Subwavelength-sized plasmonic structures for high-resolution optical imaging

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**NANYANG
TECHNOLOGICAL
UNIVERSITY**

**SUBWAVELENGTH-SIZED PLASMONIC STRUCTURES
FOR HIGH-RESOLUTION OPTICAL IMAGING**

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2012

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ABSTRACT

Optics does not naturally suit the nanometer world because the optical length scale is on the order of several hundred nanometers due to diffraction limit. However, exploiting the hybrid nature of surface plasmons (SPs) offers a way to hurdle this diffraction limit and thus enable a route to subwavelength optics. SPs are essentially light waves coupled to free electron oscillations in a metal, which are strongly confined in the nanoscale region of metal/dielectric interface. The nature of two-dimensional evanescent waves bestows SPs the unique capabilities in tailoring optics by subwavelength-sized surface structures to be incorporated into nanoscience for various applications. Specifically, these plasmonic fields hold exceptional promise to miniaturize optical components as well as to provide wide-field view in the field of super-resolution microscopy.

The work contained in this thesis is directly towards the design, fabrication and characterization of subwavelength-sized plasmonic structures for integration of the excitation, modulation and interference of SP waves into nanoscale. The subdiffraction SP interference patterns can be served as the excitation profiles in the proposed novel super-resolution fluorescence microscopy.

At first, the thesis discusses the design of subwavelength-sized slit array embedded in the metal film to excite SP waves propagating along metal/dielectric interface. The design benefits from the compact configuration for potential applications in on-chip plasmonic devices. The surface relief dielectric elements deposited on the metal surface are used to investigate the phase modulation of SPs by

using finite-difference time-domain (FDTD) method. Due to the exponential decay property of SPs, the length, width and thickness of dielectric structure have different effects on SP phase modulation, respectively. A modified effective-refractive-index model is proposed to optimize the three-dimensional size of dielectric element in SP phase modulation, with consideration of SP losses. Specifically, an in-plane phase modulation Fresnel zone plate is constructed by using this model to realize the totally constructive interference of SPs into subwavelength-sized focal point. This subdiffraction SP spot, as a virtual probe, is of significant interest in the resolution enhancement of the integrated in-plane plasmonic microscopy.

Subsequently, the design of four perpendicular metallic subwavelength-sized slit arrays is demonstrated to generate the SP interference patterns. By rotating the polarization of incident beam, one-dimensional or two-dimensional high contrast SP standing waves are generated in the center area of slit structure. These SP interference fringes are of high spatial frequency and confined in the vicinity of the metal film surface. Therefore, they can be served as the excitation profile of fluorescence imaging for lateral resolution enhancement.

Last but not least, a wide-field high-resolution fluorescence microscopy is demonstrated by using the phase-shifted SP standing waves illumination. The numerical and experimental results show that the lateral resolution of reconstructed fluorescence density image is enhanced to $0.28\lambda_{\text{SP}}$ in two dimensions, which is twofold better than that of conventional high numerical aperture (NA=1.42) fluorescence microscopy. With such standing wave excitation nature, it helps to break through the spatial resolution beyond the diffraction limit. The proposed SP standing wave fluorescence microscopy also benefits from compact SP coupling configuration to offer a high level of miniaturization for applications in integrated lab-on-chip wide-

field super-resolution imaging. In addition, since the SP excitation is contactless, this approach allows direct access to biological specimens in their living environment. In the future, a nanometer scale resolution is possible by introducing fluorophore nonlinearities to extended techniques into the nonlinear regime.

In summary, novel subwavelength-sized plasmonic structures are proposed in this thesis to tailor SP fields for applications in fluorescence imaging, which will open a new chapter of on-chip microscopy to realize super-resolution imaging.

SYMBOLS AND ABBREVIATIONS

List of Symbols

λ : wavelength

r : radius

Δr : lateral resolution

Δz : axial resolution

n : refractive index

θ : angle

k : wavevector

ε : permittivity

ω : frequency

E : electric field

D : electric displacement field

L : propagation length

G : grating constant

m : number

Λ : pitch

O : exposure dose

A : area

C : beam current

t : thickness

w : width

l : length

φ : phase

d : distance

I_{sp} : SP field intensity

I : image function

o : object function

P : point spread function

f : weighting function

α : arbitrary phase

List of Abbreviations

NA: numerical aperture

1D: one-dimensional

2D: two-dimensional

3D: three-dimensional

SEM: scanning electron microscopy

SIM: structured illumination microscopy

STED: stimulated emission depletion

PALM: photo activation localization microscopy

STORM: stochastic optical reconstruction microscopy

SSIM: saturated structured illumination microscopy

PSF: point spread function

SPP: surface plasmon polaritons

SPs: surface plasmons

NSOM or SNOM: near-field scanning optical microscope

EBL: electron beam lithography

ATR: attenuated total reflection

TM: transverse magnetic

TE: transverse electric

MIBK: methyl isobutyl ketone

IPA: isopropanol

ITO: indium tin oxide

PMMA: polymethyl methacrylate

HSQ: Hydrogen silsesquioxane

TMAH: tetramethyl ammonium hydroxide

PMT: photo-multiplier tubes

FIB: focus ion beam

Au: gold

Ag: silver

Al: aluminum

FDTD: finite-difference time-domain

PML: perfectly matched layer

FZP: Fresnel zone plate

PMFZP: phase modulation Fresnel zone plate

AMFZP: amplitude modulation Fresnel zone plate

FWHM: full width half maximum

DOF: depth of focus

SP-SWF: surface plasmon-standing wave fluorescence

CCD: charge-coupled device camera

SPCE: surface plasmon-coupled emission

CMOS: complementary metal oxide semiconductor

CHAPTER 1

INTRODUCTION

1.1 Background

1.1.1 Surface plasmon subwavelength optics

The performance of electronic devices, circuits and components fundamentally depends on their physical dimensional size. The advances of fabrication techniques continue to improve the miniaturization and integration of modern electronic devices for information processing. However, electronic devices are now approaching their fundamental speed and bandwidth limitations, which is an increasingly serious problem to further improve the performance. Photonics offers an effective solution to this problem by replacing electron with light as information carriers. Unfortunately, the low levels of integration and miniaturization of photonic devices, due to the diffraction of light in dielectric media, limit their applications in dense chip-based integration. According to the Abbe theory [1], the diffraction limit of light is equal to 0.5λ , where λ is the wavelength of light, which means the photonic device does not naturally suit to be miniaturized into nanoscale regions much smaller than the wavelength of light in the material.

One of the most feasible ways of circumventing this problem is to exploit the hybrid nature of surface plasmons (SPs). Their particular near-field characteristics and associated field enhancement are of great interest to a wide range of researchers from physicists, chemists and materials scientists to biologists. Most recently, a renewed interest in SPs stems from the developments of fabrication techniques, which make it possible to structure thin metallic film on the nanoscale for tailoring SPs beyond the diffraction limit. As a result, a new branch of nanophotonics, namely “plasmonics”, is developed to study the propagation, localization and modulation of strongly localized SPs using metallic nanostructures. It opens up a way to develop new types of subwavelength-sized plasmonic devices in applications of integrated circuits, light sources and nanolithography, as well as in nano-resolution optical imaging [2-4].

In the following sections, we are concerned primarily with surveying the different types of subwavelength structures that have been explored in the context of plasmonic optics.

(a) Plasmonic chips

The strong confinement of SP fields at a metal/dielectric interface indicates the feasibility of SP-based quasi-two-dimensional optics. Such subwavelength-sized plasmonic components could someday replace electronic circuits to overcome the size-compatibility problem. The plasmonic optoelectronic circuit will consist of various components such as waveguides, switches, modulators, and couplers.

Fig. 1. 1 Plasmonic nanowire waveguide. (a) Topography of a silver nanowire. (b) Near-field image of SP propagation in nanowire. [5]

Plasmonic waveguide has been configured by using various geometries. As shown in Fig. 1.1, nanowires, with lateral dimension much smaller than the wavelength of SPs, guide SP waves propagating over a distance of a few micrometers [5]. An array of nanoparticle has also demonstrated a decay length longer than $50\text{ }\mu\text{m}$ by guiding the SPs localized in one dimension [6].

Fig. 1. 2 Electric current control of SPP transmission through and reflection from a gallium stripe. [7]

In plasmonic circuits, an active plasmonic was achieved by exploiting light-induced nanoscale structural transformations in the metal-on-dielectric waveguide [7], as shown in Fig. 1.2. Some other active plasmonic devices have also been reported, such as the one by combining thin polymer films containing molecular chromophores with a thin silver film [8], the one by light-on-light modulation using SPs thermal effects in a thin gold foil [9].

Fig. 1. 3 Nanodots plasmonic coupler and nanostrip waveguide. (a) SEM image of the focusing nanodot array and Ag strip guide. (b) NSOM image of the SP intensity showing focusing and guiding. [10]

Plasmonic couplers with high conversion efficiency are necessary for far/near-field optical signal transmission. A promising solution for this feature was reported by combining a quarter circle arranged nanodots and metal nanostrip waveguide [10], as shown in Fig. 1.3. A similar promising coupler was also obtained by using hemispherical metallic nanoparticles as a plasmonic condenser and a nanodot array as a plasmonic waveguide [11].

(b) Light source

Fig. 1. 4 Beaming light from subwavelength apertures. (a1) FIB micrograph image of a bull's eye Ag structure surrounding a cylindrical hole. (a2) Optical image of the sample of (a1) illuminated from the back. (b1) FIB micrograph image of Ag slit aperture with parallel grooves. (b2) Optical image of the sample of (b1) illuminated from the back. [12]

SPs have been displayed as a key role in the enhanced transmission through subwavelength single aperture [12], as shown in Fig. 1.4, or hole arrays [13]. The enhanced optical transmission originates from subwavelength plasmonic structures associated with SPs. By texturing the metallic surface with a subwavelength pattern at microwave, millimeter wave, and THz frequencies, enhanced transmissions are also observed by the resonant tunneling of SPs [14, 15].

(c) Nanolithography

Fig. 1. 5 SEM image of sub-100-nm lines by surface plasmon polariton lithography. [16]

SPs also provide an alternative solution to breakthrough diffraction limit of optical lithography by using its strongly enhanced nanoscale spatial distribution of electric field near the metal surface. Using this technique, sub-100-nm lines have been patterned at a wavelength of 436 nm by designing corrugated surface of the metallic mask [16], as shown in Fig. 1.5. Furthermore, theoretically simulation predicts as small as 30 nm features can be produced using interfering surface plasmons launched by 1D grating. [17].

In addition, the unique properties of SPs, such as near-field characteristic and associated field enhancements, combined with the advancement in nanoscale fabrication techniques, pave a way for novel applications in super-resolution imaging. In the following section, we review the mechanism of optical nanoscopy by means of surface plasmons, which offer the exciting possibility of super-resolution microscopy.

1.1.2 Super-resolution microscopy

Scientists have long dream of an optical microscopy to see samples with nanometer resolution. Modern optical microscopes have been developed with many complex designs aiming to improve the resolution and sample contrast such as utilization of dark field [18], phase contrast cross-polarized light illumination [19] and confocal scanning mechanism [20]. However, the diffraction nature of light limits optical microscopes to reach a spatial resolution beyond the value comparable to the wavelength of light. According to the Abbe diffraction limit [1], the focused light radius (r) in optical microscopy is equal to

$$r = \frac{0.61\lambda}{n \sin \theta} \quad (1.1)$$

where λ is the wavelength of light and $n \sin \theta$ is numerical aperture (NA) in microscope. Hence, the straightforward method to increase the resolution is to increase the NA or decrease the wavelength. However, the highest theoretical numerical aperture obtainable with immersion oil is 1.51, and the light wavelength can not always be lowered. The resolution of far-field light microscopy is still limited to $\Delta r > 200$ nm of lateral resolution and to $\Delta z > 450$ nm along the optical axis. It has been a main barrier to the achievable resolution of fluorescence microscopy for a long time.

(a) Optical super-resolution microscopy

There are a number of optical techniques proposed to dramatically enhance the obtainable resolution, enabling the researchers to inspect the finer details of samples. The purely optical approaches, such as 4Pi microscopy [21], I⁵ microscopy [22] and structured illumination microscopy (SIM) [23], extend the diffraction resolution to the limit. The advantage of these approaches is that they do not use special fluorophore.

However, the best resolution using these methods can only be improved by a factor of 2. In order to obtain much higher image resolution it would involve the nonlinearity of fluorophore. Current super-resolution fluorescence microscopy methods can be divided into two approaches. One is based on single molecule localization, as of stimulated emission depletion (STED) [24, 25], photo activation localization microscopy (PALM) [26] and stochastic optical reconstruction microscopy (STORM) [27]. However, they essentially suffer from point by point scanning nature or take hours to collect the data, which are not suitable to study dynamically. The other kind of super-resolution microscopy is based on spatially patterning the excitation light, as of saturated structured illumination microscopy (SSIM) [28]. However, in practice only a finite number of harmonic components are strong enough to be distinguished from the noise. Furthermore, this nonlinear phenomenon requires high-power pulsed lasers with an increased risk of photobleaching and photodisruption in comparison with conventional fluorescence imaging.

(b) Metallic tip

A nano-sized metallic probe tip can excite localized modes of SPs which generates a nanoscale spot of light. With this tiny light spot as the light source of an optical microscopy, extremely high spatial resolution can be obtained. Kawata *et. al* first demonstrated this concept through an apertureless near-field scanning optical microscope (NSOM; also known as SNOM) in 1994 [29]. Recently, tip-enhanced Raman spectroscopy (TERS) combining Raman scattering with the NSOM was reported to image single-walled carbon nanotubes with a spatial resolution of 25 nm [30]. X. S. Xie *et. al* also demonstrated a tip enhanced fluorescence image of photosynthetic membrane fragment with spatial resolution of the order of 20 nm by using a gold nano tip [31]. Metallic tip has played an extremely important part in the

area of nano-imaging. However, the contact scanning mechanism limited its applications in living cell imaging.

(c) Superlens and hyperlens

In 2000, J. Pendry predicted that the decay of evanescent waves can be offset by amplifying them in a negative refractive index material [32]. Theoretically, evanescent waves, instead of decaying, can be enhanced through the slab of noble metal film material by exciting SPs at optical frequencies. Following Pendry's proposal, several superlenses have been built to take evanescent waves from a surface and transmit them to produce a high-resolution image [33, 34]. However, the image exists only at the near-field of the final layer.

