil that can be focused into an optimal spot with arbitrary desired state of polarization. I propose to solve the problem in an inverse manner. The schematic setup for the proposed 3D state of polarization and field distribution control is shown in Fig. 3.1. An electric dipole situated at the focus of a high NA objective lens is aligned in the same direction as the desired polarization. The dipole radiation has the well known angular patterns of field strength and local polarization distribution. The radiation from this dipole is collected and collimated by an aplanatic objective lens and the field distribution at the lens pupil can be found in conjunction with Richards-Wolf vectorial diffraction theory. The field at the lens pupil plane will be a vector field in general with nonuniform amplitude distribution. If we use this field distribution in the pupil plane as illumination and reverse the propagation, the field of the electric dipole should be reconstructed up to the propagating components at the focal point, which should give the desired 3D polarization we began with. An optimal spot with desired 3D linear polarization will be achieved. Hence, such method should provide a systematic way of obtaining the field distribution at the lens pupil.
Figure 3.1 Schematics of proposed method. An electric dipole situated at the focus of a high NA lens is aligned in the direction of the desired 3D linear polarization, which mimics a radiation source to reconstruct the input field at the pupil for generating the desired polarization.

For an electric dipole polarized in xz plane with an angle $\theta_1$ measured from the optical axis z, the dipole radiation angle to point A, which is also the angle between OA and dipole orientation, $\theta'$ can be expressed as

$$\theta' = 2\sin^{-1}\left[\sqrt{(1 - \sin\theta\sin\theta_1\cos\varphi - \cos\theta\cos\theta_1)/2}\right],$$

(3.1)

where $\theta$ is the focusing angle between the beam and the optical axis, and $\varphi$ is the azimuthal angle. Then the dipole radiation field at point A will be

$$\vec{R}(\theta') = C\sin(\theta')\vec{a}_{\theta'},$$

(3.2)

where $C$ is a constant related to the dipole strength, and $\vec{a}_{\theta'}$ is the unit vector of the radiation field at point A. The angle $\varphi'$ between $\vec{a}_{\theta}$ and $\vec{a}_r$ is given by

$$\cos\varphi' = \frac{-\cos\theta'\cos\theta + \cos\theta_1}{\sin\theta\sin\theta'},$$

(3.3)

Then the dipole radiation field at point A can be decomposed into radial components $\vec{a}_r$ and azimuthal component $\vec{a}_\varphi$ on the spherical surface after the objective lens

$$\vec{E}_o(\theta, \varphi) = R(\theta')(\cos\varphi\vec{a}_r - \sin\varphi\vec{a}_\varphi),$$

(3.4)
Considering the bending effect, the field on the spherical surface can be expressed in the Cartesian coordinates of the image space as

\[
\vec{E}_o(\theta, \varphi) = R(\theta')[(\cos\varphi'\cos\theta\cos\varphi + \sin\varphi'\sin\varphi)\vec{i} \\
+ (\cos\varphi'\cos\theta\sin\varphi - \sin\varphi'\cos\varphi)\vec{j} + \cos\varphi'\sin\theta\vec{k}], 
\]

(3.5)

The input field \(\vec{E}_i(\theta, \varphi)\) can be written in Cartesian coordinate system of the pupil plane as

\[
\vec{E}_i(\theta, \varphi) = R(\theta')[(\cos\varphi'\cos\varphi + \sin\varphi'\sin\varphi)\vec{x} + (\cos\varphi'\sin\varphi - \sin\varphi'\cos\varphi)\vec{y}] ,
\]

(3.6)

Note that the field is expressed in terms of the refraction angle \(\theta\) in the image space and the azimuthal angle \(\varphi\). In order to express the input field in the pupil plane spatial coordination \((r, \varphi)\), the projection function of the objective lens needs to be considered [50]. For an objective lens that obeys sine condition \(r = f \cdot \sin \theta\), where \(r\) is radial position in the pupil plane and \(f\) is the focal length of the objective lens, the projection function from the \((r, \varphi)\) space to the \((\theta, \varphi)\) space is \(\sqrt{\cos \theta}\). Consequently, the input field at the lens pupil will be

\[
\vec{E}_i(r, \varphi) = \vec{E}_i(\theta, \varphi)/\sqrt{\cos \theta} ,
\]

(3.7)

where \(\theta = \sin^{-1}(r/f)\).

Now the desired illumination field at the pupil to generate the prescribed 3D linear polarization at the focus can be found as

\[
\vec{E}_i(r, \varphi) = R(\theta')[(\cos\varphi'\cos\varphi + \sin\varphi'\sin\varphi)\vec{x} + (\cos\varphi'\sin\varphi - \sin\varphi'\cos\varphi)\vec{y}]/\sqrt{\cos \theta}
\]

(3.8)
The Richards-Wolf vectorial diffraction method can be applied to verify whether this input field will produce a focal spot with the desired polarization. The electric field \( \vec{E} \) at any point \( P(r_p, \psi, z_p) \) in the vicinity of the focal point can be calculated by vectorial Debye integral as

\[
\vec{E}(r_p, \psi, z_p) = \frac{i}{\lambda} \int_0^{\theta_{\text{max}}} \int_0^{2\pi} \vec{E}_\theta(\theta, \varphi) e^{-ikr_p \sin \theta \cos (\varphi - \psi) - i k z_p \cos \theta} \sin \theta d \theta d \varphi , \tag{3.9}
\]

Here \( \vec{E}(r_p, \psi, z_p) \) is the electric field vector at point \( P \); \( \lambda \) is the wavelength of illumination; \( \theta_{\text{max}} = \sin^{-1}(NA) \), where \( NA \) is the NA of the aplanatic lens and is set to be 1 in the following calculations. \( \vec{E}_\theta(\theta, \varphi) \) is the field distribution after the refraction of the lens given by equation (3.5).

Let us consider two special examples of an electric dipole situated at the focus and polarized in different directions. If \( \theta_1 = 0 \), which means that the dipole is oscillating in longitudinal direction. By solving the inverse problem of finding the field distribution in the lens pupil plane due to the radiation of \( z \)-polarized dipole, one finds that the resulting field at the pupil is radially polarized (Fig 3.2). When focused with a high NA lens, radially polarized beam creates a strong longitudinal field in the vicinity of the focus [39]. If \( \theta_1 = 90^\circ \), which means that the dipole is oriented in \( x \)-direction, the resulting input field at the lens pupil plane is mainly linearly polarized in the \( x \)-direction (Fig. 3.3), which is expected.
Figure 3.2 Field distribution and polarization pattern at the lens pupil plane for generating desired 3D linear polarization at the focus. (a) Input field is radially polarized for generating longitudinally polarized field at the focus. (b) Projection of intensity and state of polarization distributions on three orthogonal planes in the focal region.
Figure 3.3 Field distribution and polarization pattern at the lens pupil plane for generating desired 3D linear polarization at the focus. (a) Input field is mainly x-polarized for generating x- linearly polarized field at the focus. (b) Projection of intensity and state of polarization distributions on three orthogonal planes in the focal region.
Figure 3.4 Input field at the lens pupil plane for generating desired 3D linear polarization at the focus. Amplitude and polarization distributions are shown.

Arbitrary 3D linear polarization can be obtained through changing the dipole orientation. For example, in order to produce a 45° linear polarization in xz plane, one can simply set $\theta_1=45^\circ$. The input field designed to produce the desired 3D linear polarization at the pupil plane $\vec{E}_i(r, \varphi)$ is given by equation (3.8). The amplitude and polarization patterns at the lens pupil plane are shown in Fig. 3.4. The input field is vector beam with spatially variant amplitude and polarization distribution. The field after the refraction of lens, $\vec{E}_o(\theta, \varphi)$, can be obtained by equation (3.5). By inserting $\vec{E}_o(\theta, \varphi)$ into equation (3.9), the field distribution in the vicinity of the focal point can be calculated. Fig. 3.5 illustrates the 3D slice projection of the focal spot in three orthogonal planes. Intensity and local polarization distributions are both shown. A homogeneous spot with uniform polarization is obtained in the vicinity of the focal point. The FWHM of the focused spot are 0.513\lambda in xz plane and 0.414\lambda in yz plane. The DOF is calculated
to be $0.966\lambda$. At the focal plane, the conversion efficiency from the input power to the main focal spot is 60.6%. Using the criterion given by Sales [68], for diffraction limited cases, the product of transverse size ($G_T$) and the axial size ($G_A$) should satisfy $G_T G_A \geq g$, where $g$ is approximately 0.47 for a circular aperture. In our case, using the average spot size in transverse plane, we have $G_T G_A = 0.45$. This value is slightly smaller than 0.47 is due to the non-uniform amplitude across the circular pupil. Nevertheless, this clearly demonstrates that a diffraction limited spot with the desired state of polarization has been obtained.