Fig. 1. 6 Plasmonic hyperlens. (a) Schematic of the experimental set up for the first hyperlens demonstration [35]. (b) Schematic of the magnifying superlens for surface plasmons [36].

In 2006, X. Zhang [35] and I. Smolyanivov [36] successfully created hyperlenses. Both of these designs utilize anisotropic metamaterials with hyperbolic dispersion to realize magnification in a cylindrical geometry, as shown in Fig. 1.6. As such, a magnified image is projected onto the surrounding dielectrics outside this cylindrical medium and to be resolved with a conventional optical microscopy

typically with a resolution of 60–90 nm. Other improved designs, such as impedance-matched [37], flat [38], oblate [39, 40], and aperiodic [41] hyperlens have been proposed to magnify subdiffraction objects to the far-field in one dimension. More recently, spherical hyperlens for two-dimensional subdiffraction limited real-time imaging at visible frequencies was presented [42]. However, superlens has some practical limitations in obtaining high-resolution due to the surface roughness and the absorption of plasmons in metal which resulting in the loss of information. Since very specific techniques are required to fabricate multilayer nanoscale thin film, and the imaging targets are required to be projected in the near-field region, superlens are relatively not feasible for most of the biological applications.

(d) Super-oscillation

Recently, the idea of achieving super-resolution without evanescent fields has been demonstrated by N. I. Zheludev, based on the fact that band-limited functions are able to oscillate arbitrarily faster than the highest Fourier components that they contain, a phenomenon known as super-oscillation [43, 44]. An optical generator of super-oscillating fields has been demonstrated with the use of a Penrose type quasicrystal array of nano-holes in a thin metal film. When illuminated with a coherent light source, it creates a complex diffraction pattern on the other side of array and leads to well-defined, sparsely distributed subwavelength light localizations at few tens of micrometers distance away [45, 46]. Because such subwavelength localizations are formed by propagating far-fields, they can be projected to the far-field by a conventional lens or used as a proper far-field to far-field super-resolution lens to achieve a subwavelength resolution. In comparison with conventional near-field scanning optical microscopy with near-field probes, the super-oscillating mask will allow for the placement of the object at a considerable distance from the mask.

For many biomedical imaging applications, this is crucial advantage over NSOM. However, the serious barrier to the development of optical super-oscillation generators is manufacturing accuracy at the nanometer scale [47].

(e) Standing wave surface plasmon resonance fluorescence microscopy

Fig. 1. 7 SW-SPRF microscopy. (a) Experimental setup (b) SW-SPRF image with linear deconvolution to reduce side lobes and its point spread function profiles. [48]

Standing wave surface plasmon resonance fluorescence (SW-SPRF) microscopy was developed based on total-internal-refraction (TIR) microscopy by Peter T. C. So *et al.* in 2009 [48]. In this configuration, two beams fixed at the angle of SP excitation are incident into TIR microscopy, as shown in Fig. 1.7. The counter-propagating SP waves launched through a high-NA objective lens interfere each other and excite fluorescence samples on the gold-coated coverslip. The point spread function (PSF) profiles of SW-SPRF demonstrate that lateral resolution of approaching 100 nm in one direction is achieved. Two-dimensional imaging with extended resolution is possible by manually rotating sample stage. SW-SPRF also shows a significant increase in signal-to-background-noise ratio due to strong fluorescence excitation field of SPs. However, the using of complicated phase control

feedback system or huge size of TIRF configuration blocks these conventional SW-SPRF microscopes from applications in on-chip biological imaging. Furthermore, the TIR coupling configuration, suffering from its limited incident angle, can not be used to excite high frequency SP waves for further resolution improvement.

(f) Nanostructured surface plasmon illumination microscopy

Fig. 1. 8 Nanostructured SP illumination microscopy. (a) The schematic of grating-assisted TIRFM. (b) Simulation result of the reconstructed density of fluorophores obtained with the linear inverse procedure from the 24 images. [49]

Recently, a potential SP based super-resolution microscopy was proposed with the design of a periodically nanostructured glass slide by A. Sentenac *et. al* in 2006 [49-51]. As shown in Fig. 1.8, the sample is deposited on a nanoscale grating substrate. The permittivity of grating substrate is designed to generate a high-frequency SP grid as the fluorescence excitation profile. A linear inversion scheme is utilized to reconstruct the fluorophore's density distribution from a series of images with varying azimuthal incident angles. The numerical study points to the feasible improvement of resolution of $\lambda/10$ [52]. However, due to the requirement of high quality grating fabrication, this grating assisted SPF microscopy is realized only in numerical experiment.

Most recently, Z. W. Liu *et. al* numerically demonstrated a plasmonic structured illumination microscopy by tuning the incident angle of the excitation light

to realize the phase-shift of SP interference pattern for the reconstruction of high-resolution image [53]. However, the requirement of precise control on incident angle (0° , 4.6° , 8°) makes it much difficult to experimentally realize super-resolution imaging. Additionally, the orientation of the illumination pattern is also needed to be rotated to yield several intermediate images for the two dimensional resolution improved image, which is time consuming and might induce vibrations.

1.2 Motivations

Studies of the dynamical process of biological samples always require real-time, non-invasive, and high-resolution imaging with sufficient contrast. Although most of the present super-resolution imaging approaches, such as STED, PALM and SSIM, complement each other and emerge as effective tools for nanoscale biological imaging, such platforms require moderately complex, bulky and expensive architectures which partially hinder their wide applications. The miniaturization of conventional optics is restricted by diffraction to about half the light wavelength. In order to push the miniaturization of optical elements and devices, new concepts out of the box are needed.

SPs, due to their unique properties, enable light localization and guiding in nanoscale for applications in super-resolution microscopy. As surface waves, the fields of SP propagate along the metal/dielectric surface and decay exponentially in directions perpendicular to the SP carrying interface. The strong confinement of SP fields at a metal/dielectric interface indicates the possibility of SPs modulation by using surface defects, similar to modulation of light beams, but in two dimensions. Because the SPs carry a larger wave vector compared to that of the excitation photon, by a ratio of $|k_{SP}|/|k_0|$ where $|k_{SP}|$ and $|k_0|$ are the magnitude of the wave vectors of SPs

and excitation photon, respectively. This ratio can be tuned up to as high as 10^2 by adjusting the frequency of the excitation light to approach the SP's resonant frequency. As a result, the coupling of free space light into SPs on a planar surface should lead to subdiffraction resolution features.

Very recently, some SP-involved approaches have achieved subdiffraction limited resolution. The superlens microscopy has shown its potential in resolution enhancement with a flat configuration, but it requires samples to be placed in a specific location where it is relatively not feasible for most of the bio-applications. As for NSOM scanning scheme, it is notorious for its time consuming and not suitable for soft samples. As for SP based standing wave microscopy, it suffers from both the huge size of TIR configuration and limited incident angle, also unsuitable to be integrated on-chip for super-resolution biological sample detection. Although the present SP based nanostructured microscopes have demonstrated their potential nanoscale resolution in theory without compromising a wide-field configuration, the high quality requirements of structure fabrication or illumination beam control make them difficult to be implemented in practice. Therefore, in this thesis, we go beyond the concepts to propose alternative subwavelength-sized surface structures for excitation, modulation and interference of SPs into subdiffraction patterns as the excitation profiles in high-resolution fluorescence image. To take advantages of SPs' unique potentials, the proposed plasmonic devices in this work provides much feasibility to miniaturize microscopy.

The dispersion of SPs is sensitive to the dielectric properties of the interface, which make it feasible to spatially modulate the SPs in phase with surface relief dielectric structures. Compared to tailoring SPs with metallic surface defects, the phase modulation methods make the devices capable of varying the optical properties

while achieving better modulation performance. Therefore, subwavelength dielectric structures deposited at the side of SP grating coupler is proposed to investigate the phase modulation of SPs for interference. The totally constructive interference of in phase SP waves will form a subdiffraction limited focal spot, which can be served as a virtual probe for resolution enhanced in-plane plasmonic microscopy. Due to its compact excitation scheme, the grating coupler helps to miniaturize plasmonic devices for lab-on-a-chip techniques.

Additionally, wide-field high-resolution surface plasmon microscopy is possible with the design of slit array structures to excite and modulate SPs to form standing waves as the excitation profiles. The SP interference patterns with high-spatial-frequency information excite the fluorescent beads on the metal plate, thereby improving the imaging resolution and providing wide-field view. Due to the nature of evanescence wave, SPs are highly confined near the interface and exponentially decay into surrounding dielectric for several hundreds nanometers. The limited penetration of SP field at the metal/dielectric interface may efficiently suppress the background noise in fluorescence imaging. Likewise, the proposed slit array surface structures provide a novel method to miniaturize the SP standing wave microscopy by integrating the generation of excitation profiles with the main fluorescence imaging procedure on a chip.

Last but not least, the development of nanofabrication techniques such as electron beam lithography (EBL), ion-beam milling, self-assembly and nanoimprint lithography, plus with modern nanocharacterization techniques such as near-field optical microscopes, have opened up new ways to pattern metals surfaces and control SP propagation and dispersion at the nano scale. To taking full advantage of SPs' potentials, the focus of our efforts and this thesis are the study the excitation and

manipulation of SPs, and further applications of subdiffraction SP interference patterns in super-resolution fluorescence imaging.

1.3 Objectives

This thesis describes the development of apparatus and methods to realize the excitation and modulation of SPs by subwavelength-sized surface structures in applications of on-chip high-resolution SP fluorescence imaging.

Several aims will be pursued in this thesis:

1. *To explore the characteristic of grating coupler for SP excitation.* To design subwavelength-sized silt array as the SP coupler to miniaturize plasmonic devices.
2. *To investigate phase modulation of SPs with subwavelength-sized surface relief dielectric structures.* To develop a theoretical model for optimization of dielectric structures in phase modulation of SPs and to experimentally demonstrate the totally constructive interference of SPs to form a subdiffraction limited focal point on the metal surface, as a virtual probe for potential applications in the in-plane plasmonic microscopy.
3. *To investigate modulation of SP interference to standing wave patterns with subwavelength-sized metallic structures.* To design, fabricate and characterize subwavelength-sized silt arrays for excitation and modulation of SPs to form one-dimensional (1D) or two-dimensional (2D) high contrast SP standing waves, acting as excitation patterns in wide-field high-resolution fluorescence imaging.

4. *To demonstrate the first proof of principle of wide-field high-resolution SP fluorescence microscope technique.* To integrate the SPs coupling and fluorescence excitation procedures on a compact metallic chip and to break through the diffraction limited spatial resolution by phase-shift of SP standing waves as the illumination source.

The proposed SP manipulation methods and the subdiffraction limited SP patterns in this thesis are also expected to be useful in applications beyond the scope of this thesis, for example, nanolithography, particle manipulation and data storage.

1.4 Major contributions of the thesis

This thesis makes three major contributions to the research fields of excitation and modulation of SPs with subwavelength-sized surface structures for high resolution optical imaging.

1. *Development of in-plane Fresnel zone plate lens to focus SPs into subwavelength focal spot.*

Subwavelength-sized slit arrays are fabricated to excite SPs propagating along metal surface. A modified effective-refractive-index model is proposed for the design of relief dielectric structures deposited on the right side of SP grating coupler to realize phase modulation of SPs. With this model, the length of the dielectric structure is optimized to obtain phase shifting between the SP waves transmitted through dielectric structure and those directly propagated with the consideration of SP losses. Specifically, a one-dimensional dielectric Fresnel zone plate is realized by employing the proposed model to demonstrate phase modulation of SPs into a subwavelength focal point.

2. *Design of four perpendicular subwavelength slit arrays to generate SP standing wave patterns.*

Four perpendicular subwavelength-sized slit arrays are designed to excite and modulate SPs on the metal surface. The counter-propagating SPs constructive interfere to form 1D or 2D SP standing waves in the center region of the structure. NSOM is employed to examine intensity distributions of the generated SP interference patterns. The full width half-maximum (FWHM) of 0.24λ in 1D and of 0.34λ in 2D standing waves are achieved by varying the polarization states of the incident beam, which shows good agreement with finite-difference time-domain (FDTD) simulation results.

3. *Demonstration of wide-field high resolution fluorescent microscopy using phase shifted SP standing wave illumination.*

A wide-field super-resolved optical microscopic imaging technique is demonstrated, based on subwavelength-sized slit arrays embedded in a thin silver film to generate SP standing wave interference patterns. These fringes carrying high spatial frequency information serve as excitation profiles to excite the nanoscale fluorescence objects. The super-resolved fluorescence density distribution is reconstructed from a weighted sum of a series of fluorescence images with differently phase-shifted SP standing wave illumination. Simulation and experimental results show that the lateral resolution of the reconstructed fluorescence density image is enhanced by $0.28\lambda_{SP}$ in two dimensions, which is twofold better than that of conventional high numerical aperture fluorescence microscopy.

1.5 Organization of this thesis

This work is organized as the following:

Chapter 1 briefly introduces subwavelength surface plasmon optics, the super-resolution optical microscopy techniques and SP involved high-resolution techniques that are related to this work. The motivations, objectives as well as the organization of this thesis are also presented in Chapter 1.

Chapter 2 presents the background knowledge of SPs that include overview, SP wave equations and unique properties of SPs, as well as the SP modulation methodologies by subwavelength-sized dielectric and/or metallic structures.

Subsequently, in Chapter 3, we discuss the fabrication and characterization methods that were used in this work such as EBL and NSOM.

Chapter 4 discusses phase modulation of SPs by dielectric element to totally constructive interference to form a subwavelength size SP focal point as a virtual probe.

In Chapter 5, subwavelength-sized slit arrays are utilized to interfere SPs to form standing wave pattern, which contains high frequency and can be act as the wide-field excitation profiles in fluorescence imaging.

Chapter 6 demonstrates the achieved 2D wide-field high-resolution SP fluorescence images by reconstruction of a series of images with phase-shifted SP standing wave illuminations.

Finally, in Chapter 7, a brief summary concludes this work, describing all important findings and offer an outlook for possible routes to follow this work.

CHAPTER 2

OVERVIEW OF SURFACE PLASMONS AND THEIR MODULATION SCHEMES

2.1 Introduction

In order to modulate SPs for applications in on-chip high-resolution microscopy by using subwavelength-sized plasmonic structures, it is necessary to explore the unique properties of SP waves as well as their interactions with surface structures.