Figure 3.5 Projection of intensity and state of polarization distributions on three orthogonal planes in the focal region. The field at focus is $45^\circ$ linearly polarized in xz plane.
In order to generate 3D elliptical polarization, one can introduce a second electric dipole polarized in yz plane, with certain field strength ratio $\eta$ and phase shift $\phi$ with respect to the dipole oscillating in xz plane. For the second dipole situated at the focus and polarized in yz plane, the azimuthal angle is

$$\varphi_2 = \varphi - \pi/2 \ , \quad (3.10)$$

Radiation angle $\theta'_2$ and $\varphi'_2$ can be expressed as

$$\theta'_2 = 2 \sin^{-1}\left[\sqrt{(1 - \sin\theta\sin\theta_2 \cos\varphi_2 - \cos\theta \cos\theta_2)/2}\right] \ , \quad (3.11)$$

$$\cos\varphi'_2 = \frac{-\cos\theta'_2 \cos\theta + \cos\theta_2}{\sin\theta \sin\theta_2} \ , \quad (3.12)$$

The illumination after the refraction of lens due to the second dipole is

$$\vec{E}_{\theta_2}(\theta, \varphi) = \eta e^{i\phi} \times R\left(\theta'_2\right) [\left(\cos\varphi_2 \cos\theta \sin\varphi_2 - \sin\varphi_2 \cos\varphi_2\right)i]$$

$$+ \left(\cos\varphi_2 \cos\theta \sin\varphi_2 + \sin\varphi_2 \sin\varphi_2\right)j + \cos\varphi_2 \sin\theta k] \ , \quad (3.13)$$

Now the illumination field at the lens pupil plane due to the second dipole can be written as

$$\vec{E}_{\theta_2}(r, \varphi) = R\left(\theta'_2\right) [\left(\cos\varphi_2 \cos\theta \sin\varphi_2 + \sin\varphi_2 \sin\varphi_2\right)i]$$

$$+ \left(\cos\varphi_2 \sin\varphi_2 - \sin\varphi_2 \cos\varphi_2\right)j]/\sqrt{\cos\theta} \ , \quad (3.14)$$

This field will be coherently superimposed with the field computed with equation to generate the illumination that can create the desired 3D elliptical focal field polarization. As an example, I show the results for a 3D polarization state that its projected polarization is $45^\circ$ linear in xz plane, circular in xy plane and yz plane. The pupil illumination required to produce such polarization can be found by setting $\theta_1=45^\circ$, $\theta_2=90^\circ$, the field strength ratio $\eta=\sin(\theta_1)$ and phase shift $\Phi=\pi/2$ (shown in Fig. 3.6). The intensity and state of polarization distributions of the focused spot are projected to three
orthogonal planes as shown in Fig. 3.7. The FWHM of the focused spot are $0.468\lambda$ in xz plane and $0.470\lambda$ in yz plane. The DOF is calculated to be $0.938\lambda$. At the focal plane, the conversion efficiency from the input power to the main spot is 72.3%. The product of transverse and axial spot sizes is $G_TG_A=0.44$. Again, a diffraction limited spot has been obtained. In comparison, taking the best results given by the low-order harmonics method in Reference 66, the corresponding FWHMs of the focal spot obtained by are $0.522\lambda$, $0.703\lambda$ and $2.554\lambda$, and the power conversion efficiency is only 41.7%. The product of transverse and axial spot sizes is $G_TG_A=1.56$, more than three times higher than the criterion for diffraction limit.

Figure 3.6 Input field at the lens pupil plane for generating desired 3D elliptical polarization at the focus. Amplitude and polarization distributions are shown.
Figure 3.7 Projection of intensity and state of polarization distributions in the focal region to three orthogonal planes. The projected polarization of the field in the focus is circular in xy plane, 45° linear in xz plane and circular in yz plane.

3.3 Proposed Experimental Solutions

NA equal to 1 is used in the previous calculation. Such high NA can be achieved with reflective type of parabolic objectives [69]. This ensures that radiation of dipole emission into half of the space can be collected and recovered. The method also works for a refractive objective with lower collection angle. Fig. 3.8 shows the relationship between the dipole orientation and the polarization direction of the focused field with an aplanatic
lens of NA=0.95. There is a small deviation from the linear relationship. This is due to the fact that the contribution from the illumination edge has been cut off, which is mostly converted to the longitudinal component of the focused field in the vicinity of the focal point. Therefore, in order to obtain a focused field with desired 3D polarization using lower NA objective lens, one can simply choose a dipole with orientation angle slightly smaller than the desired inclination angle as the start to calculate the input field at the entrance pupil.

![Figure 3.8 Deviation from linear relationship with lower NA lens.](image)

In principle, the field necessary for 3D polarization control has nonuniform spatial distribution in polarization, magnitude and phase. Such complicated field requires full control of the spatial distribution of light field. This is becoming possible with the advances in nanofabrication and spatial light modulators. Here I propose one practical method using liquid crystal spatial light modulators (LC-SLMs). As shown in Fig. 3.9(a), a pure polarization rotator has been demonstrated by sandwiching a SLM between two
orthogonally oriented $\lambda/4$ waveplates [20-22]. The fast axis of the SLM has an angle of $\pi/4$ with respect to the two waveplates. The Jones matrix of this rotator is

$$T = R \left( -\frac{\pi}{2} \right) \left[ \begin{array}{cc} 1 & 0 \\ 0 & -j \end{array} \right] R \left( \frac{\pi}{2} \right) R \left( -\frac{\pi}{4} \right) \left[ \begin{array}{cc} e^{-j\varphi_{xy}/2} & 0 \\ 0 & e^{j\varphi_{xy}/2} \end{array} \right] R \left( \frac{\pi}{4} \right) \left[ \begin{array}{cc} 1 & 0 \\ 0 & -j \end{array} \right]$$

$$= -je^{-j\varphi_{xy}/2} R \left( \frac{\varphi_{xy}}{2} \right), \tag{3.15}$$

where $\varphi_{xy}$ is the spatially distributed retardation from the SLM. This polarization rotator is independent of the initial state of polarization. The amount of rotation is determined by the retardation of the SLM. Vector beams with amplitude distribution can be obtained using three SLMs (Fig. 3.9(b)). The first SLM generates a pure phase pattern of $\delta_{xy}/2$ for the whole aperture. The combination of the first pure polarization rotator and a linear polarizer provides the necessary amplitude modulation. Linearly polarized beam with desired amplitude distribution is obtained after the linear polarizer. The second pure polarization rotator then rotates the local polarization to the desired vector field direction. The Jones matrix of the system is

$$T = -je^{-j\varphi'_{xy}/2} R \left( \frac{\varphi'_{xy}}{2} \right) \left[ \begin{array}{cc} 1 & 0 \\ 0 & 0 \end{array} \right] \left( -je^{-j\varphi_{xy}/2} \right) R \left( \frac{\varphi_{xy}}{2} \right) \left[ \begin{array}{cc} e^{j\delta_{xy}/2} & 0 \\ 0 & e^{-j\delta_{xy}/2} \end{array} \right] \left[ \begin{array}{cc} 1 & 0 \\ 0 & 0 \end{array} \right]$$

$$= -j\cos \left( \frac{\varphi_{xy}}{2} \right) e^{-j\left(\varphi_{xy} + \varphi'_{xy} + \delta_{xy}\right)/2} R \left( \frac{\varphi'_{xy}}{2} \right), \tag{3.16}$$

The phase retardations $\varphi_{xy}/2$ and $\varphi'_{xy}/2$ due to the two pure polarization rotators are compensated by the first SLM.
Figure 3.9 Setup for generating vector beams with amplitude distribution. (a) Pure polarization rotator (PR) using SLM sandwiched between two orthogonally oriented $\lambda/4$ waveplates. F: fast axis. (b) Generation of arbitrary polarization and amplitude distribution by using a SLM and two pure polarization rotators.

In our method, the focused field has maximum intensity in the focal point, and the spot size is confined to near diffraction limit. This is very valuable for applications such as particle trapping and manipulation. For example, for a dielectric particle with higher dielectric constant than the ambient, the gradient force of the focused field tends to pull and trap the particle to the highest intensity region of the focal spot. Recently, there is a strong interest in manipulating and trapping anisotropic particles, either geometrical or
optical anisotropy, such as carbon nanotubes, nanorods, ferroelectric materials or birefringent crystals. If the tightly focused beam is linearly polarized, the nano- or micro-particles will be trapped and aligned in the plane of polarization. Arbitrary 3D linear polarization control makes it possible to manipulate these particles by changing the polarization of the tightly focused beam. The real time polarization control with SLMs allows real time control of the trapping force, and manipulates the particles in desired trajectory. If the focused beam is 3D elliptically polarized, the anisotropic particles will experience a spinning torque [70]. When the torque is big enough, it will cause the particle to rotate along the axis. The tightly focused beam with the desired 3D polarization provides strong alignment force or spinning torque for particle manipulation.

3.4 Conclusions

In conclusion, I have demonstrated arbitrary 3D state of polarization control with an optimal spot size in the vicinity of the focal point of a high NA lens. The input field for generating arbitrary 3D linear polarization is found by solving the radiation field of an electric dipole situated at the focus and aligned in the same orientation as the desire polarization. Through introducing a second electric dipole with appropriate phase and amplitude difference located at the focal point and polarized in an orthogonal plane, input field for creating arbitrary 3D elliptical polarization can be obtained. Vectorial diffraction method is applied to verify that the input fields solved in this way can be focused into the desired 3D state of polarization with optimal spot size. A homogeneous spot with uniform polarization is achieved in the vicinity of the focal point when the input field is focused with an aplanatic lens. This technique provides analytical methods to generate any 3D polarization with diffraction limited spot size. An experimental setup using liquid
crystal spatial light modulators (LC-SLMs) has been proposed as a potential realization method. The proposed focal field polarization control method may find important applications in single molecular imaging, tip enhanced Raman spectroscopy, high resolution optical microscopy, and particle trapping and manipulation.
CHAPTER 4
SURFACE PLASMON RESONANCE

Surface plasmons are collective oscillations of free electrons that can be excited by TM polarized light at dielectric/metal interface. Since its first observation by Wood in 1902 [71,72], surface plasmon has been widely applied in surface enhanced Raman spectroscopy, subwavelength optics [73-74], super-resolution imaging [75-77], nanolithography [78], high harmonic generation [79], waveguiding [80], near field imaging and sensing [81]. A new emerging field called plasmonics, which sets up a bridge connecting photonics and electronics, remains continuous interests to the optics and photonics community.