This chapter reviews the research history in SPs and presents an electromagnetic theory of surface plasmons based on Maxwell wave equations' analysis. The main characteristics of SPs along planar metal/dielectric interface are introduced, and methods for optical excitation and detection of SPs are discussed. This chapter also briefly describes the methods of SP modulation with subwavelength-sized surface metallic and dielectric structures. These discussions form the groundwork for the research in the subsequent chapters of this thesis.

2.2 Overview of surface plasmons

2.2.1 Historical milestones in surface plasmons research

The first scientific studies of SPs date back to the beginning of the early twentieth century. In 1902, Robert W. Wood observed the dramatic drop in the optical reflection spectra measurements from metallic diffraction gratings over a small change in incident angle [54]. Although Wood had no idea at that time to explain this anomaly due to SPs, his work marked the beginning of the SP research area. In 1956, these anomalies were attributed to the excitation of surface waves by Robert Fano [55]. A major advance in the study of SP was made in 1968 when Andreas Otto as well as Erich Kretschmann and Heinz Raether presented the attenuated-total-reflection (ATR) method for the optical excitation of SPs on metal films [56, 57]. These contributions make experiments on SP easily accessible to many researchers, which are the now most widely used device geometry.

Most recently, with the advancement of nano-scale fabrication technique and the invention of scanning probe techniques, the systematic investigations lead to a revival of the research on surface plasmons. In 1997, Junichi Takahara and co-workers suggested that metallic nanowires enable the guiding of optical beams with a nanometer scale diameter [58]. In 1998, Thomas Ebbesen and coworkers reported on the extraordinary optical transmission through subwavelength metal apertures [59], and in 2001 John Pendry suggested that a thin metallic film may act as a “perfect lens” [32]. All these findings have motivated a tremendous amount of new research, captured in a number of exciting review articles [2, 4, 60].

The term “surface plasmons” was first used to describe the interaction of the plasma oscillations on the surfaces of metal foil [61]. The term “surface plasmon

polariton” (SPP) is used frequently to emphasize the connection of the electronic excitation in the solid to its associated electromagnetic field. However, in academic literature, the “surface plasmon polaritons” and “surface plasmons” are always used interchangeably to describe the coherent oscillation of free charge on the planar surfaces [62]. For simplicity in this thesis, we refer to them as SPs.

2.2.2 Surface plasmons mode equation solution

Due to the nature of electromagnetic wave, the wave solution of SPs can be obtained via a straightforward solution of Maxwell’s equations with appropriate boundary conditions [63].

We consider an infinite metal/dielectric interface plane as a xy plane at $z=0$. The metal is characterized with a complex frequency-dependent permittivity function $\varepsilon_1(\omega)$ where the dielectric permittivity $\varepsilon_2(\omega)$ is real. We are looking for homogeneous solution of Maxwell’s equations for the metal/dielectric interface. The Maxwell’s equation is demonstrated by a general wave equation form. Mathematically, SP is the solution of

$$\nabla \times \nabla \times E(r, \omega) - \frac{\omega^2}{c^2} \varepsilon(r, \omega) E(r, \omega) = 0 \quad (2.1)$$

where $\varepsilon(r, \omega) = \varepsilon_1(\omega)$ if $z < 0$ and $\varepsilon(r, \omega) = \varepsilon_2(\omega)$ if $z > 0$.

Firstly we consider p-polarized or transverse magnetic (TM) plane waves solution in half-space $j=1$ and $j=2$. The electromagnetic field can be written as

$$E_j = \begin{pmatrix} E_{j,x} \\ 0 \\ E_{j,z} \end{pmatrix} e^{ik_x x - i\omega t} e^{ik_{j,z} z} \quad j=1, 2 \quad (2.2)$$

The situation is depicted in Fig. 2.1.

Fig. 2. 1 Wavevectors and associated field for p-polarization (TM mode).

The wavevector parallel to the interface is conserved, which leads to the following relations

$$k_x^2 + k_{j,z}^2 = \epsilon_j k^2 \quad j=1, 2 \quad (2.3)$$

where $k=2\pi/\lambda$, and λ is the vacuum wavelength. The electric displacement fields in both half-spaces have to be source free, which leads to the boundary condition

$$k_x E_{j,x} + k_{j,z} E_{j,z} = 0 \quad j=1, 2 \quad (2.4)$$

The parallel component of the electric fields E and the perpendicular component of the electric displacement field D are required to be continuous, which yields other boundary condition

$$\begin{aligned} E_{1,x} - E_{2,x} &= 0 \\ \epsilon_1 E_{1,z} - \epsilon_2 E_{2,z} &= 0 \end{aligned} \quad (2.5)$$

The Eq. (2.4) and Eq. (2.5) lead to nontrivial solution only if

$$\epsilon_1 k_{2,z} - \epsilon_2 k_{1,z} = 0 \quad (2.6)$$

Combination with Eq. (2.3) and Eq. (2.6) leads to the component of wavevector along the propagation direction (k_x), which is expressed as

$$k_x^2 = \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} k^2 = \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \frac{\omega^2}{c^2} \quad (2.7)$$

The normal component of the wavevector $k_{j,z}$ is expressed as

$$k_{j,z}^2 = \frac{\varepsilon_j^2}{\varepsilon_1 + \varepsilon_2} k^2 \quad j=1, 2 \quad (2.8)$$

The conditions for existence of such a surface electromagnetic wave are the following

$$\begin{aligned} \varepsilon_1(\omega) \cdot \varepsilon_2(\omega) &< 0 \\ \varepsilon_1(\omega) + \varepsilon_2(\omega) &< 0 \end{aligned} \quad (2.9)$$

The ε_2 is always positive for dielectric constant. Equation (2.9) means that ε_1 needs a negative real part and an absolute value exceeding ε_2 . Noble metals such as gold and silver have such large negative real part of permittivity with a small imaginary part. Therefore, at the interface between a noble metal and a dielectric, such as air, oil or glass, localized modes at the metal/dielectric interface can exist.

For s-polarized or transverse electric (TE) light, the electric field vector is perpendicular to the plane of incidence. The electromagnetic field can be written as

$$E_j = \begin{pmatrix} 0 \\ E_{j,y} \\ E_{j,z} \end{pmatrix} e^{ik_y y - i\omega t} e^{ik_{j,z} z} \quad j=1, 2 \quad (2.10)$$

Considering the continuity relations of the tangential components,

$$E_{1,y} = E_{2,y} \quad (2.11)$$

and

$$H_{1,y} = H_{2,y} \quad (2.12)$$

From Eq. 2.1 and Eq. 2.10, they lead to

$$k_{1,z} + k_{2,z} = 0 \quad (2.13)$$

Since the confinement to surface requires both $\text{Re}[k_{1,z}]$ and $\text{Re}[k_{2,z}]$ bigger than 0. Therefore, there is no surface modes existed for s-polarization, and SPs can only be excited by p-polarized light.

2.2.3 Spatial extension of surface plasmons

For metals, ε_1 is complex and frequency-dependent due to the dispersion, as can be described as the following

$$\varepsilon_1 = \varepsilon_1' + i\varepsilon_1'' \quad (2.14)$$

The dielectric medium has a real permittivity, ε_2 . According to Eq. (2.7), k_x is naturally a complex number, $k_x = k_x' + ik_x''$. The real part k_x' determines the SP wavelength, while the imaginary part k_x'' accounts for the internal absorption of the SPs. In this case, the k_x can be determined from Eq. (2.7) under the assumption that

$$|\varepsilon_1''| \ll |\varepsilon_1'|$$

$$k_x = k_x' + ik_x'' \approx k_0 \sqrt{\frac{\varepsilon_1' \varepsilon_2}{\varepsilon_1' + \varepsilon_2}} + ik_0 \left(\frac{\varepsilon_1' \varepsilon_2}{\varepsilon_1' + \varepsilon_2} \right)^{\frac{3}{2}} \frac{\varepsilon_1''}{2(\varepsilon_1')^2} \quad (2.15)$$

For the SP wavelength, it can be thus decided as

$$\lambda_{SP} = \frac{2\pi}{k_x} \approx \sqrt{\frac{\varepsilon_1' + \varepsilon_2}{\varepsilon_1' \varepsilon_2}} \lambda \quad (2.16)$$

With the complex permittivity of metal $\varepsilon_1(\omega)$, the SP frequency is described as $\omega = \omega' - i\omega''$, where both ω' and ω'' are real. SP waves in half-spaces ($j=1$ and $j=2$) can be written as

$$E_j = \begin{pmatrix} E_{j,x} e^{-|k_x''|x} \\ 0 \\ E_{j,z} e^{-|k_{j,z}|z} \end{pmatrix} e^{i(k_x'x - \omega't)} e^{-|\omega''|t} \quad j=1, 2 \quad (2.17)$$

From Eq. (2.17), we can conclude that SP waves are intrinsically electromagnetic waves that propagate along the interface between a metal and a dielectric. Its property is defined by the characteristic of the metal and dielectric medium. The localization at the interface is characterized by the exponentially decaying of electromagnetic fields away from the interface into both half-spaces, as shown in Fig. 2. 2.

Fig. 2. 2 (a) Charges and electromagnetic fields of SP propagating along x direction on a surface. (b) The exponential dependence of the field $|E_{j,z}|$.

When introducing the purely imaginary wavevector component into the expression equation for a plane wave, the SP propagating at the metal/dielectric interface are equivalent to the free-space plane waves with a spatial decay component $\exp(-|k_x''|x - |k_{j,z}|z)$, $j=1, 2$ and the temporal decay component $\exp(-\omega''t)$. Therefore, the SP wave carries a wavevector and behaves like a propagating wave. Reflection,

diffraction or scattering can also occur for SP waves interacting with surface metallic structures. Specifically, the dependence of SP dispersion on the permittivity of the dielectric makes it highly sensitive to the variation of the dielectric properties at the interface. Small variations of dielectric property thereof will cause efficient change of SP properties. Therefore, fabrication of dielectric structures on the metal surface allows tailoring SPs with desired properties.

The SPs decaying along the interface can be described by k_x'' which is responsible for an exponential damping of the electric field amplitude. The $1/e$ decay length of the electric field density is defined as propagation length L_x .

$$L_x = \frac{1}{2|k_x''|} \quad (2.18)$$

This damping is caused by Ohmic losses of the electrons participating in the SPs and finally results in heating of the metal.

The normal component of SP field amplitude decreases exponentially at the order of $\exp(-|k_{j,z}|z)$. The depth away from the interface of the SP electric fields dropping to $1/e$ can be obtained from Eq. (2.8).

$$L_{j,z} = \frac{1}{|k_{j,z}|} \quad (2.19)$$

According to Eq. (2.7), the dispersion relation of SP propagating along an interface between the metal and the dielectric is described as

$$k_{SP} = \frac{\omega}{c} \left(\frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2} \right)^{1/2} \quad (2.20)$$

Due to negative permittivity of metal, the dispersion curve of the SP always lies to the right of the free space photon k_0 for all wave vectors, as shown in Fig. 2.3. With the increase of frequency ω , the k_{SP} dispersion deviates further from k_0 , which

leads to larger wavevectors and shorter SP wavelengths. From this dispersion relation, it is obvious that SPs cannot be directly transformed from free space photon and the momentum mismatch must be satisfied to couple the light with SPs.

Fig. 2. 3 Dispersion relations of free space photons (A), free photons propagating in dielectric (B) and the surface plasmons (C).

2.2.4 Excitation and detection schemes of surface plasmons

(a) Coupling between surface plasmons and far-field radiation

An important feature of SPs is that in visible frequency the wavevector k_{SP} is larger than that of light in free space, which is obvious from Eq. (2.7) and also from Fig. 2.3. This means that SPs on a plane interface cannot be excited by light that propagates in free space. Excitation of SPs by light is only possible if the wavevector component of the exciting light can be effectively increased over its free-space value. There are several approaches for overcoming this difficulty, including using ATR scheme via prism coupling, launching SPs by employing near-field optics, or using a diffraction grating to directly match the wavevector of the incident photon to that of the SPs.

(a1) ATR couplers

A dielectric medium ($\epsilon_2 > 1$) makes it possible to increase the wavevector of light. If the p-polarized light is reflected with angle θ_0 in the dielectric medium, its wavevector becomes $\omega/c \sqrt{\epsilon_2}$ instead of ω/c . The in-plane wavevector component of the light is $k_x = \omega/c \sqrt{\epsilon_2} \sin\theta_0$. If the SP excitation matching condition is satisfied,

$$k_{SP} = \sqrt{\epsilon_2} \sin\theta_0 \frac{\omega}{c} = \sqrt{\frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2}} \frac{\omega}{c} \quad (2.21)$$

, the photons of light can be coupled to SPs located at the interface between metal and dielectric.

Fig. 2. 4 ATR couplers. (a) The Otto configuration. (b) The Kretschmann-Raether configuration. (c) Objective configuration to excite SPs.

There are two configurations that can realize ATR couplers by using prism to excite SPs at the metal-dielectric interface: the Otto configuration proposed by Andreas Otto in 1968 and the Kretschmann-Raether configuration proposed by Kretschmann and Raether shortly after Otto [56], which are shown in Fig. 2.4 (a) and (b). Both configurations are similar except for the location of metal film with respect to the prism. Since the Otto configuration is difficult to control the distance between a prism and a metal, the Kretschmann configuration is used more often. A minor modification to the Kretschmann configuration makes use of a high numerical aperture

oil immersion objective lens [64], as shown in Fig. 2.4 (c). The index matching oil is used to optically couple metal coated glass slides to the objective lens. This configuration allows for exchange of the metal film, making it is well suited for applications in biosensing. However, due to the limitation of incident angle, the ATR method can only excite low frequency in-plane SP waves propagating in one direction. The huge system size is also not suitable to apply in optical integrated devices.

(a2) Near-field couplers

Fig. 2. 5 Apparatus of launching SPs on a metallic film by a near-field tapered fiber probe.

The alternative method is the near-field optical microscopy techniques [65], as shown in Fig. 2.5. The typical geometry is a probe tip of aperture size much smaller than wavelength of incident beam illuminating the surface of a metal film in the near-field. Due to the small aperture size, the light transmitted from the tip will consist of increased wavevector components, thus allowing phase-matched excitation of SPs. Specifically, for a surface with defects like shallow pits and holes, the additional momentum for direct excitation of the SPs can be provided. With these approaches, the region of the area for launching SPs can be greatly reduced. However, the launching efficiency is generally lower than that of ATR technique and light is typically incident from the above metal surface, which could be troublesome in biosensors and fluorescence imaging.