4.1 Physical Principles

Surface plasmon is free electron oscillation due to the interaction of light and metal [82]. As shown in Fig. 4.1, when light illuminates from a dielectric material into metal, using Maxwell boundary conditions at the dielectric-metal interface we will have

\[ k^x_d = k^x_m ; \quad k^z_d \varepsilon_m = k^z_m \varepsilon_d , \]

where \( k^x_d \) and \( k^z_d \) are x- and z- direction wave vectors in dielectric material, \( k^x_m \) and \( k^z_m \) are x- and z- direction wave vectors in metal, and \( \varepsilon_d \) is the dielectric constant of the dielectric. Also,
\begin{align}
(k_d^x)^2 + (k_d^z)^2 &= \varepsilon_d (\frac{\omega}{c})^2 ; \quad (k_m^x)^2 + (k_m^z)^2 &= \varepsilon_m (\frac{\omega}{c})^2 \\
\text{(4.2)}
\end{align}

Combining equations (4.1) and (4.2), we can obtain

\[ k_m^x = \frac{\omega}{c} \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}} = k_m^x + ik_m^z , \quad \text{(4.3)} \]

where \( k_m^x \) is the real part of \( k_m^x \) and corresponding to the surface plasmon polariton wavelength, while \( k_m^z \) is the imaginary part and determines the evanescent decay nature.

Assuming that the metal has dielectric constant \( \varepsilon_m = \varepsilon_m^\prime + i\varepsilon_m^\prime \), where \( \varepsilon_m^\prime \) and \( \varepsilon_m^\prime \) are real,

\[ k_m^x = \frac{\omega}{c} \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}} , \quad \text{(4.4)} \]
Figure 4.1 (a) Surface plasmon polaritons along the dielectric-metal interface excited with a TM polarized light. (b) Decaying nature of surface plasmon polaritons.

The wavelength and 1/e decay length of surface plasmon polariton is

\[ k_m^* = \frac{\omega}{c} \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d} \frac{\varepsilon_m \varepsilon_d}{2 \varepsilon_m + \varepsilon_d}}, \]  

(4.5)

The wavelength and 1/e decay length of surface plasmon polariton is

\[ \lambda_{SPP} = \frac{2\pi}{k_m^*} \approx \lambda \sqrt{\frac{\varepsilon_m + \varepsilon_d}{\varepsilon_m \varepsilon_d}}, \]  

(4.6)

\[ \delta_m = \frac{1}{2k_m^*}, \]  

(4.7)

Where \( \lambda \) is the wavelength of the excitation light.
Figure 4.2 shows the dispersion map for light propagating in free space, dielectric material, and the surface plasmon mode propagating along the dielectric-metal interface. In order to couple propagating light into surface plasmon mode, both energy conservation and momentum conservation conditions must be satisfied. The angular frequency $\omega$ is proportional to energy of photon, and the wave vector $k$ accounts for momentum of photon. It is obvious that for every angular frequency $\omega$, the wave vector $k$ of propagating light in air is always smaller than that of surface plasmon mode, which means that the energy conservation and momentum conservation conditions cannot be fulfilled simultaneously. Thus, free space light cannot be couple into surface plasmon mode. For light propagating in high index dielectric material, we can see that at certain frequency $\omega_{sp}$, surface plasmon can be excited along the dielectric-metal interface. Moreover, the wave vector of excited surface plasmon polariton is much larger than that of light in free
space, which means that the effective wavelength of the surface plasmon polariton can be much shorter than the excitation light in vacuum. Though appropriate design of the excitation geometry, it is possible to achieve effective wavelength approaching X-ray region with optical frequency excitation.

4.2 Surface Plasmon Excitation Configurations

Two well known geometries for surface plasmon excitation are Otto configuration [83] and Kertschmann configuration [84] (Figure 4.3). In the Otto configuration, there is an air gap between the metal and dielectric prism. The laser beam with an incident angle larger than critical angle illuminates from the prism side. An evanescent wave is created at the air-dielectric interface, and couples into the surface plasmon mode at the air-metal interface if the gap is smaller than its decay length. This method is inconvenient due to the strict control of the air gap. Kretschmann proposed to coat the prism bottom with metal thin film, and surface plasmon wave can be excited at the air-metal interface.

In a typical attenuated total reflection (ATR) arrangement (such as the Kretschmann or Otto configuration), when the incident angle of a monochromatic TM polarized light satisfies the phase matching condition, a SPR phenomenon is observed as a sharp decrease in reflectivity within a very narrow incident angle range. Figure 4.4 shows the reflection and transmission coefficient curve versus incident angle of TM polarized light (Kretschmann configuration with 50nm silver film). It can be seen that the surface plasmon polariton condition is satisfied at $\theta_{sp}=44.1^\circ$. Maximum field enhancement about 8 can be obtained with 532 nm excitation. The surface plasmon waves are associated with shorter effective wavelengths and strong field enhancement effects, making them very attractive for a variety of applications. Due to its shorter
effective wavelength, surface plasmon wave can be focused into a highly confined spot with size beyond the diffraction limit.

Figure 4.3 Excitation of surface plasmon with (a) Otto configuration. (b) Kretschmann configuration.
Figure 4.4 Calculated (a) reflection coefficient and (b) transmission coefficient with respect to incident angles for TM polarized illumination. Field is enhanced at the surface plasmon excitation angle $44.1^\circ$. 
CHAPTER 5

SURFACE PLASMON RESONANCE WITH RADIAL POLARIZATION: THEORY

Optimal plasmonic focusing can be achieved through a combination of radially polarized illumination and axially symmetric plasmonic structures [42-46,85]. If radially polarized beam is focused onto axially symmetric plasmonic structures, unlike linearly polarized beams, the entire beam is TM polarized with respect to the interface, enabling surface plasmon excitation from all directions and homogeneous plasmon focusing through constructive interference of these plasmon waves. The challenges these applications facing are the optimization of the focus shape, size and strength. Here, three types of plasmonic focusing have been studied. Plasmonic lens with planar interface, conical shape and annular rings under radial polarization illumination are discussed.

5.1 Generation of Evanescent Bessel Beam

Bessel beams are diffraction-free solutions to the Maxwell’s wave equations that were first proposed by Durnin et al in 1987 [86,87]. These beams, which can be either propagating or evanescent, have found important applications in radar communications, nonlinear optics [88] and biophotonics [89]. The theoretical analysis and experimental
generation of Bessel beams remain continuous interests [86-92], while most of the proposed setups were complicated and difficult to implement.

In this work I demonstrate a simple approach to generate evanescent Bessel beam through combining surface plasmon resonance (SPR) excitation with a tightly focused radially polarized beam, eliminating the widely used conical devices.

Bessel beams can be generated with conical shape optical elements or axicon type computer generated holograms [86-92]. These techniques usually require complicated elements and precise alignment of the system. The combination of SPR excitation with a tightly focused radially polarized beam illumination provides a simple approach to generate an evanescent Bessel beam. In order to couple TM polarized light into surface plasmon modes, both energy and momentum conservations must be satisfied. The momentum conservation requirement is usually associated with a narrow resonance in the angular domain. This angular selectivity of the SPR excitation can be exploited within a rotationally symmetric optical system to mimic the function of an axicon for evanescent nondiffracting Bessel beam creation.

The proposed setup for the evanescent Bessel beam generation is illustrated in Fig. 5.1. An aplanatic oil immersion lens (NA=1.25) focuses the radially polarized beam onto a glass/silver interface. Immersion oil with refractive index matched to the glass substrate ($n_s=1.516$) is filled between the lens and the substrate. A 50 nm silver film ($\varepsilon=-10.18-0.824i$) is deposited onto the glass substrate. The medium above the silver layer is air ($n_m=1$). The optical excitation wavelength was chosen to be 532 nm. The surface plasmon excitation angle for this geometry is calculated to be 44.10° with a FWHM angular width of 0.89°. Due to the radial symmetry of the setup, a dark ring due to SPR excitation can
be obtained at the back focal plane of the objective lens (Fig. 5.2). The center part of the illumination corresponding to the incident angle below the critical angle $\theta_c = \sin^{-1}(n_m/n_s) = 41.27^\circ$ is blocked by a photomask. When a radially polarized beam is strongly focused onto this planar plasmonic structure, the entire beam is TM polarized with respect to the glass/silver interface, providing an efficient way to excite highly focused surface plasmon with constructive interference at the center and creating an evanescent spot with strongly enhanced localized field.

![Diagram of the proposed setup for evanescent Bessel beam generation](image)

Figure 5.1 Diagram of the proposed setup for evanescent Bessel beam generation. A radially polarized beam is focused onto a glass-silver interface by a higher NA lens. An immersion oil with the same index of refraction as the glass substrate is filled between the lens and the substrate. Surface plasmons are generated at all directions because the entire beam is TM polarized with respect to the interface.
Figure 5.2 Intensity distribution at the back focal plane of the objective lens after reflection. The dark ring corresponds to the SPR excitation.

The 2D intensity distributions at the silver/air interface are numerically studied with the Richards-Wolf vectorial diffraction model [42,47,48]. The radial component $E_r$ and longitudinal component $E_z$ after the silver/air interface can be expressed as

$$E_r(r,\varphi, z) = 2A \int_{\theta_{\text{min}}}^{\theta_{\text{max}}} \cos^{1/2}(\theta) P(\theta) t_p(\theta) \sin \theta \cos \theta \times J_1(k_1 r \sin \theta) \exp[i(z(k_2^2 - k_1^2 \sin^2 \theta)^{1/2})] d\theta,$$

$$E_z(r,\varphi, z) = i2A \int_{\theta_{\text{min}}}^{\theta_{\text{max}}} \cos^{1/2}(\theta) P(\theta) t_p(\theta) \sin^2 \theta \times J_0(k_1 r \sin \theta) \exp[i(z(k_2^2 - k_1^2 \sin^2 \theta)^{1/2})] d\theta,$$

where $\theta_{\text{min}}$ and $\theta_{\text{max}}$ are the minimum and maximum incident angles on the glass/silver interface corresponding to the annular illumination; $\theta_{\text{max}} = \sin^{-1}(\text{NA})$ is determined by the NA of the objective lens; $\theta_{\text{min}}$ is given by the photomask size; $P(\theta)$ is the pupil apodization function; $t_p(\theta)$ is the transmission coefficient of TM-polarization at the incident angle of $\theta$; $J_m(x)$ is the $m$th order Bessel function of the first kind; $k_1$ and $k_2$ are the wave numbers in the glass and air, respectively.
The calculated results are shown in Fig. 5.3. A homogeneous spot is generated at the geometric center with radially polarized beam illumination. The total intensity $|E|^2$ is the sum of longitudinal component $|E_z|^2$ and radial component $|E_r|^2$. The longitudinal intensity distribution shows a central peak, while the radial component has a donut shape. The ratio between the peak intensities of $|E_z|^2$ and $|E_r|^2$ is about 7.8:1. Thus the longitudinal component is much stronger than the radial component and dominates the total field distribution. The FWHM for the main lobe is 195 nm and breaks the diffraction limit, while the FWHM for the longitudinal component is about 171 nm. The total intensity distribution is broadened by the radial component. In contrast, if a linearly polarized light is used instead, the surface plasmon fields will cancel out at the center due to destructive interference of the longitudinal components and the total field will be much weaker (Fig. 5.4). Furthermore, only part of the focused linearly polarized beam is TM polarized with respect to the interface, leading to an inhomogeneous plasmonic focus.

If metal coated fiber NSOM probe with a nano aperture is used to map the intensity distribution, the detected signal is proportional to $|\nabla \cdot E_z|^2$ due to the symmetry of the $HE_{11}$ mode propagating in the fiber core of the probe. This expected NSOM signal is calculated and shown in Fig. 5.3d, which shows a donut pattern. However, due to the finite aperture size, certain amount of radial component will also be detected even if the probe is placed at the center of the focus.

The transverse profiles of the total intensity distributions at different distances from the silver/air interface are measured to show the nondiffracting nature of the beam (Fig. 5.5). From the normalized profiles in Fig. 5.5b, all of the main lobes nearly overlap with each other, indicating that the beam maintains its shape while the intensity drops as the
distance from the surface increases. As shown in Fig. 5.6, the intensity exponentially decays in the direction normal to the surface, with a decay length of 171 nm for $1/e^2$ of the maximum intensity. Therefore, a nondiffracting evanescent Bessel beam can be generated with the proposed simple setup.

Figure 5.3 Calculated results of surface plasmon intensity distribution at silver-air interface with radially polarized illumination. (a) Total intensity $|E|^2$, homogeneous spot with a strongly enhanced field at the center is obtain. (b) Longitudinal component $|E_z|^2$, which is much stronger than $|E_r|^2$ and dominates the total field distribution. (c) Radial component $|E_r|^2$, which has a donut shape at the center. (d) $|\nabla_\perp E_z|^2$ distribution, which is proportional to the detected power of apertured NSOM.
Figure 5.4 Surface plasmon intensity distribution with focused linearly polarized excitation. An inhomogeneous spot with much weaker peak intensity is resulted.
Figure 5.5 Nondiffracting property of the evanescent Bessel beam. a, Transverse profile of the total intensity $|E|^2$ at different distances from the silver/air interface. The intensity decays along the z axis. b, Normalized plot of a. The shape of the beam remains almost constant.

Figure 5.6 Calculated decaying nature of the evanescent Bessel beam. The decay length is about 171 nm.
5.2 Plasmonic Focusing with Multiple Rings Structure

In the above example, plasmonic focusing was realized with the help of high NA optical objective lens. The high NA objective lens is used to provide the phase matching for plasmon coupling. The same plasmonic focusing effect can also be achieved with flat plasmonic lens without the use of high NA optical objective lens. Flat plasmonic lens can be fabricated with spatially arranged subwavelength metal structures, such as notches, holes and slits in a metal thin film. For example, focusing with circular slit has been investigated and demonstrated recently by X. Zhang et al [100]. The scattering from these subwavelength structures provide the necessary coupling from optical energy into plasmonic waves. By making circular subwavelength slit into a silver film, the plasmon waves excited at the edges of the slit will have a curved wavefront and be focused towards the geometrical center. However, again due to the mismatch between the polarization of the excitation beam and the plasmonic structure, only part of the beam is TM-polarized with respect to the flat plasmonic lens and the focus generated this way is inhomogeneous. In addition, if the incident linear polarization is aligned with the center of the circular plasmonic lens, the plasmon waves generated at the opposite side of the circular slit will have opposite phase and consequently destructive interfere and creating a dip in contrary to a strong peak in the center.

I have demonstrated that radial polarization can also be used to improve the excitation and focusing for these plasmonics flat lens. I study the plasmonic lens with annular rings (Fig. 5.7) under radial polarization illumination. A 200nm silver film is deposited onto a quartz substrate. The width of annular slit is 250nm. The input field has a amplitude distribution of \( E = r \exp(-r^2/w^2) \), where \( w \) is the beam waist and is set to be 3 \( \mu \)m in our
simulation. The excitation wave length is chosen to be 632.8 nm. First, I explore the electric field enhancement factor at the geometric center with single ring. I adjust the radius to maximize the field enhancement at the focus. The optimum radius for the assumed illumination is found to be 2.488 µm.

Figure 5.7 Diagram of plasmonic lens with multiple annular rings

The calculated results are shown in Fig. 5.8. Due to rotational symmetry of both the plasmonic lens and excitation geometry, only half of the structure is shown. A strongly enhanced homogeneous spot is generated at the geometric center with radially polarized
beam illumination. The ratio between the peak intensities of $|E_z|^2$ and $|E_r|^2$ is about 48:1, which is much larger than the previous plasmonic focusing of planar thin film.

Figure 5.8 (a) Electric energy density $|E|^2$ distribution at air/silver interface of plasmonic lens with three annular rings. (b) Corresponding longitudinal electric field $|E_z|$ distribution. (c) Radial component $|E_r|$ distribution.
with high NA lens. The longitudinal component is much stronger than the radial component and dominates the total field distribution. The FWHM for the main lobe is 222 nm and breaks the diffraction limit.

In the case of multiple concentric rings structures, if the location and the rings are chosen to satisfy the circular Bragg condition for plasmonic wavelength, it is found that as more rings are added to the plasmonic lens structure, the peak intensity of the plasmonic center gets stronger while the focal spot remains approximately the same size. The location of each slit to the center is shown in Table 5.1. Fig. 5.9 shows the electric field density and polarization map distribution with three annular rings. From numerical simulations, the plasmonic focusing is clearly observed for single circular slit carved into metal thin film as well as plasmonic lens structures consists of multiple concentric rings (Bull’s eye). Surface plasmon waves constructively interfere at the center of the annular structure under normal radial polarization illumination, providing a strong enhanced field with size smaller than the diffraction limit. The field enhancement factor and spot size at the center of the plasmonic lens are shown in Table 5.1. The field enhancement factor can be increased by adding more rings to the structure which is because of more surface plasmon source with multiple slits. The spot size is almost unchanged with the increasing number of rings. The FWHM of the spot is about 222nm, which is 1/2.85 of the optical excitation wavelength.

As shown in the numerical simulation (Fig. 5.9), the electrical field distribution inside of the metal film clearly confirms the electron density oscillation at the metal dielectric interface. There are several other interesting properties need to be pointed out here. The electric field near the vicinity of the focus are purely polarized along the longitudinal
direction that is normal to the interface, generating a kind of optical “needle” field similar to that reported recently using a combination of multiple zone DOE and high NA objective lens. The Bull’s eye flat plasmonic lens provides and much more compact and simpler alternative to this approach. In addition, on both sides of the plasmonic lens the vector electric field shows polarization vortex structures near the slits that are worthy of further investigation.