(a3) Diffraction grating couplers

Fig. 2. 6 Grating configuration to excite SPs.

The additional light momentum to excite SPs can be provided by light scattering at a periodic corrugation on a metal surface, as shown in Fig. 2.6. The incident plane wave is diffracted by such a periodic structure into many orders. Some orders correspond to propagating plane wave. Others are of evanescent wave which can drive surface charge oscillations to form SPs when the incident light is p-polarized and momentum match condition is satisfied. For this type of SP coupler, the increase of the wavevector necessary to match the SP momentum is achieved by adding an additional grating momentum to the free-space wavevector. This requires that the metal surface is structured with a period structure with pitch Λ , such as grating or holes array. For the simple one-dimensional grating structure, the SP phase matching takes place whenever the condition $k_{SP} = k_0 \sin \theta \pm mG$ is fulfilled, where θ is the angle of incident light, $G = 2\pi/\Lambda$ is the grating constant and $m = (1, 2, 3 \dots)$. This method can preferentially excite a particular SP mode with high efficiency. The compact configuration makes the grating coupler well suitable for applications in on-chip optic-electro plasmonics devices. In this thesis, subwavelength slits are employed to launch SPs propagating along the metal/dielectric interface.

(b) Detection of surface plasmons

SPs are bound to the metal/dielectric interface, which brings complexity to detect them. Imaging of their propagation with far-field optics will be available only when the SPs are coupled back to photons first. Three main methods have been reported in literature to detect SPs.

(b1) Leakage radiation microscopy

Fig. 2. 7 The scheme of leakage radiation microscopy

The first method is to detect the leakage radiation [65-67]. If light can couple into the SP mode through the substrate, light from the SP mode is able to couple out through the substrate as well. If the surface is rough, roughness will scatter SP into far-field radiation that can be detected with a microscope, as shown in Fig. 2.7. Drawback of this technique is that the scattered field is measured, rather than the SP itself.

(b2) Fluorescence imaging

The second technique is aided by fluorescence [68]. With sufficient low concentrations, the fluorescence does not perturb the SP under investigation. Therefore, with this technique, one can visualize the SP mode. This technique

provides a useful tool to study the propagation of SPs and their interaction with surface structures and optical components. However, the resolution of this technique is limited by diffraction, plus with the shortcoming that all phase information is lost.

(b3) Scanning near-field optical microscope

The most common method of studying SP propagation is the application of NSOM [69, 70]. A NSOM system composes of a near-field detector and far-field illumination. The SP waves have a typical decaying length in the order of hundreds nanometers. A bare tapered or metal-coated glass fiber is dipped into the SP field to couple some of the light into the probe, where SPs are converted into propagating modes and guided towards a detector. The resolution of this technique is limited by the size of the tip aperture, which can approach diameters of 100 nm or even less when etching techniques are used.

In this thesis, we apply NSOM to record the intensity distribution of SPs propagating at the metal/dielectric interface. The details will be demonstrated in the following chapters.

2.3 Overview of surface plasmons modulation

As mentioned in the previous section, the behaviors of SPs on metal surface are similar to light waves in two-dimensional spaces. This implies that plasmonic components can be achieved by employing the same principle of electromagnetic waves. SPs can be modulated either in amplitude, via scattering, reflection and diffraction by surface submicron and/or nanometer scale structures, or in phase velocity modulation when deterministic surface relief dielectric structures on the metal film are designed and optimized quantitatively.

2.3.1 Modulation of SPs by metallic structures

Many achievements have been demonstrated the tailoring of SPs by scattering and reflection of SPs at the defect-mediated metal surface to realize conventional counterpart elements.

Fig. 2. 8 Nanoparticle SP beamsplitter. (a) SEM image of nanoparticles build beamsplitter. (b) Corresponding fluorescence image.[71]

In 2002, Ditzlbacher *et al.* described a scheme to control SP in a way we do control light waves with nanoparticles [71]. Figure 2.8 shows a SP Bragg mirrors with a reflectivity of about 90%, which is realized by depositing five lines of gold nanoparticles on the metal surface. In addition to SP optical elements, SP beamsplitter with well controlled transmission/reflection ratios was built from single lines of closely spaced nanoparticles [72]. SP interferometer also displayed the similar properties as a conventional interferometer. The detected maximum of the interference either on the left side or on the right side of the beamsplitter is due to a phase difference between the reflected and the transmitted part of SPs, in analogy to the conventional photonic beamsplitter.

In 2001, Sergey I. Bozhevolnyi *et al.* demonstrated the realization of plasmonics crystals by depositing 400-nm-period triangular lattices of gold bumps on

a gold thin film [73-75]. SPs are excited at the nanostructured gold/air thin film interface for incident free-space wavelengths in the range of 740-820 nm. Using NSOM, SPs are observed to be reflected by triangular lattice and guided along line defects at 782nm, as well as significant deterioration of these effects at 815 nm, thereby directly demonstrating the SP band gap effect and showing first examples of SP channel waveguides in surface band gap structures.

Fig. 2. 9 Plasmonic lens for SP focusing. (a) The scheme of SPs focusing by circular structures milled into optically thick metallic films. (b) NSOM imaging of plasmonic lens. [76]

Other designs of subwavelength-sized metallic structures were demonstrated to manipulate SPs propagating by diffraction of surface structures. Plasmonic lens for SP focusing is reported by diffraction of incident light from circular and elliptical subwavelength structures milled in metallic films, as shown in Fig. 2.9 [76]. Enhanced optical transmission and surface plasmon resonant cavity effects were also studied with subwavelength aperture arrays surrounded by rectangular-shaped Bragg mirrors [77].

Subwavelength-sized metallic surface structures hold a lot of promises for the next generation of integrated photonic elements. However, SPs have low reflection coefficients, less than 30%, by designing metallic surface defects such as groove

arrays as multilayer mirror [66]. The amount of energy reflected is likely to be insufficient for some specific applications. Furthermore, it remains a challenge to modify the optical properties of metals by external modulations for applications in active optical devices. Therefore, the design of subwavelength-sized dielectric structures for phase modulation of SPs is one of the alternative approaches to solve the above problems.

2.3.2 Modulation of SPs by dielectric structures

The interaction of SPs with surface dielectric structures is interesting, for a variety of reasons, such as high modulation efficiency, simple fabrication process and possibility for direct external modulations. Recently, the dielectric structures of defined geometry size deposited on top of a metal film have been demonstrated to realize SP focusing, bending, refraction, and total internal reflection by controlling spatial variation of the SP velocity [67, 78]. Several dielectric based plasmonic components have been reported, such as SP waveguides [79] and superlens for subdiffraction limited imaging [80].

Fig. 2. 10 Dielectric structures for SP phase modulation. (a) Locally launched SPs interacting with a cylindrical and (b) a triangular SiO₂ structure. [67]

Figure 2.10 illustrates a thin cylindrical SiO₂ structure deposited on the Au film to focus SP with a FWHM of 5 μm and a triangular SiO₂ structure for refraction and reflection of SP waves by R. Aussenegg *et. al* [67]. Other designs of dielectric structures shows the similar SP phase modulation behaviors, such as strips on gold for bending [78] and polymer prisms for SPs guidance [79]. Furthermore, a magnifying superlens based on multilayer photonics metamaterials, which consist of alternating layers of positive and negative refractive index, was designed to achieve a resolution on the order of 70 nanometers [36, 80].

Over the years, several of new SP modulation schemes have been developed, such as thermal effect modulation [81] and electro-optic modulation [82]. As these techniques are not referred to in the later portion of our research area, in-depth methodologies will not be discussed in this thesis.

2.4 Chapter summary

This chapter presents a detailed theoretical background of surface plasmons and modulation methods by using subwavelength-sized surface structures. The introduction of these unique properties of SPs and modulation methods forms the groundwork for further research work done in this thesis. Phase modulation of SPs by dielectric subwavelength-sized structures will be explored in chapter 4, and realization of SP standing wave patterns will be studied by using of subwavelength-sized metallic slit array structures in chapter 5.

CHAPTER 3

FABRICATION AND CHARACTERIZATION METHODOLOGIES

3.1 Chapter introduction

From previous section 2.2.4, we have seen that one way for momentum matching between light and SPs is to introduce periodic structure on the metal surface. To obtain large-scale gratings in metal films for this purpose, holographic photolithography techniques have been widely used. However, local excitation and the control of SPs require grating pitch of sub-micro scale as well as sharp edge of metallic slits. Instead of optical lithography methods, electro beam lithography (EBL) has been proven to be a very powerful tool because of their ability to define diverse structures with high-resolution. Therefore, we apply the EBL, in combination with thermal evaporation and lift-off techniques to fabricate subwavelength metallic gratings. In this chapter, EBL fabrication techniques are introduced. In particular, a method combining dose and pattern shape correction is utilized to minimum the proximity effect in our EBL fabrication.

Moreover, as discussed in previous section 2.2.4, since SPs are surface waves and highly confined into regions of space much less than the free space light, it can

not be observed before it is scattered into light by surface defects or special functional structures. One possibility to exploit the near-field field for high-resolution optical imaging is NSOM. This chapter also introduces the imaging of SP fields by NSOM and understanding of different near-field probes.

3.2 Fabrication of subwavelength structures by using electron beam lithography

3.2.1 Introduction of electron beam lithography methodology

EBL is a high-resolution lithographic technique for nanostructure fabrication. The primary advantages of EBL include the ability to fabricate features much smaller than optical techniques which are limited by the wavelength of light used. The first experimental electron beam writing system was developed in the 1960s to take advantage of the high-resolution electron beam resist, polymethyl methacrylate (PMMA) [83].

(a) EBL system

In brief, the EBL technique is the practice of scanning electron beam across photoresist and selectively removing some part of regions of the resist to create very small structures in the resist. Currently, the EBL system is variously used for photolithography mask creation, advanced prototyping of integrated circuits and nanoscale scientific research. The critical dimension of pattern transferred by EBL can reach below 10 nm.

Fig. 3. 1 e_LiNE - Ultra high-resolution electron beam lithography and nanoengineering workstation (Raith GmbH) [84].

Figure 3.1 illustrates a typical commercial electron beam lithography system including the column, chamber, control electronics and computer [84]. The column is modified by a set of electrostatic and/or magnetic lenses for focusing the electron beam. The electromagnetic deflection coils are used to scan the beam across the working field which is typically $100\text{ }\mu\text{m}$ -1 mm on a side. A set of electrostatic beam blanking plates is used to deflect the e-beam out of the electron optical path when needed. Underneath the column is the vacuum chamber containing a laser-interferometer stage to move the sample between the exposures of different working fields and allows sample loading and unloading. A set of controlling electronics supplies power and signals to the system. The computer system handles loading and unloading the sample, aligning, focusing the electron beam and an exposure job.

EBL system usually uses thermionic sources with low resolution. However, in high-resolution EBL systems, a thermal field emission source such as heated W/ZrO emitter is preferred to offer bright and stable stream of electrons. Typically the tungsten tip of the field emission source is heated to around 1800 K to provide very high electric fields. It requires a vacuum in the range of 10^{-8} Torr, which is usually achieved by an ion pump to the thermal emitter.

In EBL, the electron wavelength has no impact on the resolution of developed resist pattern [85], which is limited by additional factors such as electron beam size, forward and backward electron scattering effects, molecule size of any given resist.

(b) Electron beam resists

The usual electron beam resists are divided into two types: positive and negative. A positive resist is a type of resist that the electrons induce chemical chain scission, such as PMMA and ZEP 520. After developing, the exposed portion of the positive resist is removed away. Whereas in the case of negative resist, such as Hydrogen silsesquioxane (HSQ) and SU8, the exposed portion remains after development due to chemical chain cross-linking during irradiation. A positive tone resist may be employed if the pattern to be transferred is fairly sparse. Negative tone resist might be used if large area of resist needs to be removed away.

Fig. 3. 2 The sensitivity curves of photo resists. (a) Gradation curve of a sensitive (A) and less sensitive (B) negative resist. (b) Gradation curve of a sensitive (C) and less sensitive (D) positive resist. [86]

The general requirements for electron resist materials include high-resolution, sensitivity and contrast [86]. The sensitivity of a resist defined by the dose is strongly influenced by many parameters, including beam energy, resist thickness, substrate material, proximity effects and developer temperature. Generally, slow resists are utilized to obtain high-resolution. Contrast is defined in terms of the rate of a resist undergoes chain scission or cross-linking as a function of dose. Higher contrast resists are very desirable for obtaining better edge definition and higher resolution. Figure 3.2 shows the gradation curve of positive and negative resist. As a general rule,

positive electron resists exhibit better contrast than negative resists. Therefore, positive resist PMMA is utilized to fabricate sharp edge subwavelength structures in this thesis.

Positive resists are developed in a good solvent for the removal of irradiated areas, followed in a combination of good and poor solvent for the removal of unirradiated polymer. In experimental, a predetermined good-poor solubility ratio solvent is usually utilized to minimize the exposure dose. Under most developer conditions, the solubility of a uniformly irradiated polymer is linear with the time. Negative resist development is only required to choose a good solvent to remove the unexposed resist because the cross-link areas are not usually soluble. The strength of the developer is also important to determine the resolution of negative resist.

PMMA (MicroChem) is one of the most common used positive tone e-beam resist because of its very high-resolution and long shelf life. There are two typical kinds of molecular weight PMMA resists for e-beam lithography, including 495K and 950K, which both can be diluted in either anisole or chlorobenzene for spin-coating. After resist spinning, PMMA layer is typically hot-plate baked several minutes or heated in oven several hours to remove solvent. When exposed to high energy electrons, the chemical chain of PMMA is broken, which makes it more soluble in a developer solution. The mixture of methyl isobutyl ketone (MIBK) and isopropanol (IPA) (1:3) is an high contrast developer for nanolithography in PMMA [87]. Presently, the best resolution of obtained feature is on the order of 10 nm with thin PMMA resist and high electron beam voltage [88, 89].