Figure 5.9 Electric energy density $|E|^2$ distribution and polarization map confirm the surface plasmon oscillation inside the silver film.

Table 5.1 Field enhancement factor and spot size of plasmonic lens with multiple annular rings.

<table>
<thead>
<tr>
<th>Slit</th>
<th>Distance of slit edge to the plasmonic lens center (µm)</th>
<th>Field enhancement factor</th>
<th>FWHM spot size (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.488</td>
<td>7.34</td>
<td>220.6</td>
</tr>
<tr>
<td>2</td>
<td>3.122</td>
<td>13.32</td>
<td>224.0</td>
</tr>
<tr>
<td>3</td>
<td>3.632</td>
<td>17.53</td>
<td>222.8</td>
</tr>
<tr>
<td>4</td>
<td>4.208</td>
<td>25.99</td>
<td>222.2</td>
</tr>
<tr>
<td>5</td>
<td>4.884</td>
<td>30.45</td>
<td>221.6</td>
</tr>
</tbody>
</table>
I also studied the electric field enhancement factor at the focus of single annular metallic ring under radially polarized illumination. Due to the rotational symmetry of both metallic ring and illumination geometry, a 3D axial symmetry finite element method model can be applied to investigate the field enhancement at the geometric center of the metallic ring. I explored the field enhancement factor with respect to the ring inner radius \( r \), which is changing form 1 \( \mu \)m to 60 \( \mu \)m with step size 0.5 \( \mu \)m. From the calculated result shown in Fig. 5.10, we can see the field is always enhanced no matter how big the ring radius is. The maximum enhancement is about 13.7, occurred when \( r = 22.5 \) \( \mu \)m. The surface plasmon intensity is proportional to the ring area, which is about \( 2\pi r d \). Therefore, the intensity is increasing linearly with respect to the ring radius \( r \). On the other hand, the propagating loss of surface plasmon wave is also proportional to \( r \). The loss along the air/silver interface is calculated to be 2.28% for every step (0.5 \( \mu \)m). While for \( r \) larger than 22.5 \( \mu \)m, the increased surface plasmon intensity ratio is less than 2.28%. Thus the field enhancement starts to drop. Moreover, though the field enhancement is increasing from \( r = 1 \) \( \mu \)m to 22.5 \( \mu \)m, the ring area is larger for bigger \( r \), which means the illumination cost is higher. This is due to the large propagating loss along the air/silver interface for bigger ring. As shown in Fig. 5.11, we can see the efficiency is always dropping. In order to maximize the efficiency, the incoming light should be focused into a tighter spot with metallic rings structures matching to the illumination spot.
Figure 5.10 Field enhancement factor with respect to the ring radius from the center.

Figure 5.11 Coupling efficiency from input power to surface plasmon focusing drops with increasing the radius of annular ring.
5.3 Apertureless Near Field Scanning Optical Microscope Probe

Near field scanning optical microscope (NSOM) is one type of scanning probe microscopes (SPM) that can provide optical information with spatial resolution beyond the diffraction limit [93]. Combined with various feedback techniques, NSOM can offer topographical and optical information simultaneously and has found many applications in the characterization of sub-wavelength structures and devices. Most widely used NSOM probes are metallic coated tapered glass fiber with the end left uncoated to form a nanometric aperture. The resolution of apertured NSOM, determined by the size of the aperture and tip-sample distance, is generally limited to about 50 nm ~ 100 nm. The spatial resolution cannot be reliably increased further due to technical obstacles such as a weakly detectable signal, large background scattering noise and rigorous tip-sample distance control [94,95].

To overcome the drawback of apertured NSOM, various NSOM techniques using apertureless metallic probes were developed. Most of these techniques utilize the field enhancement effect arising from surface plasmon excitation. The principle of the local field enhancement in an apertureless NSOM is similar to the electrostatic lightning-rod effect. Surface plasmon, a group oscillation of electrons, can be generated at the dielectric/metal interface due to the interaction of metals with the incident light. Like free electron in electrostatics, surface plasmon can propagate to the sharply pointed structures, producing a strongly enhanced field at the end of the apertureless NSOM probe that can be used as a nanoscale near field light source. The enhancement factor strongly depends on both the structure of the probe tip and polarization state of the incident light. Most theoretical and experimental investigations of apertureless NSOM
use linearly polarized external illumination, which has the critical problem of large far field background noise [95-98].

In order to eliminate the background noise, apertureless NSOM with radially polarized internal light illumination was proposed [45,46,85]. When a radially polarized light is injected into the apertureless NSOM probe, the induced surface plasmon will converge toward the end of the tip and interfere constructively because of the rotational symmetry of the input polarization and the probe. Consequently, a strong field enhancement can be realized. Bouhelier et al. studied this field enhancement using a multiple multipole method, where the radial polarization was mimicked by placing an electric dipole parallel to the probe at the center of the input port [85]. In contrast, if a spatially homogeneously polarized light is coupled into the probe, the surface plasmon excitation will cancel out at the apex of the tip because the opposed sides on the probe surface have opposite charges. Hence, there is no field enhancement in this case. Here, I will use a finite element method model to quantitatively solve the electromagnetic field distribution at the vicinity of the tip. With an axially symmetric 3D model, I numerically investigate the field distribution and the field enhancement factor. Surface plasmon resonance excitation at the surface of thin silver film will be shown. A FWHM spot size smaller than 10 nm can be realized with 632.8 nm excitation. From our simulations, I also found that the enhancement factor strongly depends on the illumination spot size and the probe structure.

5.3.1. 3D Finite Element Modeling Structure

The probe structure to be modeled is illustrated in Fig. 5.12. A radially polarized beam is focused by an objective lens towards the bottom of a hemispherical solid
immersion lens (SIL), where a sharp conical shape tip is fabricated at the center. The SIL and the tip are not to scale to illustrate the details of the probe. The bottom of SIL and the entire tip are coated with thin silver film in order to form an apertureless probe and eliminate the strong far field background signal.

Figure 5.12 Diagram of an apertureless NSOM probe structure with radial polarization input.

A finite element method (FEM) is applied to investigate the field enhancement in the vicinity of the silver coated tip. Numerical simulation was performed with COMSOL, a commercial FEM software package. 3D axial symmetry geometry was applied to model the tapered tip as well as the illumination. Since both the structure and the illumination are axisymmetric, there are only variations in the radial \( r \) and vertical \( z \) directions and no dependence in the angular \( \theta \) direction. One can then solve a quasi 2D problem in the \( r-z \) plane instead of the full 3D problem, which can save considerable amount of memory and computational time.

In the model, the SIL material was chosen to have a dielectric constant of \( \varepsilon_r=2.2 \) at the excitation wavelength of 632.8 nm. The domain of calculation for the tip is chosen to be 1.2 \( \mu m \) in the \( r \)-direction and 3.86 \( \mu m \) in the \( z \)-direction. The tip has a half cone angle
of 16.4° and a radius of curvature of 20 nm. The entire tip is coated with a 50 nm thin silver film, and the radius of curvature of the silver film at the apex is set to be 5 nm. The dielectric constant of silver is chosen to be -15.8779 - 1.0765i. The input radially polarized beam is modeled as

$$E(r) = r \exp[-(r/w)^2] e^{i\varphi},$$

where \( r \) is the radial coordinate, and \( w \) is the beam waist of the focused incident light at the bottom of the SIL. In the following simulations, the beam waist is chosen to be 0.4 \( \mu \)m. After carefully setting the maximum finite element mesh sizes and boundary conditions, the field distribution within the whole domain of calculation can be rigorously solved.

### 5.3.2. Simulation Results for Field Enhancement and Spot Size

Fig. 5.13 shows the logarithmic 2-D and 3-D plots of electric energy density distribution in the vicinity of the tip. From the plots, we can see that the surface plasmon propagates along the air/silver interface and constructively interfere at the tip apex, leading to strong localized field.
Figure 5.13 (a) 2-D Electric energy density distribution at the end of probe tip. (b) 3-D Electric energy density distribution at the end of probe tip.
The distributions for the radial \((E_r)\) and longitudinal \((E_z)\) field components are shown in Fig. 5.14. Please notice that the color scale for the movies has been intentionally saturated in order to illustrate the details of the propagation.

Figure 5.14 Distribution of (a) longitudinal, and (b) radial, components of the electric field. Only half of the cone is shown in this figure.

Due to the rotational symmetry of both the illumination and the tip, there is no transverse electric field along the symmetry axis. From the movie, we can clearly see the propagation of surface plasmon towards the end of the tip. A high field enhancement at the tip apex is observed due to the constructive interference of the surface plasmon. In this case, the electric energy enhancement is about 102,400, corresponding to an electric field enhancement of about 320. The value of the enhancement is given by the ratio between the field intensity at the end of the tip and the incoming field peak intensity. The
spot size of the strong localized field is analyzed through calculating the FWHM of the intensity distribution at different distances from the tip. The results are shown in Fig. 5.15. The FWHMs of 7.3 nm, 16.9 nm and 27.3 nm are calculated at distances of 0 nm, 5 nm and 10 nm from the tip apex, respectively.

![Normalized electric energy density distribution](image)

Figure 5.15 Normalized electric energy density distribution at 0, 5, 10 nm from the tip.

5.3.3. Optimal Condition for Field Enhancement

I further explore different taper angles of the tip, illumination spot sizes, metal coating material and thickness, tolerance of alignment, and find that the enhancement factor has strong dependence on these parameters. In order to determine the optimal taper angle for maximum field enhancement, I numerically studied the enhancement factor for different half cone angles with the same illumination spot size. The results of this study are shown in Fig. 5.16. Due to the diffraction and multiple reflections upon the metal/dielectric interfaces, the relationship between the enhancement and taper angles exhibits a strong oscillating nature, which means that the enhancement factor is very
sensitive to the tip taper angle. Though the field enhancement is sensitive to the taper angle, in practice, we can choose an angle range with relatively higher field enhancement to design the tip. For example, according to Fig. 5.16, field enhancement factor higher than 50 is generally obtainable for half taper angles ranging from 15° to 19°. Thus it would make sense to select 17° as the desired half taper angle to fabricate the tip. It should be pointed out that, although at certain angles the enhancement factors are very low, they are always higher than one, which means the field is still enhanced.

Figure 5.16 Electric field enhancement factor with respect to taper angle of probe.

Moreover, probe designs with different metal coatings (gold and aluminum) and their enhancement performance are studied as well. Fig. 5.17(a) and Fig. 5.17(b) illustrate the electric field enhancement factor dependence on taper angle of probe for gold and aluminum respectively. From these plots, we can see the performance of gold gives better performance at the excitation wavelength, which is due to the smaller absorption
coefficient. For the same reason of a big absorption coefficient, the field enhancement of aluminum is much lower with 50 nm film thickness.

Figure 5.17 Electric field enhancement factor with respect to half taper angle of probe for (a) 50 nm gold coating, and (b) 50 nm aluminum coating.
Figure 5.18 Electric field enhancement factor with respect to incident beam size.

In addition to the tip cone angle, another study was also carried out to study the effects of the illumination spot size on the field enhancement. Fig. 5.18 shows the relationship between the electric field enhancement factor and spot size $w$ with the same half taper angle of 16.4°. It is found that there is an optimal spot size of about 0.68 μm for this tip cone angle. This relationship indicates that, given a fabricated tip with fixed cone angle, we may be able to conveniently maximize the enhancement effect by adjusting the size of the focal spot through controlling the aperture of the objective lens. For example, for the minimal field enhancement occurs at 20° half cone angle shown in Fig. 5.16, I found the field enhancement factor can be increased to 50 by increasing the spot size from 0.4 μm to 1 μm.
To explore the optimal condition for the electric field enhancement, I studied the influence of the metal thin film thickness on the field enhancement. Fig. 5.19 shows the relationship between electric field enhancement factor and the silver thin film thickness. I found that the optimal thickness is about 55nm for the tip with a 16.4° taper angle. While
from Fig. 5.20, I find that the beam size is almost unchanged, which remains about 6 nm. Essentially, the spot is determined by the radius of the tip.

I also investigated the tolerance of the alignment angle, because some probe could use tapping mode for probe-sample distance regulation (illustrated in Fig. 5.21). For this study, a full 3D COMSOL model had to be used due to the break of symmetry by the deflection angle. When radially polarized beam is focused into the cone tip with a small tilt angle, which may happen because of the vibration of the tip, the enhancement factor decreases as expected. However, it remains at a relatively high value for fairly large deflection angles. Fig. 5.22 shows the normalized field enhancement factors at different tilt angles compare to that of normal incidence.

Figure 5.21 Misalignment of probe tip and illumination.
In conclusion, optical surface plasmon focusing with radially polarized beam is analyzed for three different types of structures. For planar metallic/dielectric interface, an evanescent nondiffracting evanescent beam with optimal spot size and field strength can be obtained with strongly focused radial polarization. Plasmonic lens with conical shape and annular rings under radial polarization illumination are also studied by 3-D axial symmetry FEM model. The focusing properties and field enhancement effect of these plasmonic lenses are numerically studied. For the conical shape plasmonic lens, the field distribution with a FWHM of as small as 10 nm and intensity enhancement of five orders of magnitude can be achieved with 632.8 nm optical excitation. For the plasmonic lens with annular rings, the field enhancement can be increased by adding more rings, while the spot size is almost unchanged. The high field enhancement and strong spatial confinement provided by the optimal plasmonic focusing could find many applications in nanoscale imaging and sensing.
CHAPTER 6
SURFACE PLASMON RESONANCE WITH RADIAL POLARIZATION: EXPERIMENTS

6.1 Generation of Radially Polarized Beam

The experiment setup for generating radially polarized beam is shown in Fig. 6.1. A linearly polarized beam from Coherent DPSS Nd:YAG second harmonic 532 nm green laser is collimated and passing through a SPE (Fig. 6.2). A charge +1 vortex beam is created after the element, and coupled into a few-mode fiber. The cutoff wavelength of the fiber (Thorlabs 630HP) used in the radial polarization generation is around 570 nm. Therefore, it supports the second higher order modes at our excitation wavelength 532 nm and eliminates all other higher modes. LP_{11} is the second higher mode which includes TE_01, TM_01 and HE_{21} modes, with TE_01 being the radially polarized beam, TM_01 being the azimuthally polarized beam, and HE_{21} being the hybrid mode. All these modes have a donut intensity distribution pattern, and can be distinguished from each other by a linear analyzer. Generally, it is difficult to excite a pure radially or azimuthally polarized beam with high efficiency at the end of the fiber without generating the fundamental Gaussian mode. A radially polarized beam with high efficiency and stability can be generated through coupling a charge +1 vortex beam into a few-mode fiber.
Figure 6.1 Experimental setup for generating radially polarized beam.

Figure 6.2 SPE for creating a +1 vortex beam.

Figure 3 shows the donut pattern of the radially polarized beam generated at the fiber output. Pictures of the beam after it passes through a linear analyzer oriented at different angles are also captured to confirm the radial polarization distribution. The two lobe patterns follow the rotation of linear analyzer.
Figure 6.3 Radially polarized light pattern generated at the end of the fiber, and pictures of the beam after it passes through a linear analyzer oriented at different angles shown as the arrows. The patterns follow the rotation of the linear analyzer.

6.2 Realization of Evanescent Bessel Beam

Most previous experimental works of plasmon focusing used linearly polarized beam, which usually resulted in a minimum longitudinal field at the geometric focus due to destructive interference between counter-propagating surface plammon waves and inhomogeneous plasmon focal spot owing to the symmetry mismatch between the incident polarization and the plasmonic structures [99-101].

When a radially polarized beam is strongly focused onto a glass/silver interface, the entire beam is TM polarized with respect to the interface and capable of exciting surface plasmon waves. The interference of these surface plasmon waves creates an evanescent Bessel beam after the silver thin film with strongly enhanced localized field and spot size beyond the diffraction limit. The excitation of surface plasmon is confirmed by the observation of a narrow dark ring at the back focal plane. The rotationally symmetric angular selectivity of SPR excitation mimics the function of axicon, leading to a very simple approach to generate evanescent Bessel beam. 2D intensity distributions at
different distances from the sample surface are mapped by a collection mode NSOM to verify the nondiffracting and decaying natures of the evanescent Bessel beam.

![Experimental setup diagram](image)

Figure 6.4 Experimental setup for evanescent Bessel beam generation. PM: photomask, OBJ: objective lens.

The diagram of the experimental setup is illustrated in Fig. 6.4. A radially polarized beam is generated at the end of the fiber as mentioned above. A circular photomask placed after the collimated radially polarized beam provides an annular illumination that blocks the incident angles below the critical angle. The beam is tightly focused onto the glass/silver interface by an oil immersion objective lens with an NA of 1.25. A 50 nm silver film ($\varepsilon=-10.1786-i0.8238$) is deposited onto the glass substrate. The medium above
the silver layer is air. The surface plasmon intensity distribution is directly imaged by a collection mode NSOM (Veeco Aurora 3) using a metal coated fiber probe with an aperture size of 50-100 nm. The fiber probe is mounted on a tuning fork and shear force feedback mechanism is applied to regulate the probe/sample distance.

The lateral vibration of the tuning fork is parallel to the sample surface, which is beneficial to the plane by plane near-field scanning in this experiment. On the other hand, normal force feedback probes vibrating vertically would make it difficult to accurately map the evanescent wave at several distances from the sample surface due to the fact that the probes move in and out of the near-field.

The reflected intensity distribution at the back focal plane of the objective lens can be captured with a CCD camera by inserting a beam splitter in the optical path to observe the SPR coupling (Fig. 6.5). Due to the fact that the entire beam is TM polarized with respect to the glass/silver interface, a dark ring corresponding to SPR excitation by the focused radially polarized beam is observed, indicating that surface plasmons are excited for all directions. Instead, two dark arcs will be observed if a linearly polarized beam is used for excitation. The excitation angle is measured to be 45.51° with a FWHM angular width of 1.28°. Only the incident beam corresponding to the dark ring is coupled into surface plasmon modes. Thus, the SPR excitation with highly focused radial polarization performs a rotationally symmetric angular filtering function for the transmitted field that mimics an axicon device. The discrepancy between the experimental and the theoretical excitation angles is largely attributed to the differences between the actual refractive index of the deposited silver film and the handbook index value used in the theoretical calculation. Moreover, the beam splitter has different reflectivity for TE and TM
polarization, slightly breaking the symmetry and causing the horizontal part of the reflection image appear a little darker. The beam splitter is used for alignment and observation of the SPR excitation only. Before taking the NSOM images, the beam splitter is removed from the setup after the photomask is inserted and aligned in the beam path.