Hydrogen silsesquioxane (HSQ) is one of the high-resolution negative tone e-beam resists. It is available from Dow Corning under the product code Fox -12 or Fox -23. After exposure, polymer chemical bonds cross link. Unexposed area can be

removed in tetramethyl ammonium hydroxide (TMAH) developers. The sub-10-nm resolution has been reported with HSQ [90]. A major advantage of HSQ lies in its etch resistance to oxygen, which makes it ideal in a bilayer resist scheme. The key issue of HSQ is its ability to spontaneously cross link in small amounts, causing calamity in pattern transfer.

SU-8 (MicroChem) is another commonly used epoxy-based negative electron resist. It can be spun to the thickness from 0.1 micrometer up to 2 millimeters, allowing the fabrication of structures with a wide range of thicknesses. The aspect ratio of SU-8 pattern can reach to 20 [91]. Furthermore, SU-8 is highly transparent in the ultraviolet region and high stability to chemicals and radiation damage. It is now mainly used in the fabrication of microfluidics, micro-electromechanical system parts. The typical electron dose for SU8 is about an order of magnitude less than for PMMA. It can be used as EBL resist as well as near UV photoresist in low cost fabrication. Adhesion of SU-8 is also usually good. Therefore, SU8 is utilized to fabricate optical phase element in this thesis.

(c) Voltage and current

In EBL system, the energies of the electrons are determined by the beam voltage. Since the electrons are negatively charged particles, the electron beams have a broaden tendency due to the interparticle electrostatic repulsion. High beam voltages result in a relatively tight beam due to the short electron interval time. The typical beam voltage is about 20 kV.

The beam current influences the quality of the beam. Lower beam currents produce high beam compactness and good focusing. However, it leads to longer exposure times.

(d) Proximity effect

Fig. 3. 3 Proximity effect by formation of an excitation volume in electron beam lithography [86].

In EBL, the resolution does not accurately depend on the diameter of focused electron beam. Electrons penetrating a conventional resist generate forward and backscattering due to interaction with the atoms in that material. Since most forward scatter of the electrons undergoes only small angles scattering, it has not much influence on the electron beam and can be minimized by reducing the resist thickness and utilizing a high accelerating voltage. However, backscattered electrons from substrate will continue to interact with the resist and contribute to the exposure for a relatively long distance. This effect is a major contributing factor to broaden the incident current distribution. The deposition of electron beam energy around the actual beam area due to scattering is named proximity effect of electron beam lithography [86], as shown in Fig. 3.3. The proximity effect results in gradient decrease of the exposure starting from the center of the region. The range of the proximity effect in the substrate depends on both the energy of the primary electrons

and type of the substrate. Low atomic number substrate materials give less backscattering. Large primary electron dose results to more backscattering. Proximity effect is usually the main resolution limitation factor in electron beam lithography. However, it is possible to partially compensate this effect by utilizing higher exposure dose at the edges of the structures or a small structure, compared to lower dose inside extended areas. Alternative compensation method is the pattern correction based on altering the shapes of the patterns to correct resist exposure. In this thesis, a technique which combines dose control and alteration of pattern shapes is utilized as a very flexible method of correcting proximity to achieve adequate pattern fidelity.

(e) Pattern transfer methods

In high-resolution lithography, one of the most critical operations is the transfer of pattern in the resist to the substrate. There are many different kinds of transfer processes such as solution etching, plasma etching, lift-off, electro plating and Ion sputtering. For each particular process, the critical variables are totally different. However, some general characteristics have emerged from the literature. For a given pattern transfer material, one type of transfer processes may be more effective than others. For example, the lift-off technique is often used in the fabrication of very high-resolution metal patterns.

Fig. 3. 4 The lift-off technique used in defining metal patterns: (a) the PMMA resist after development; (b) the structure after metal deposition; (c) the desired metal pattern left on the substrate after resist removal.

Figure 3.4 shows the common lift-off technique. The resist is irradiated and developed to produce an undercut profile. Metal evaporation methods, for example, electron beam evaporation, Sputtering evaporation or thermal evaporation, are utilized to deposit metal film over the entire substrate. After the metal film was evaporated on the patterned resist, the sample is dissolved in acetone. When the remaining resist is dissolved with the removal of metal on top, desired metal pattern stays firmly on the substrate. With this technique, positive resist is preferred to provide an undercut profile as long as the dose is sufficiently high. Whereas, negative resists typically have round profiles and are not suitable in lift-off. Moreover, for good lift-off technique, vertical material deposition process is necessary. In addition, the substrate must be kept below the resist transition temperature. Therefore, in the experiments described in this thesis, all metal films are vacuum-deposited by thermal evaporation due to its low temperature and vertical evaporation conditions.

The poor adhesion of metal film to the glass substrate is especially important in lift-off technique. The standard addressing to it is to evaporate several nanometers Ti or Ge film before target metal deposition. However, these materials have an influence on the SPs mode at metal/dielectric interface. Therefore, in this thesis, gold or silver metal materials are directly thermal evaporated on the substrates to fabricate subwavelength-sized plasmonic structures.

3.2.2 Fabrication of subwavelength grating structures

From previous discussion, in order to minimize the proximity effect of EBL, a method combining dose and pattern shape correction is utilized in our fabrication of designed subwavelength grating coupler structures.

Fig. 3. 5 GDSII design pattern for proximity effect correction

Figure 3.5 displays the corrected GDSII design pattern of subwavelength-sized slit structures. At the end of slit, the pattern shape and exposure dose are optimized in order to get rectangular-shaped undercut PMMA structures, which is helpful in the following lift-off process. Cross-shaped features in the corner of pattern are utilized to align the second layer exposure.

Fig. 3. 6 Flow chart of fabrication process.

Figure 3.6 shows the flow chart of EBL fabrication process. After the substrate is cleaned, PMMA (A4, MicroChem) is spin-coated on an indium tin oxide (ITO) glass substrate at a spin speed of 1500 rpm in order to achieve a film thickness of 350 nm. Next, the film is pre-baked on a hotplate at 180°C for 90 seconds to remove solvent. After that, the PMMA photoresist is exposed to a scanning electron beam lithography machine (Raith e_LiNE) with accelerating voltage of 20 KeV and beam current of 160 pA. This high electron beam voltage and low current are helpful in order to get undercut shaped PMMA slits for easy lift-off. The film is then dissolved in the developer (MIBK: IPA = 1:3) for 120 seconds and rinsed with IPA for 40 seconds to remove the exposed PMMA areas.

Fig. 3. 7 (a) SEM imaging of PMMA grating structure. (b) SEM imaging of metal grating structure.

Part of the developed PMMA grating structure is shown in Fig. 3.7 (a). Rectangular-shaped grating end is preferred to the following lift-off process. After that, the sample is thermally deposited with Au film of 100 nm thickness by using physical vapor deposition machine at 0.1 nm/s speed. Such a low evaporation speed is preferred to get smooth metal surface, which is important to reduce SP propagation loss. Finally, the film is immersed into acetone solvent for lift-off to remove the Au on non-exposed PMMA areas. The SEM imaging in Fig. 3.7 (b) shows that the subwavelength-sized slit array is obtained through proximity correction. The sharp edge of slit is critical to ensure high SP excitation efficiency when design subwavelength-sized slit arrays as the SP coupler in plasmonic devices.

The subwavelength-sized dielectric and metallic structures used in this thesis are all fabricated by EBL with the similar process. The detail results will be discussed in the respective chapters.

3.3 Characterization of SPs by using near-field scanning optical microscopy

3.3.1 Introduction of near-field optical scanning microscopy

NSOM is a microscopic technique for surface feature investigation that breaks through the far-field diffraction limit by utilizing probe much close to the specimen surface for excitation or collection of diffraction in the near-field. The idea of employing near-field optics to achieve images beyond the diffraction limit was first discussed in 1928 by E. H. Synge [92]. Several tens years later, Pohl and Lewis introduced the usage of a subwavelength metal coated optical tip and a feedback mechanism to achieve resolution as low as 25 nm [93, 94]. In NSOM system, the resolution is limited by the aperture size of the detectors instead of the wavelength of the illuminating light. NSOM is a powerful technique for studying nano scale optical interactions.

There are three basic NOSM operational modes: illumination mode, reflection mode and collection mode. In illumination mode operation, the light is coupled into the optical probe as the source to illuminate a sample in the near-field. A high numerical aperture objective lens is positioned under the sample to collect the transmitted light and focus it into a photodetector. Reflection mode operation has the similar principle as illumination mode except that the photodetector is set to collect the light reflected from the opaque sample surface. In collection mode operation, the light is coupled into objective lens to illuminate the sample. The NSOM probe collects the electromagnetic field on the sample surface and delivers it to the photodetector through the fiber.

Fig. 3. 8 (a) Aperture-type NSOM system (NT-MDT). (b) Tuning fork with glued probe. [95]

Figure 3.8 (a) displays a commercial aperture-type NSOM (NT-MDT NTEGRA Solaris) [95], incorporated into an inverted optical microscope, with far-field excitation and near-field detection. The optical image of sample surface is collected point to point by scanning either the probe or the sample stage. The scanning routine can either work at a constant height or regulated height by using a feedback mechanism. Tuning fork share-force feedback technique, as shown in Fig. 3.8 (b), is used to control the distance between NSOM probe and sample surface. Then, light propagating through the fiber is directed to sensitive detector, such as photo-multiplier tubes (PMT). After that, the PMT signal is transmitted to the computer where image processing is employed to obtain the final optical image.

Fig. 3. 9 (a) The metal-coated tapered fiber. (b) The apertureless metal tip. [96]

The most commonly used NSOM probes are apertureless type and aperture type, as shown in Fig. 3.9. They are positioned close to a surface to couple the evanescent electromagnetic field into the far-field, which are collected by a photodetector. The apertureless metallic tips are very sharp (5 to 15 nm in diameter). It is operated by scattering the radiation. Generally, the aperture size determines the resolution of the NSOM images. The spatial resolution of an apertureless metallic tip can be much higher than that of an aperture probe, because of the small radius of the tip apex. However, the tip-surface interaction significantly affects the local electromagnetic fields. The use of apertureless metallic tips close to a metal surface results in a local SP field enhancement by several orders of magnitude, which cause an artifact in the imaging [96]. In this case, apertureless metallic tips are not usually used to detect SP fields above the metal surface.

Fig. 3. 10 (a) Straight NSOM fiber probe (b) Cantilevered NSOM fiber probe. [96]

An aperture fiber probe has a transparent core with/without a metal coating, and can be fabricated with either a straight or bent tip, as shown in Fig. 3.10. Bent probes are normally operated in tapping-mode feedback, while the straight optical fiber ones are operated in shear-force mode feedback.

Fabrication of aperture probe is usually done with either chemical etching or heated pulling. The end of tip is generally from 50 to 100 nm in diameter. A metal coating is subsequently deposited on the walls of the probe to reduce light loss through the tapered region of the fiber. Aluminum or aluminum/chromium layers are often used for metal coating because of small penetration depths of the electromagnetic field in the visible range. After aluminum evaporation, focus ion beam (FIB) is commonly used to precisely shape the tip end to get different aperture diameters. Compared to coated fiber tips, uncoated fiber probe brings with much smaller perturbations in the measurement of electromagnetic field on the metal surface. However, the light leakage from bare fibers would lead to poor signal collection efficiency as well as low signal-to-noise ratio [96].

3.3.2 Characterization of surface plasmons

The near-field scanning optical microscope used in this thesis for characterization of SPs is a commercial model (NTEGRA Solaris) from NT-MDT with collection mode. The near-field SP image is built up by the scanned light intensity detection in the near-field as the probe scans over the sample surface.

Fig. 3. 11 The schematic setup of NSOM experimental setup.

The schematic setup for detection of SP field distributions on the metal surface is displayed in Fig. 3.11. The excitation light originated from a laser of wavelength $\lambda=633\text{nm}$ (0.5 mm beam diameter, 30 mW). The telescope system is utilized to modulate the wavefront of beam. Polarizer and half wave plate are used to select the desired polarization of laser light. After reflected by the prism, the laser light is collimated normal incident onto the sample to excite SP waves by an objective lens

(4×, NA=0.13). On the other hands, NOSM fiber probe is mounted on the piezo-tube to be controlled to scan just above the sample surface. The evanescent field of the SP is frustrated and converted into guided modes within the fiber. The output end of the probe is connected to a PMT to amplify the signal. The control of the scanners and data acquisition are usually driven through additional electronics and computer. After the image information is processed in computer, we finally obtain the near-field image of the SP field on the metal surface.

In order to detect SPs with high efficiency, an aluminum coated NOSM fiber tips is used in experiments. The results show that the artifact perturbation from SP interaction with metal surface is not obvious. The main reason is that the SP field signal is very strong. The metal coated fiber with optical transmission of 5×10^{-5} and aperture size of 100 nm, as well as low scanning speed of 10 $\mu\text{m/s}$, also helps to suppress artificial noise.

In our NSOM system, the tune fork glued with fiber tip is derived to resonate at the frequency of 190K Hz. Compared to optical frequency 5×10^{14} Hz, it has much lower speed fluctuation. The signal collected by NSOM probes is time averaged to build up near-field image. Therefore, the lateral vibration of probe would not have much effect on the measurement accuracy. Additionally, feedback control system is used to control fiber probe vertical fluctuation above the metal surface in 8 nm region. Compared to several hundreds of SPs penetration depth, the effect of small vertical fluctuations between probe and surface on the measurement accuracy is estimated to be less than 3%, which can be negligible.

The characterization results of SP field distributions will be discussed in the following chapters for the different design of plasmonic structures.

3.4 Chapter summary

This chapter presents a description of the electron beam lithography methodology for fabrication of subwavelength-sized plasmonic structures and the near-field scanning optical microscopy for detection of SP waves. These methodologies will be utilized for the further work done in the thesis. The detailed fabrication and characterization results will be displayed in the following chapters.

During the fabrication of subwavelength-sized structures, Madam Bu Jing gave me many instructions and valuable suggestions on electron beam lithography. I am grateful for her kind help.

CHAPTER 4

PHASE MODULATION OF SURFACE PLASMONS BY USING SUBWAVELENGTH-SIZED DIELECTRIC STRUCTURES

4.1 Chapter introduction

The most attractive properties of SPs lie in the fact that they are surface waves highly confined at a metal/dielectric interface, thus light manipulation can be restricted in two dimensions. This significantly simplifies the procedure of modulation of SPs into nanoscale region and offers much feasibility of developing new types of on-chip photonic devices. Many achievements have been made in tailoring SPs for specific applications by utilizing metal nanostructures to control the scattering of SPs at the surface [71, 97, 98]. For example, plasmonic Fresnel zone plate, consisting of metallic nanostructures covered with a dielectric layer, demonstrated numerically the ability of focus beams into subdiffraction with high intensity [99]. Another similar design of plasmonic micro-zone-plate structure embedded in silver film is numerically presented to super-focus beams at a working distance at the micrometer scale with elongated depth of focus [100, 101]. However,

most of these metallic plasmonic structures indicate a rather weak interaction of SPs with surface defects because of poor experimental control of the defect geometry. Therefore, dielectric structures have been proposed as a promising alternative in order to facilitate the implementation of plasmonic devices. By utilizing the dielectric elements instead of metallic elements, it is also possible to improve the performance of plasmonic devices and simplify the fabrication process.

To date, many efforts have been devoted to the control of the propagating properties of the SP light using dielectric structures, and some plasmonic components have been reported [67, 79, 102]. These results demonstrated the manipulation of SP by dielectric structures through phase modulation. However, little is known about the relationship between dielectric elements and the induced SP phase change, which is very important to realize modulation of SPs in phase for totally constructive interference, in order to improve the performance of dielectric plasmonic devices. It is therefore necessary to quantitatively investigate the phase modulation of SPs with dielectric structures readily implemented on a metal film. In this chapter, with the consideration of SP losses, a novel modified effective-refractive-index model for optimization of dielectric structure's dimensional size is proposed in realization of phase modulation of SPs to constructive interfere. Furthermore, plasmonic phase modulated Fresnel zone plate is experimentally realized to integrate the excitation, modulation and totally constructive interference of SPs to a subwavelength spot, which potentially act as a virtual probe to excite fluorescence dyes for high-resolution fluorescence imaging.

4.2 Phase modulation of surface plasmons

4.2.1 Theoretical modeling

(a) Design of grating coupler

Despite of various techniques have been introduced to couple the free space light into SP waves through ATR configuration or near-field optical microscopy, these techniques require a complex experimental system. In this thesis, grating coupler, of its compact size, planarity and stability, is utilized to excite SPs on the metal surface.

Noble metals, such as gold (Au) and silver (Ag), are usually deposited on the substrate for SP excitation and propagation. However, Au is chemically stable compared to silver material, thus suitable to be deposited on the substrate as the first layer subject to sustain the high temperature in second layer fabrication process. In order to manipulate SPs in more efficient ways, it is important that the SPs have a propagation length of at least several times of their wavelength. Although the ohmic damping of SPs is strong, propagation lengths of a few tens of micrometers are reported for SPs excited at optical frequencies on the Au film [63]. It is sufficiently large enough for manipulation of SP waves on the Au surface with the aid of structured patterns. Therefore, we choose Au film, where the complex permittivity (ϵ_m) is approximately $-9.51 + 1.13i$ of wavelength 633 nm, to be deposited on a glass substrate for SP excitation and propagation.

Since the laser beam possess a Gaussian profile, the distribution of excitation angles will have a maximum around 0 degrees. As the previous discussion in section 2.2.4, the maximum SP coupling efficiency would therefore be expected on the

grating with a pitch (Λ) comparable to the SP wavelength (λ_{SP}) under normal incidence, as the following

$$\Lambda = \lambda_{\text{SP}} \quad (4.1)$$

The calculated wavelength of SPs is 598 nm with 633 nm excitation light. According to Eq. (4.1), grating pitch is designed as 600 nm to match the momentum of SPs under normal illumination.

The thickness of the Au film is designed to be 100 nm, which is sufficiently thick to block the directly transmitted light, ensuring that all electromagnetic fields at the Au/dielectric interface beyond the gratings originate from SP waves.

Slit width plays a vital role in the design of grating coupler. Commercial software XFDTD (Remcom Incorporated) for vectorial three-dimensional electromagnetic calculation is used to analyze the SP field excitation efficiency by using grating couplers of different slit widths.

Finite-difference time-domain (FDTD) methodology is a grid-based differential time-domain numerical modeling method. It was proposed by Kane Yee since the 1966 [103], but has gained great popularity in recent years with the increased performance from computers. With the FDTD approach, both space and time are divided into discrete segments. The electric and magnetic fields are calculated using a leapfrog scheme where first the electric fields, then the magnetic are computed at each step in time. FDTD is capable of simulating a wide variety of electric and magnetic materials. The most basic material of course is the free space. Frequency independent dielectric materials are defined by their parameters of relative permittivity and conductivity. More details of FDTD's working principle can be found in Ref. [104].

In XFDTD program, the complex permittivity of Au material is described by the expansion of Drude dispersion relation as $\varepsilon(\omega)=\varepsilon_{\infty}+(\varepsilon_s-\varepsilon_{\infty})/(1+j\omega t_0)+\sigma/(j\omega\varepsilon_0)$, where $\varepsilon_{\infty}=9.0157$ is the infinite frequency relative permittivity, $\varepsilon_s=-1.29569\times 10^4$ is the static relative permittivity, $t_0=8.41395\times 10^{-15}$ s is the relaxation time, $\sigma=1.36458\times 10^7$ S/m is the electrical conductivity and $\varepsilon_0=8.854\times 10^{-12}$ F/m is the free space permittivity. All these parameters are set up to match the empirical data of the real and imaginary parts of the gold's complex permittivity over a wavelength range from 0.3 μm to 1.2 μm . Liao numerical radiation boundary is used to absorb all radiated or scattered electromagnetic fields with very little reflection from the boundary.

Liao boundary is a second-order stabilized radiation boundary [105]. It allows electromagnetic fields to be radiated or scattered by the FDTD geometry and to be absorbed with very little reflection from the boundary. Compared to perfectly matched layer (PML) boundary, Liao is an estimation method. By looking into the FDTD space and back in time, it estimates the electric fields just outside the limits of the FDTD mesh. These estimated values are then used in the FDTD equations inside the space. The Liao estimation assumes that waves are allowed to travel outward from the space but not reflect back in. The Liao method works well provided that there is enough space between the radiating geometry and the outer boundary. Typical limits are at least 15 cells spacing.

A spatial monitor is set on a plane located 20 nm above the structure to obtain the electromagnetic field distributions. Figure 4.1 displayed the XFDTD simulation model of SP grating coupler.

Fig. 4. 1 XFDTD simulation model of SP grating coupler.

The simulation results showed that the amplitude of SP fields excited by grating is varied with different slit width, and the excitation efficiency reaches a peak when slit width is little smaller than half of wavelength of SPs under normal incident condition. These simulation results are in consistent with grating diffraction theory where a majority of the light is diffracted when the slit width is designed to smaller than half of the incident wavelength of the illumination light [1]. Therefore, the slit width of grating coupler is designed to be 245 nm in our case.

(b) Modeling of phase modulation of SPs

As discussed in the previous section 2.3.2, one approach of controlling SP propagation is to use phase modulation with deterministic surface relief dielectric structures deposited on the metal film. It is necessary to quantitatively determine the relationship between the SP phase change and the three-dimensional sizes of dielectric structures. Therefore, a numerical experiment is implemented on phase modulation of SP where the geometric sizes of the surface relief structures are varied and finally optimized.

In our proposed simulation model, as shown in Fig. 4.2, a subwavelength-sized slit array is embedded in the Au film as a SP launcher. A linearly polarized Gaussian beam ($\lambda=633\text{nm}$) is lightly focused by objective lens ($\text{NA}=0.13$) onto the

grating to excite in-phase SP propagating along two opposite directions at the Au/dielectric interface.

Fig. 4. 2 XFDTD simulation model of phase modulation of SPs.

A PMMA structure with a definite thickness (t), width (w), and length (l) is loaded above the Au film to modulate the phase of SP waves. Similar to conventional optical phase devices, the phase difference ($\Delta\phi$) between the transmitted SPs through the dielectric structures and the directly propagating SPs is determined by the size of the dielectric structures and the effective-refractive-index (n_{eff}) difference, as

$$\Delta\phi = \frac{2\pi}{\lambda_{SP}} \left[(n_{eff}(t, w, l) - 1) \right] \times l \quad (4.2)$$

As previously discussed in chapter 2, due to Ohmic losses of the electrons, SPs decay along the interface, and due to its native evanescent waves, the normal component of SPs amplitude exponentially decreases away from the interface. Therefore, it can be predicted that the l , t and w of the PMMA structure exert different

effects on the SP phase modulation, and that the $n_{eff}(t, l, r_m)$ varies with the dimensional sizes of the PMMA structure due to exponential decay behavior of SP waves. Theoretically, for a very thin dielectric layer deposited on the Au film, n_{eff} is close to that of SPs propagating at the Au/air interface. Whereas for a very thick layer, normally much thicker than the penetration depth of SPs in dielectric, n_{eff} is close to that of SP propagating at the Au/PMMA interface. In our case, n_{eff} is normalized to 1 for the SP propagating at the Au/air interface.

With this design methodology, perfect phase modulated SPs devices are possible. The dielectric structures can be designed so that the transmitted SPs are the same in magnitude but are out of phase with the directly propagating SPs at the metal/dielectric surface. This π phase-shift condition is much useful in design of phase modulated plasmonic devices. Specifically, an in-plane phase modulation Fresnel zone plate (PMFZP) is proposed to employ the phase modulation model for SP focusing at the focal point. Our suggested PMFZP device is composed of both an embedded subwavelength-sized silt array to excite SP waves propagating along metal surface and an in-plane PMMA FZP structures deposited on the right side of the grating to spatially modulate the phase profiles of the transmitted SPs for totally constructive interference.

Fresnel zone plate (FZP) was formerly investigated by Augustin-Jean Fresnel who used it to replace a lens for an image [106]. A conventional FZP consists of a set of alternative opaque and transparent radial symmetric rings, known as Fresnel zones. The boundaries of the Fresnel zones are defined according to Huygens-Fresnel diffraction theory, with each adjacent zone of equal area, and the average phase difference of π , as the following

$$r_m = \sqrt{m\lambda_{SP}f + \frac{m^2\lambda_{SP}^2}{4}} \quad (4.3)$$

where m is an integer for the FZP's zone order and f is the designed focal length. Light passes through the transparent zone plates and diffracts around the opaque zones to form a bright focal point. A disadvantage of the conventional FZP is that only small part of the incident light contributes to the focal point [107]. However, this can be improved by replacing the opaque rings with transparent rings with a thickness equivalent to a phase change of π . This is the so-called phase reversal zone plate [108].

Fig. 4. 3 The top view of the proposed in-plane PMFZP.

In our proposed in-plane PMFZP, subwavelength-sized PMMA Fresnel zones is utilized to replace the opaque ones to retard the phase of SPs by π . Therefore, all the SPs from Fresnel zones are totally constructively interfere at the focal point. The geometry of the in-plane PMFZP is shown in Fig. 4.3. The SP focal point is designed

at $f=5\text{ }\mu\text{m}$ away from the PMFZP. For each dielectric Fresnel zone, the w is defined by Fresnel zone radius, r_m , according to Eq (4.3). Due to the remarkable attenuation of SPs at several decaying lengths away, total 26 Fresnel zones are simulated in our model, as shown in Table 4.1.

FZP: $f=5\text{ }\mu\text{m}$, $\lambda_{sp}=0.5986\text{ }\mu\text{m}$			
r_1	1.7557 μm	r_{14}	7.711 μm
r_2	2.5188 μm	r_{15}	8.0654 μm
r_3	3.1281 μm	r_{16}	8.4155 μm
r_4	3.6613 μm	r_{17}	8.7618 μm
r_5	4.1478 μm	r_{18}	9.1048 μm
r_6	4.6025 μm	r_{19}	9.4449 μm
r_7	5.0339 μm	r_{20}	9.7822 μm
r_8	5.4477 μm	r_{21}	10.1172 μm
r_9	5.8475 μm	r_{22}	10.450 μm
r_{10}	6.2360 μm	r_{23}	10.7809 μm
r_{11}	6.6153 μm	r_{24}	11.1099 μm
r_{12}	6.9868 μm	r_{25}	11.4373 μm
r_{13}	7.3517 μm	r_{26}	11.7633 μm

Table 4. 1 Parameters of Fresnel zone plate with focal length of 5 μm .

A plane SP waves are excited with the grating coupler and incident on the PMMA zones. The total amplitude at the focal point is determined by the integration over the wavefronts emitted from each zone. This phase zone plate can be treated as two separate Fresnel zone plates, one of which introduces an extra phase-shift $\Delta\phi$ by PMMA zones. The total amplitude at the focal point is the sum of these two zone

plate effects, achieving the maximum intensity because of totally constructive interference when $\Delta\phi=\pi$, and zero intensity because of totally destructive interference when $\Delta\phi=0$. In our case, the SP phase-shift between neighboring zones is modulated by varying the l and t and w of each PMMA structure.

4.2.2 Modified effective-refractive-index modeling for optimization of phase modulation

XFDTD software (Remcom Incorporated) is utilized to calculate the SP field distribution of in-plane PMFZP. The phase modulation is firstly investigated by varying the length of PMMA structures while the thickness remains constant.

Fig. 4. 4 Simulation results of PMFZP and AMFZP. Plots of electric field intensity $|E|^2$ at the focal point of the in-plane PMFZP vs. length of the PMMA structures with 100 nm thickness and AMFZP vs. length of Au zone structures.

The simulation result of SP field intensity $|E|^2$ at the focal point of the in-plane PMFZP as a function of the length of PMMA zones is shown in Fig. 4.4. It can be observed that the SP field intensity at the PMFZP focal point is gradually changed when the length of PMMA zones gradually increased while holding the thickness at 100 nm. When the length of PMMA structure is increased to 0.48 μm , the electric field intensity at the focal point reaches its maximum value. Under this condition, π phase-shift between PMMA zones and air zones results in constructive interference of SP waves from in-phase PMMA zones. Similarly, when the length of PMMA zones is increased to 0.96 μm , the PMMA zones introduce 2π phase-shift relative to air zones. The destructive interference of SPs from all zones results to a minimum intensity at the focal point. With a 1.44 μm structure length, 3π phase-shift brings with the second principal maximum of SP field intensity at the focal point, whose value is lower than the previous one mainly due to the decay of SPs along the propagating path at the metal/dielectric interface. Extrapolated from these simulation results, the continual phase modulation of SPs by PMFZP can be realized by increasing the length of PMMA structure with a specific thickness.