Figure 6.5 Intensity distribution at the back focal plane of the objective lens after reflection. The dark ring corresponds to the surface plasmon excitation. Surface plasmons are generated at all directions with radially polarized beam excitation.

The measured 2D intensity distribution of the surface plasmon focus shows the expected rotational symmetry (Fig. 6.6). The plot is in logarithmic scale for better visualization of outer rings. The surface plasmon waves generated at all azimuthal directions propagate to the geometric center, constructively interfere with each other and create a strongly enhanced local field. It can be clearly seen that the rings due to surface plasmon interference are getting stronger when they come closer to the focus center. Since the detected signal of the apertured NSOM fiber probe is proportional to $|\nabla_\perp E_z|^2$, 

the experimental results show a dark center as predicted by the theoretical calculation (Fig. 4.3d). The far field image is obtained with the probe retracted a few microns away from the surface (Fig. 6.7). Much weaker signal corresponding to leakage and scattering light is captured. The transverse profiles of the measured intensity distribution and the calculated data after the silver/air interface were plotted in Fig. 6.8. The two peaks are about 278 nm apart. Spot size as small as 195 nm ($\lambda_0/2.728$) can be obtained with 532 nm optical excitation. The surface plasmon interference period is measured to be 239 nm in good agreement with theoretical prediction. The center of transverse profile of the experimental data does not go to zero is because the finite aperture size of probe tip. The spot size of the surface plasmon field is only a few times of the probe aperture size. Consequently the scanning optical images are the convolution of the aperture function of the probe and the surface plasmon intensity distributions. Thus, when the tip is located exactly at the focus center, surface plasmon field within the radius of tip size also contributes to the detected optical signal, giving rise to the elevated signals at those minimum signal locations.
Figure 6.6 Measured near field intensity distribution of evanescent Bessel beam. Multiple rings corresponding to surface plasmon wave propagation are observed. Due to the apertured NSOM probe is more sensitive to $|\nabla \times E_z|^2$, a dark center is obtained as expected. Plot is in logarithmic scale for better visualization of outer rings.

Figure 6.7 Measured far field intensity distribution. Plot is also in logarithmic scale for comparison. Much weaker background leakage signal and scattering light are detected.
Figure 6.8 Comparison of measured and calculated transverse profile of intensity distribution. Experimental result agrees with simulated result very well. The mainlobe and sidelobe location are about the same.

The normalized transverse intensity profiles at different distances from the sample surface are plotted in Fig. 6.9. It can be seen that all curves almost overlap with each other, indicating the nondiffracting property of the beam. Then the probe is moved to the peak of the innermost ring and the signal is measured at a series of distances away from the surface. The evanescent decaying nature of the Bessel beam is clearly shown in Fig. 6.10, with a decay length of 143 nm obtained through curve fitting. These experimental results confirm that an evanescent Bessel beam is generated via surface plasmon excitation with a highly focused radially polarized beam.
Figure 6.9 Measured nondiffracting nature of the evanescent Bessel beam. The intensity decays along the z axis, but the shape of the beam remains almost constant.

Figure 6.10 Measured evanescent decaying property of the evanescent Bessel beam. Decay length is measured to be 143 nm.

6.3 Plasmonic Focusing with Concentric Rings

I also conducted experiments to verify the optimal focusing effect with the Bull’s eye plasmonic lens. A 200 nm silver film was deposited onto a glass substrate by e-beam evaporation. Annular ring patterns with single ring, 5-ring and 9-ring were fabricated
through focused ion beam (FIB, FEI dual beam SEM-FIB NOVA 200 Nanolab system) by D. C. Abeysinghe. Figure 6.11 shows the SEM image of 9-ring plasmonic lens in silver film. The innermost ring has a diameter of 9.2 µm. The slit width is 135 nm, and period is 500 nm, which matches to the surface plasmon wavelength. Surface plasmons excited at different slits have phase differences of \(2\pi n\) \((n=1,2,3,\ldots)\). Therefore, when propagating to the center of the plasmonic lens, these surface plasmon waves are in phase and will interfere constructively with each other, producing higher field enhancement than single ring plasmonic lens. The surface plasmon intensity distribution was directly imaged by a collection mode NSOM (Veeco Aurora 3).

![Figure 6.11 SEM image of a 9-ring plasmonic lens in silver film.](image)

Due to the rotational symmetry of both the annular metallic rings and optical excitation geometry, surface plasmons excited by the plasmonic lens at all azimuthal
directions interfere constructively. Therefore, a tightly focused spot with strong field enhancement is obtained at the center. There are two components contributing to the total energy density distribution $|E|^2$, longitudinal component $|E_z|^2$ and radial component $|E_r|^2$. $|E_z|^2$ is calculated to be 31.4 times higher than $|E_r|^2$, and dominates the total energy density distribution. The measured 2D intensity distribution of the surface plasmon focusing along the air/silver interface of the single ring plasmonic lens shows the expected rotational symmetry (figure 6.12). The surface plasmon waves excited at all azimuthal directions propagate towards the geometric center, creating a strongly enhanced local field with constructive interference. It can be clearly seen that the fringes due to surface plasmon interference are getting stronger when they come closer to the focus center. Since the detected signal of the apertured NSOM fiber probe is proportional to $|\nabla \times E_z|^2$, which is due to the symmetry of fundamental mode in the fiber core of a probe, the experimental results show a dark center as predicted by the normalized theoretical calculation. The transverse profiles of the measured energy density distribution and the calculated result after the silver/air interface are plotted in Figure 6.13. The two peaks are about 294 nm apart, which corresponds to a FWHM spot size of 184.4 nm (0.347\(\lambda\)). The surface plasmon interference fringe period is measured to be 255 nm, in good agreement with theoretical prediction.

The field enhancement of single ring plasmonic lens is given by the constructive interference of surface plasmon waves excited at all azimuthal directions. It is possible to further improve the enhancement through appropriately designing more concentric rings in the radial direction. In the case of multiple concentric rings structures, if the locations of the rings are chosen to satisfy the circular Bragg condition for plasmonic wavelength,
it is found that as more rings are added to the plasmonic lens structure, the peak intensity of the plasmonic center gets stronger while the focal spot remains approximately the same size.

Figure 6.12 Measured near field energy density distribution at the air/silver interface for single ring plasmonic lens. Multiple fringes corresponding to surface plasmon wave propagation are observed. Due to the apertured NSOM probe is more sensitive to $|\nabla E|^2$, a dark center is obtained as expected.

Figure 6.13 Comparison of measured and calculated transverse profiles of energy density distribution for single ring plasmonic lens. Experimental result agrees with simulated result very well.
Figures 6.14a and 6.14b show the 2D surface plasmon energy density distribution at the surface of plasmonic lens with 5-ring and 9-ring patterns, respectively. From the energy density scale bar, it is demonstrated that the peak intensity increases with respect to the number of rings. The intensity enhancement is 8.11 and 19.08 times higher than that of single ring structure for 5-ring and 9-ring plasmonic lens. While from the transversal profiles (figure 6.14c), all curves almost overlap with each other, indicating the plasmonic focal spot size remains the same. The strong spatial confinement and high field enhancement make plasmonic lenses very attractive for near-field optical imaging and sensing in material characterization and biological applications.

Strongly enhanced plasmonic fields are dominating in the near field, while in the far field, radiative fields created at the subwavelength slits start to contribute to the total field distribution. The decay length of surface plasmon is calculated to be 254.4 nm for 1/e² of the maximum intensity. Therefore, in regions located much further from the surface, radiative fields have considerable contribution to the total intensity. In order to verify this, NSOM probe is retracted 2 µm away from the surface to map the far field energy density distribution. As shown in figure 6.15, radiative fields in far field have bigger spot size and fringe spacing. The two peaks are about 346 nm away, corresponding to a FWHM spot size of 214 nm.