In order to demonstrate the significant contribution of phase modulation on SPs' focusing, we compare the electric field intensity of PMFZP at the focal point with that of conventional in-plane amplitude modulation Fresnel zone plate (AMFZP) under the same conditions but the dielectric surface structures are replaced by Au elements. Figure 4.4 displayed that the SP field intensity peak of the in-plane PMFZP is approximately 130% larger than that of in-plane AMFZP under the π phase-shift condition, which is due to the scattering loss of SPs in the opaque zones of the AMFZP.

Fig. 4. 5 Simulation results of in-plane PMFZP with different zone thickness. Plot of electric field intensity $|E|^2$ at the focal point of the in-plane PMFZP vs. l with $t=200\text{nm}$ (solid line), $t=400\text{nm}$ (dashed line) and $t=500\text{nm}$ (dotted line), respectively.

Next, the effectiveness of PMMA zones' thickness on the phase modulation of SPs is included for investigation in our numerical experiment. Figure 4.5 shows the SP intensity distributions at the focal point with different PMMA thickness of 200 nm, 400 nm and 500 nm respectively. The trends of these simulation results are similar to that of 100 nm PMMA thickness, which is shown in Fig. 4.4. It is noted that the PMFZP of larger thickness results in higher focusing efficiency, and the length of PMMA structure is smaller than that of thin PMMA zones under π phase-shift condition. This can be explained by the fact that more SP waves are confined within the thicker dielectric structures, resulting in higher effective-refractive-index.

The previous discussions have demonstrated that the effective-refractive-index $n_{eff}(t,w,l)$ of the dielectric strip is a variable of the 3D size of PMMA zones. The SP guided modes are excited when SPs propagate into a PMMA strip. The characteristics of such a waveguide configuration can be modeled by using the effective-refractive-index approximation [109]. The $n_{eff}'(t,w)$ can be roughly determined as a function of the cross-sectional geometric characteristic of the dielectric strip (w and t) while assuming an infinite length l . However, due to the SP inherent exponential decaying along the propagation direction, the conventional waveguide effective-refractive-index model needs further modification before being perfect. For instance, when dielectric structures are applied to provide a π phase-shift between the transmitted SPs and directly propagating SPs in the adjacent Fresnel zones, the energy loss difference between these two paths needs to be considered. Moreover, a shorter dielectric length l should be foreseen to offset the loss difference and thus to improve the performances. As a result, the parameter l of the individual dielectric strip is optimized to obtain the desired phase-shift value, as well as to provide an equivalent amplitude distribution of the transmitted SP fields from dielectric strips and directly propagating SPs in other areas.

The SP waveguide mode is mostly confined on the metal/dielectric interface. The roughness of the Au/PMMA interface is quite small with a root mean square roughness of less than 10 nm due to thermal evaporation process. Then, the estimated SP scattering loss at the Au/PMMA interface is less than 0.1%. Additionally, electron beam lithography process is adopted to fabricate PMMA waveguide structures. It can be estimated that the scattering loss on the dielectric/vacuum boundary is less than 0.1% due to smooth PMMA sidewall. Therefore, the scattering loss of PMMA waveguide can be negligible when compared to that from the SP ohmic loss.

The estimation of the energy losses in the waveguide comprises two main parts: SP propagation loss and reflection loss. The difference ratio of propagation loss between the adjacent Fresnel zones can be calculated to be $\exp(-2 \cdot \Delta k_{SP}'' \cdot l)$, where $\Delta k_{SP}''$ is the imaginary part difference of SP wavevectors existed in dielectric waveguide compared to other areas. The SP reflection energy loss ΔI_{SP} in waveguide can be estimated as

$$\Delta I_{SP} = I_0 R_o R_c \exp(-4 k_{SP}'' \cdot l) \quad (4.4)$$

where R_o and R_c are the SP reflectivities at the opened and closed ends of dielectric elements, respectively. By taking the calculated propagation and reflection losses into account, we finally determine the $n_{eff}(t, w, l)$ as

$$n_{eff}(t, w, l) = \frac{\alpha \times n_{eff}'(t, w)}{\exp(-2 \Delta k_{SPP}'' l) \times (1 - R_o R_c \exp(-4 k_{SPP}'' l))} \quad (4.5)$$

where α is the trial and error factor ranging from 0.95 to 1.05. By substituting Eq. (4.5) into Eq. (4.2), the phase-shift value $\Delta\phi$ of the SPs induced by dielectric structures can be calculated for any surface dielectric structures with definite 3D geometry size.

Fig. 4. 6 Calculation results of PMMA elements of different thickness, width and length in order to realize the π phase-shift of SP waves.

Figure 4.6 shows the calculated results of PMMA structures for different t , w , and l in order to realize the π phase-shift of SP waves. As expected, an increase of the t and w of the dielectric structures results in an increased of the SP field intensity confined in the dielectric region, which leads to an increment of SP loss due to Ohmic loss. As a result, the parameter l has to be slightly reduced in order to compensate for this increase of SP loss. Therefore, the length of the PMMA structures can be optimized for different w and t in order to get the same phase modulation condition by using the modified effective-refractive-index model.

4.2.3 In-plane phase modulation Fresnel zone plate

In order to compare modified effective-refractive-index model with conventional waveguide effective-refractive-index method, numerical simulations are conducted by using XFDTD (Remcom Incorporated).

Figure 4.7 (a1) to (c1) show the simulation results of PMFZP electric field intensity distribution at specific 100 nm, 200 nm, and 300 nm PMMA thickness, respectively, while length of PMMA is designed by $\lambda_{SP}/2/(n_{eff}'(t,w)-1)$ according to conventional waveguide effective-refractive-index method to realize π phase-shift. Simulation results show that the SP waves focus weakly or even out-of-focus at the designed FZP focal position. In Fig. 4.7 (a2) to (c2), where length of PMMA is optimized with the modified effective-refractive-index methods, simulation results show that SP waves strongly focus for different PMMA thickness. We can conclude that the modified effective-refractive-index model is much more accurate to optimize the physical length of each dielectric strip in the design of SP based PMFZP for maximum focusing efficiency. As opposed to the conventional FZP, the above PMFZP enjoys the advantage of separately controlling each phase-shift by changing dielectric structure's dimensional size.

Fig. 4. 7 FDTD simulation results of the PMFZP intensity distribution on a plane located 20 nm above the Au surface with 100 nm, 200 nm, and 300 nm of PMMA thickness, respectively. (a1)-(c1) length of PMMA are designed according to the conventional waveguide effective-refractive-index method. (a2)-(c2) length of PMMA are designed according to the modified effective refractive-index model. The length scale is 1 μm .

4.3 Constructive interference of SPs by using PMFZP

4.3.1 Simulation results of PMFZP

To date, various dielectric structures for phase modulation of SPs to interfere at the focal point have been demonstrated, such as the one with dielectric planar lenses based on the Kretschmann launching configurations [67, 110]. In this kind of devices, SPs are generated on the top surface of the metal film and propagate towards the dielectric planar lens. The curved surface of the dielectric is simply shaped in a parabolic manner to refract SP towards a focus. Another dielectric structure realized the focus of SP fields at a metal/air interface by using SiO₂ based SP Fresnel zone plate on an aluminum (Al) film integrated with a pair of nanohole arrays for excitation and detection of the SP fields [111]. The SP fields from each Fresnel zone interfere at the expected focal point to obtain the focusing with threefold intensity enhancement. These results have demonstrated the SP focusing by dielectric structures through phase modulation, but the focal point size is only in micrometer scale due to partial constructive interference of SPs.

By using proposed modified effective-refractive-index model, it is feasible to design dielectric structures to optimize the SP phase modulation effect to realize the totally constructive interference of SPs for nanofocusing. In our proposed PMFZP with $t=100$ nm, the l of each PMMA zone is calculated by using modified effective-refractive-index model to range from 450 nm to 600 nm with the increased zone order, as shown in Table 4.2. SPs from each zone have equal amplitude and phase, and totally constructively interfere at the focal point of PMFZP.

PMFZP: $t=100$ nm, $f=5$ μ m	
l_1	450 nm
l_2	486 nm
l_3	511 nm
l_4	531 nm
l_5	545 nm
l_6	558 nm
l_7	568 nm
l_8	576 nm
l_9	584 nm
l_{10}	589 nm
l_{11}	593 nm
l_{12}	596 nm
l_{13}	601 nm

Table 4. 2 The length of PMMA zones in PMFZP with thickness of 100 nm.

Figure 4.8 (a) shows the FDTD simulation result of the electric field of PMFZP at focus when the length of PMMA structures is optimized for a π phase-shift between the adjacent zones, while thickness is designed as 100 nm. Similarly to conventional light waves, SP waves pass through the PMMA structure and totally constructive interfere to focus at the focal point of the in-plane PMFZP. As shown in Fig. 4.8 (b), full width half maximum (FWHM) and the depth of focus (DOF) are approximately 256 nm and 433 nm, respectively by line-scanning the longitudinal and transverse of $|E|^2$ distribution through the focus point in Fig. 4.8 (a). From the

theoretical calculation by using Rayleigh criterion in free-space, the diffraction limit is approximately $\lambda/2 \approx 316$ nm; $\text{DOF} = \pm 0.5\lambda/NA^2 \approx \pm 369$ nm, where $NA = r_m/(r_m^2 + f^2)^{1/2}$ and m is the outermost zone order 26. This simulation result shows that the FMFZP has the focal point beyond the diffraction limit. The main reasons of beyond diffraction limited focusing lie in the fact that the SPs have shorter wavelength than free-space electromagnetic waves and that the SP fields are strongly confined on the metal surface. This subdiffraction sized SP spot is very interesting to be applied in in-plane high-resolution SP microscopy due to its flat configuration.

Fig. 4. 8 FDTD simulation results of the PMFZP designed with modified effective-refractive-index model. (a) The focal point SP intensity distribution of optimized PMFZP with 100 nm PMMA thickness on a plane located 20 nm above the Au surface. (b) Plots of FWHM and DOF of the focus point.

4.3.2 Fabrication of PMFZP

Our proposed subwavelength-sized PMFZP device is fabricated by a standard EBL process. Firstly, the subwavelength-sized slit array is fabricated as the SP coupler, as shown in Fig. 4.9. The pitch, width and thickness of slit array are 600 nm, 245 nm and 100 nm, respectively.

Fig. 4. 9 SEM imaging of metal grating structure.

Next, PMMA polymer is spin-coated on metal film surface for the fabrication of relief PMFZP features with the similar EBL exposure process. Four cross-shaped marks in the corner of device are designed to align the second layer of PMMA FZP fabrication with less than 20 nm alignment error. Finally, PMMA zone plate structures are deposited on the metal surface, 0.7 μm away from the right side of the grating coupler. From the consideration of fabrication, the thickness of PMFZP is typically chosen as 100 nm in experiment. Figure 4.10 displays the SEM image of such a PMFZP.

Fig. 4.10 Realization of PMFZP. SEM image of the PMFZP profile.

4.3.3 Experimental results and discussions

The SP field distribution of PMFZP device is characterized by using NSOM system (NT-MDT NTEGRA Solaris) with the collection mode. As described in section 3.3.2, in our experimental setup, a 633 nm laser beam is polarized at 90° to the gratings and is lightly focused through $4\times$ microscope objectives ($\text{NA}=0.13$) onto the metal grating structure. The SP modes $[\pm 1, 0]$ are excited at the metal surface. After transmitting through the PMMA Fresnel zones, SP waves totally constructively interfere with the propagating SPs from the bare zones to focus at the designed focusing position.

(a)

(b)

Fig. 4. 11 Performance of the plasmonic Fresnel zone plate. (a) NSOM image of the PMFZP in the focal area. The insertion is the simulation result of SP focal spot. (b) Plots of FWHM and DOF by line-scanning the longitudinal and transverse of intensity distribution through the focus point.

Figure 4.11 (a) shows the near-field SP distribution at the right side of PMFZP. The SP focal point is clearly observed at 5 μm away from the PMFZP. Cross section

profiles of the focal point in Fig. 4.11 (b) display that the FWHM and DOF are 340 ± 5 nm and 608 ± 5 nm, respectively.

Compared to simulation results in Fig. 4. 8, a slightly enlarged FWHM and DOF of SP focal spot are observed. The reasons for these small discrepancies are probably due to the shape error and displacement of Fresnel zones in EBL fabrication. These errors can be reduced greatly with optimized fabrication process in the future. In experimental results, the interference fringes with the pitch of λ_{SP} are observed around the focal spot. They can be attributed to interference between the SPs focused by PMFZP and the light transmitted directly through the metal film [112].

4.3.4 Surface plasmon virtual probe

For optical fluorescence microscopy, a smaller excitation spot size allows the achievement of better resolution, such that smaller features and analytes can be observed. Therefore, tight focus of SPs is attracting much interest as the virtual probe in applications of enhancing resolution microscopy. Recently, the progress in nanotechnology and the theory development of plasmonics have led to rapidly growing interest in the implementation of plasmonic nanostructures for light super-focusing in application of super-resolution imaging. The imaging properties of plasmonic Fresnel zone plate launching lenses, including the resolution power, object-image relationships and aberrations are investigated by using a simulation method based on Huygens-Fresnel principle with SP point source model [113]. A 1D plasmonic zone plate lens consisting of nano-slits within a metal film introduces a phase delay distribution across the planar device surface by the modulation of the slit widths and positions to achieve light super-focusing [114]. Moreover, a SP virtual probe excited by radial polarization light in the femtosecond domain was demonstrated to image the vascular transport structure in the transverse section of leaf

petiole sample [115], and a single molecule fluorescence image was obtained with SP virtual probe [116].

In this chapter, by using our proposed modified effective-refractive-index model, it is feasible to design dielectric structures to optimize the SP phase modulation effect. The simulation and experimental results have demonstrated the optimized PMFZP for totally constructive interference of SPs in focusing as a subdiffraction virtual probe.

In practice, the scanning scheme of SP virtual probe is required in the applications of in-plane scanning fluorescence microscopy. Some references have demonstrated that the phase of SPs excited by subwavelength-sized structures can be modulated by tailoring the phase of illumination light [117, 118]. Therefore, it is possible to realize scanning of SP virtual probe by using structured illumination light, such as optical vortex beam, to introduce initial phase of SPs interaction with PMFZP. The position of SP virtual probe is also possible to be controlled by the intensity profile of the external light when dielectric zone plate is made of nonlinear materials whose permittivity depends on the intensity of the light [119]. These phase-controlled strategies can be used in in-plane microscopy applications, for example, the virtual probe combining microfluidic chip technology allow us to on-chip real-time high-resolution bio-imaging away with bulky optical elements [120]. Furthermore, advanced nanoimprinting technique offers opportunity to cost-effectively fabricate such on-chip plasmonic imaging devices.