In conclusion, optimal plasmonic focusing is experimentally demonstrated for single circular slit carved into metal thin film as well as plasmonic lens structures consists of multiple concentric rings. When a radially polarized beam illuminates the plasmonic lenses, the entire beam is TM polarized with respect to the dielectric/metal interface. Surface plasmon excited by the radially polarized light propagates toward the center of
plasmonic lenses and interferes constructively, creating a strongly enhanced focal spot with spot size beyond the diffraction limit. In the case of multiple concentric rings structures, if the location and the rings are chosen to satisfy the circular Bragg condition for plasmonic wavelength, it is found that as more rings are added to the plasmonic lens structure, the peak intensity of the plasmonic center gets stronger while the focal spot remains approximately the same size. Optimal plasmonic focusing using radially polarized illumination offers tremendous advantages in terms of the plasmonic focus shape, peak intensity and spatial localization.
Figure 6.14 Measured near field energy density distribution at the air/silver interface for 5-ring (a) and 9-ring (b) plasmonic lens. The intensity enhancement is 8.11 and 19.08 times higher than that of single ring for 5-ring and 9-ring plasmonic lens, respectively. (c) Comparison of measured transverse profiles of energy density distribution for single ring, 5-ring and 9 ring plasmonic lens. The shape of curves overlap each other, indicating that the focal spot remains the same.
6.4 Preliminary Experiments on Field Enhanced Raman Spectroscopy with Apertureless NSOM Tip

For this work, 50 nm silver film was deposited onto a NSOM fiber probe to form an apertureless tip. The SEM picture of the probe tip is shown in figure 6.16. The experimental setup is illustrated in figure 6.17. The fiber for generating radially polarized light was spliced with the apertureless NSOM fiber probe tip. Though the probe tip is apertureless, there is still some leaking light [figure 6.18(a)]. The polarization at the tip end was observed with a transmission camera of NSOM system. Due to the limited resolution of the objective lens, I had to defocus the tip end slightly to view the transmission image. A donut spot corresponding to radially polarized light was obtained [figure 6.18(b)]. A linear analyzer was used in the transmission image path and manually adjusted to exam the polarization pattern. The images after the linear analyzer are shown
in figure 6.18(c). Two lobe patterns were observed. Therefore, radially polarized light was successfully coupled into the apertureless fiber probe.

Figure 6.16 SEM picture of apertureless NSOM fiber probe tip

Figure 6.17 Near field Raman spectroscopy using apertureless NSOM probe tip with radially polarized illumination.
Figure 6.18 (a) Observed apertureless fiber tip with transmission camera of NSOM system. (b) Leaking light shows radial polarization. (c) Two lobe patterns observed by linear analyzer.

Strongly enhanced field is expected at the end of the tip due to the optimal plasmonic focusing effect described in the previous chapter. A standard silicon sample half covered with metal with a sharp transition edge was used for the investigation of the field enhancement. Reflected light including the Raman signal was collected by an objective lens with a NA of 0.2. A notch filter was used to block the laser light of 532nm. Raman signal was collected by a Horiba Jobin Yvon iHR550 spectrometer.
First, we located the tip above the silicon area, and collected both near field and far field spectra (figure 6.19). It is found that the required collection time was significantly shorter than the cases with aperture probes. With aperture probes, 300 seconds integration time was necessary to collect a decent spectrum with sufficient SNR. With the apertureless probe and successful radial polarization coupling, spectrum with similar SNR can be observed with 1 second integration time. The spectrum with 10 seconds integration time is clearly comparable with the 300 seconds cases with an aperture probe. It is also worthy pointing out that the current collection optics has a low NA of 0.2. The collection efficiency can be improved 8 times if an objective lens with NA of 0.55 is used.

In the collected Raman spectra, the broad shoulder was confirmed to be coming from the Raman scattering of the glass core of the fiber. In order to verify this, I moved the probe tip to the metal area, the broad shoulder was still observed, while the Raman peak due to silicon disappeared.

When the tip is retracted, it is found the Raman signal gets stronger first. This is owing to the reduction of the shadowing effect. To correct this, the near field and far field spectra are normalized using the SiO$_2$ signal. With this correction, an enhancement of the Silicon Raman peak from the near field signal was found. An example with 60 sec integration time is shown in Fig. 6.19(b).

Note that the far field signal comes from a much larger spatial area, while the near field signal comes from area that is presumably on the same order of the tip radius. From the topographic image we obtained with the probe tip (Fig. 6.20), the radius of the tip was estimated to be 49.4 nm. I assume the far field excitation area to be 2 um, and the penetration depth is assumed to be the same (obviously, most likely the penetration depth
of far field signal will be larger and leads to a larger excitation volume). From the figure, the far field signal is estimated to be 135 and the near field signal is estimated to be 230-135=95. The enhancement effect can be calculated to be:

\[ EF = \frac{A_{far}}{A_{near}} \times \frac{S_{near}}{S_{far}} = \frac{2000^2}{49.4^2} \times \frac{95}{135} = 1153 \],

(6.1)

where \( A_{far} \) and \( A_{near} \) are far field and near field illumination area by the apertureless tip, \( S_{far} \) and \( S_{near} \) are far field and near field Raman signal level.

The lower field enhancement comparing to the simulated result can be explained by the bigger tip radius and surface roughness of the probe tip. In the calculation, the tip radius is assumed to be 5 nm, which will correspond to a 100 times higher field enhancement. The surface roughness of the probe is other critical issue. Scattering light due to bad surface quality results in lower field enhancement at the tip end.

![Graph showing Raman intensity vs. wavelength](image)
Figure 6.19 Collected Raman spectrum. (a) 10 seconds collection. (b) 60 seconds collection.

Figure 6.20 The scanning image obtained across the metal and silicon boarder confirmed the tip size is 49.4 nm.
CHAPTER 7

CONCLUSIONS AND FUTURE WORK

7.1 Conclusions

In summary, a 3D focus shaping technique using the combination of generalized CV beams and DOE was demonstrated. 3D flattop focus with extended DOF and optical bubble can be obtained. This research has also systematically demonstrated the first analytical method for arbitrary 3D state of polarization control with optimal spot at the focal point of a high NA lens. An electric dipole situated at the focus mimics the prescribed focal spot with desired polarization. The input field at the lens pupil can be obtained by solving the angular distribution of the dipole radiation. The availability and feasibility of spatial light modulators make real time control of both state of polarization and field distribution possible.

Optimal plasmonic focusing was proposed with rotationally symmetric illumination polarization and metallic structures. Radially polarized beam with high efficiency and stability was generated by coupling a charge +1 vortex beam into a few-mode fiber. I have demonstrated the first experimental realization of nondiffracting evanescent Bessel beam generation via surface plasmon resonance excitation with radially polarized beam. The combination of radial excitation symmetry and the SPR angular selectivity
eliminates the need for a conical device and significantly simplifies the alignment procedure simultaneously. The excitation of surface plasmon is confirmed by the observation of a narrow dark ring at the back focal plane of the high NA lens. The interference of these surface plasmon waves creates an evanescent Bessel beam with strongly enhanced localized field and spot size beyond the diffraction limit. The nondiffracting and evanescent decaying natures of the beam were verified with a collection mode NSOM.

A significantly enhanced localized electromagnetic field with a FWHM of 10 nm is obtainable at the end of an apertureless NSOM probe tip with 532 nm radially polarized excitation. The study provided the systematic knowledge for manufacturing and optimizing the probe for apertureless NSOM. The strong field enhancement at the end of the apertureless probe tip was preliminarily confirmed by collecting the Raman spectrum of silicon sample. Both near field and far field spectra are collected. The collection time is significantly reduced compared with the case using aperture probes. With aperture probes, 300 second is required to collect a decent spectrum with sufficient SNR, while similar spectrum can be obtained with 1 second integration time using the new apertureless probe under radial polarization excitation. Plasmonic lenses with single and multiple annular rings under radial polarization illumination is also studied both numerically and experimentally. Experimental results confirmed that higher field enhancement factor can be obtained with increasing number of rings while the spot size remains almost unchanged.
7.2 Future Work

In the experiment for confirming evanescent Bessel beam generation, an apertured NSOM is applied to measure the near field and far field intensity distribution, and the detected signal is proportional to $|\nabla \cdot E_{\perp}|^2$ due to the detection mechanism of the apertured fiber probe. An approach for direct observation of longitudinal component and transverse component is desirable. One possible method is to use an apertureless probe to scatter the near field evanescent wave into propagating wave, and then collect the signal with an objective lens. The other potential way is to attach fluorescent molecule to the tip end of an aperture probe tip, and the fluorescent light can be simultaneously collected by the fiber through the open aperture, and by the objective lens at the reflection arm.

I have shown that enormously field enhancement at the end of an apertureless NSOM probe tip theoretically and experimentally in the visible range. The enhancement is believed to be lower with infrared light excitation due to the reduced plasmonic properties of metals in the wavelength range. However, experimental parameters can be optimized for the optimization of field enhancement at infrared range, including the probe structure, probe material, metal coating material, illumination geometry, etc. With these optimizations, tip enhanced Raman spectroscopy with silicon cantilever probe could be explored in a transmission type setup. In addition, in the near field Raman spectroscopy experiment, we fixed the tip at certain point above the sample to test the near field enhancement. In order to obtain stress, temperature, chemical and structural distribution of a semiconductor or biological sample, it is necessary to perform near field scanning Raman image with apertureless NSOM tip under radially polarized illumination.
The nondiffracting evanescent Bessel beam generated via surface plasmon excitation has strong near field enhancement with spot size breaking the diffraction limit, and maintains constant size as the observation plane moves away from the surface. Such evanescent non-spreading fields may have important applications in high density data recording, biosensing and semiconductor metrology. For current near field optical characterization techniques such as NSOM, the illumination spot size is determined by the open aperture, and also the throughput is extreme low due to the nano aperture. The distance between the sample and the aperture need to be rigorously controlled, otherwise the spot will rapidly expanding and the size could be increased many folds. Furthermore, the size of the open aperture generally becomes bigger after several scans, which will also cause the expansion of illumination size and degradation of spatial resolution. Near field imaging using the nondiffracting evanescent Bessel beam can overcome these problems. Its application to biosensing, such as measurement of nanoscale molecular size and distance, could be explored as well.
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