In reference [47], N. I. Zheludev group demonstrated an optical mask that creates constructive interference of wave known as super-oscillation, leading to a subwavelength focus of prescribed size and shape in a field of view beyond the evanescent fields, when illuminated by a monochromatic wave. In the mask design,

the desired subwavelength hot-spot is presented as a series of prolate spheroidal wave functions, which is transferred into a series of plane waves using the scalar angular spectrum description of light propagating from the mask to the super-oscillating feature. Then the required complex mask transmission function can be derived. In our proposed PMFZP, phase modulation of SPs with dielectric structures has demonstrated higher focusing efficiency than amplitude modulation with metallic structures. Therefore, once we adopt the design of phase mask instead of the one of diffraction mask to generate the required series of plane waves for superoscillation, it is possible to get a complex phase mask to generate a subwavelength concentration of SPs far away from phase mask to act as a virtual probe.

4.4 Chapter summary

In summary, the subwavelength-sized metal grating is explored as the SP coupler to shrink the size of plasmonic device. The relief subwavelength-sized dielectric structures are deposited at the side of grating coupler for phase modulation of SP. A modified effective-reflective-index model is proposed for the optimization of relief dielectric structures in SP phase modulation. As a suitable elementary structure, the in-plane phase modulation Fresnel zone plate is investigated with this model for totally constructive interference of SPs to focus. This subdiffraction limited size SP focal point may act as a virtual probe to help improve the resolution of SP based in-plane microscopy. The proposed plasmonic device with integration of SP generation and phase modulation in a compact domain is a great interest to researchers of on-chip plasmonic microscopy.

The simulation and experimental results of phase modulation of SPs by using subwavelength-sized dielectric structures have been published in *Opt. Express* 16, 19271 (2008), and *Opt. Lett.* 35, 4196 (2010), respectively.

CHAPTER 5

SURFACE PLASMON STANDING WAVE GENERATED BY USING SUBWAVELENGTH- SIZED METALLIC STRUCTURES

5.1 Chapter introduction

In order to image fluorescence objects of the subdiffraction feature size, the illumination fields must be modulated to contain high spatial frequency. Effective phase modulation of SPs for totally constructive interference into a single subwavelength-sized spot by using of relief dielectric structures has been demonstrated in the previous chapter. Except for phase modulation, SPs can also be manipulated by the defection of metal surface [71, 72, 76, 77]. Recently, isolated subwavelength energy hot spots are created and positioned by coherent control of nanoscale light localization in metamaterial [121]. Active control of plasmonic fields focusing at a freely prechosen point on the surface of a nanohole array is also realized by using a digital spatial light modulator to optimize the plasmonic phases through feedback [122]. The digital addressing and scanning of subdiffraction limited SP hot spot without mechanical motion may enable novel interdisciplinary applications of super-resolution imaging. However, like all other scanning microscopes, they all

suffers from the time-consuming point-by-point scanning schemes, which is hard to meet the requirement for real-time imaging. Therefore, a wider-field optical imaging system with resolution beyond the diffraction limit will stir considerable interests especially in the biological research. In this chapter, we propose a versatile method for manipulation of SP to interfere to 1D or 2D SP standing wave patterns with the design of subwavelength-sized metallic structures. Essentially, these subdiffraction SP standing wave features can be utilized as a wide-field fluorescence excitation profile since they contain super-diffraction-limited spatial frequencies.

The pioneering studies of SPs have investigated the generation of 1D SP standing waves by using two Gaussian beams fixed at the angle of SP excitation incident into TIRF objective lens [48]. A perceived disadvantage of ATR coupling configuration is the limited incident angle of objective lens which restricts this approach only in low frequency domains.

Some alternative studies suggested that SP standing waves at the surface of a thick metal film is formed with subwavelength surface corrugations such as a hole array [123] and slit apertures [124]. The SP waves excited at each individual aperture then propagate and interfere to form standing waves in the center area. These SP standing wave approaches offer unique advantages compared to the excitation of reflective mode SP standing wave by ATR coupler: (1) Surface structures can be designed to match SP momentum in a wide range of frequency without limitation of incident angle, (2) the transmission SP mode operation at normal incidence simplifies beam alignment, (3) subwavelength surface defect coupler is small relative to the ATR coupler, enabling eventual on-chip miniaturization and integration of plasmonics optics.

Motivated by the above advantages, in this chapter subwavelength slit arrays are explored to excite SPs on the metal surface. At the same time, the edges of slit can be thought as a line of SP point source. By patterning SP couplers with such sharp edges at the metal film, high-contrast standing wave patterns are designed by interference of SPs in the center area of slit structure. Furthermore, the subdiffraction limited SP standing wave features can also be dynamically tailored by adjusting the polarization and phase of the excitation light beam as the excitation profiles of fluorescence imaging. The profile gives a two-dimensional lateral resolution enhancement in wide-field high-resolution fluorescence imaging which will be discussed in Chapter 6.

5.2 Interference of SPs by using subwavelength-sized metallic structures

5.2.1 Theoretical modeling

The SP propagation length along the metal/dielectric interface indicates the maximum interference areas with sufficient contrasts. Due to the fact that the internal loss of SPs on the Ag surface is known to be lowest in optical frequency compared to other noble metals, Ag film based plasmonic structures are utilized to generate large-area SP interference patterns in this section.

Fig. 5. 1 The design of SP standing wave generator. (a) Cross section and (b) top view of four perpendicular one-dimensional slit array structures designed to convert free-space light into SP waves propagating along the metal/dielectric interface.

As depicted in Fig. 5.1, four perpendicular one-dimensional slit arrays are designed to be embedded in the Ag film to convert free-space light into SP waves propagating along the metal surface. The pitch of the slit array structure is 610 nm in order to match the SP momentum under normally incident illumination with the wavelength of 633 nm. Separation between two parallel slit arrays is set as 6 μm , which is much smaller than the propagation length (21 μm) of SPs for Ag with dielectric constant of $-15.87 + 1.07i$ in order to sustain the uniformity intensity of SP interference fringes.

Since the SP waves can only be excited by p-polarized incident light, with the x -polarized light normal incident on the slit array structure, SPs modes, corresponding to $+k_{SP}$ and $-k_{SP}$, are excited by left and right vertical slit array couplers and counter propagate towards the center area of structure. Consequently, the SP field intensity (I_{SP-x}) can be written as

$$I_{SP-x} = \langle [E_1 \cos(k_{sp}x - wt) + E_2 \cos(k_{sp}(d - x) - wt)]^2 \rangle \quad (5.1)$$

where d is the separation distance of slit arrays, which is much shorter than propagation length of the SP waves in order to get uniform SPs fringes in the interference area. The E_1 and E_2 are amplitude of SP fields propagating from left and right slit arrays, respectively. Since the laser beam is normally incident on the sample, the SP fields excited by slit arrays are equal in phase and amplitude. From Eq. 5.1, the SPs' interference field intensity distribution (I_{SP-x}) in the center area of slit array structure is described as

$$I_{SP-x} = I_{SP0} \left[1 + \cos \left(2k_{sp} \left(x - \frac{d}{2} \right) \right) \right] \quad (5.2)$$

where $I_{SP0} = E_1^2 = E_2^2$ is the amplitude of SP standing wave pattern. For simplicity, we ignore the SP propagating loss in Eq. (5.2). In the center area of slit arrays, 1D standing wave dominates the SP intensity distribution as a result of the interference between two counter-propagating SP waves.

By modulating the polarization of incident beam, vertical or horizontal directional SP standing wave interference patterns can be generated by x -polarized or y -polarized illumination, respectively.

Similarly, diagonal polarized illumination leads to interference of four perpendicular propagating SP waves which are excited by four slit array couplers. The SP field distribution (I_{SP}) in the center area of slit array structure can be described as

$$\begin{aligned} I_{SP} &= \left\langle \left[E_1 \cos(k_{sp}x - wt) + E_2 \cos(k_{sp}(d-x) - wt) + E_3 \cos(k_{sp}y - wt) + E_4 \cos(k_{sp}(d-y) - wt) \right]^2 \right\rangle \\ &= I_{SP0} \left\{ 2 + \cos\left(2k_{sp}\left(x - \frac{d}{2}\right)\right) + \cos\left(2k_{sp}\left(y - \frac{d}{2}\right)\right) + 4\cos\left(k_{sp}\left(x - \frac{d}{2}\right)\right)\cos\left(k_{sp}\left(y - \frac{d}{2}\right)\right) \right\} \end{aligned} \quad (5.3)$$

As a result, two-dimensional SP standing wave s assembled in the spot array pattern is obtained due to the interference of four SP waves coming from four perpendicular slit array couplers.

Due to their intrinsic standing wave nature, 1D or 2D SP standing wave interference patterns carry high spatial frequency, $K_{SP}=4\pi/\lambda_{SP}$, where K_{SP} is the wavevector of SP standing wave and λ_{SP} is the wavelength of SPs. As such, the SP standing waves offer a unique opportunity as the excitation profiles to substantially include high frequency in images, resulting in higher resolution.

5.2.2 Simulation results and discussions

Digital simulation is carried out using a method based on the finite difference time-domain (FDTD) (Remcom XFDTD). The Drude dispersion relation is used to fit silver material complex permittivity in optical frequency range with the parameters of $\epsilon_{\infty}=3.98$, $\epsilon_s=-1.7478 \times 10^4$, $t_0=1 \times 10^{-14}$ and $\sigma=1.5488 \times 10^7$ S/m.

Fig. 5.2 Simulation results of SP standing waves. (a1)-(c1) The SP electric field intensity distributions on a plane located at $z=20\text{nm}$ above metal film when the polarization of the incident light is along x (a1), y (b1) and diagonal direction (c1), respectively. (a2)-(c2) Cross sections of electrical field intensity distribution for two periods of standing intensity corresponding to (a1)-(c1).

The electromagnetic intensity distributions on the plane of 20 nm above the structure surface are shown in Fig. 5.2. Since the SP waves can only be excited by p-polarized incident light, with x -polarization illumination, the counter-propagating SP waves are excited by left and right slit arrays, which interfere each other to form 1D SP standing wave pattern along vertical direction, as shown in Fig. 5.2 (a). The period of interference pattern at the center area is equal to 305 nm. The FWHM of fringes is equal to 152 nm, corresponding to $0.24 \lambda_0$. Similarly, y -polarized light is used to generate 1D standing SP pattern along horizontal direction with the same FWHM of interference fringes as the vertical one, as shown in Fig. 5.2 (b). The average contrast of 1D SP standing wave fringes reaches up to 0.82. When the linearly polarized light is incident on the sample with the polarization along diagonal direction (indicated by white arrow), 50% of the incident light for x -direction slit arrays is p-polarized, and another 50% of incident light is p-polarized for y -direction slit arrays. Therefore, a dot array shaped 2D SP standing wave pattern is generated in the central area of slit array structure due to the overlapping of both amplitude and phase of four perpendicular propagating SP waves, as shown in Fig. 5.2 (c). The period of dot arrays is 431 nm along diagonal direction of four slit arrays with the contrast of 0.9. The FWHM is 215 nm, corresponding to $0.34 \lambda_0$.

One noteworthy point to highlight is that the intensity of 1D SP interference fields excited by either x -polarized light or y -polarized light is approximately half of the 2D SP standing wave interference fields by using diagonal direction polarized light illumination. It is because the 2D standing wave pattern is constructed by the SP wave launched from four slit array couplers, while the 1D standing wave patterns are built from two slit array couplers.

5.3 Demonstration of subdiffraction limited SP standing waves

5.3.1 Fabrication of subwavelength-sized metallic structures

The Ag film with slit array structure is fabricated by a standard electron beam lithography process. PMMA (A4, MicroChem) is spin-coated on an ITO glass substrate with a film thickness of 360 nm and expose to $180 \mu\text{C}/\text{cm}^2$ in EBL system. After developing, 100 nm thickness of Ag film is thermally evaporated on the sample. Finally, the Ag film is immersed into acetone solvent for lift-off to remove the Ag on non-exposed PMMA areas. A scanning electron microscopy (SEM) image of the subwavelength-sized slit array structure in Ag film is shown in Fig. 5.3. The pitch of slit array and slit width is 610 nm, and 265 nm, respectively.

Fig. 5.3 Realization of the surface plasmon standing wave generator. SEM image of two-dimensional slit arrays embedded in Ag film.

5.3.2 Experimental results and discussions

The distribution of the SP fields on the metal surface is characterized by a NSOM system (NT-MDT NTEGRA Solaris) with an aluminum coated fiber tip of 100 nm aperture size. As described in section 3.3.2, in our experimental setup, a linearly polarized He-Ne laser with the wavelength of 633 nm is guided into an inverted microscopy and normally incident on the substrate by an objective lens with NA equaling to 0.13. The polarization of the incident light is adjusted by a half-wave plate to modulate the SP field distribution in the center area of slit arrays.

Fig. 5. 4 NSOM images of SP standing waves. (a1)-(c1) The SP field intensity distributions of slit array structure for polarization of illumination light along x (a1), y (b1) and diagonal direction (c1), respectively. (a2)-(c2) Cross section of electrical field intensity distribution in the center area of slit array structure corresponding to (a1)-(c1).

Figure 5.4 shows the near-field distributions of SP interference patterns by using NSOM. 1D SP standing wave patterns in either the x or y direction, due to linearly polarization illumination on the sample in the x or y direction, are shown in Fig. 5.4 (a) and 5.4 (b), respectively. The interference period and FWHM is measured to be 307 ± 5 nm and 150 ± 5 nm, respectively. The average contrast of the 1D SP standing wave fringes reaches to 0.8. When the polarization of illumination light is altered along diagonal direction of four slit arrays, all the slits can be treated as SP sources and equally excite SP waves propagating toward the center of slit array structure. The four perpendicular counter-propagating SP waves along $\pm x$ and $\pm y$ directions interfere with each other in the center area of structure to assemble a 2D standing SP pattern in dot array shape, as shown in Fig. 5.4 (c). The array period approximately equals to 438 ± 5 nm along diagonal direction, with the FWHM of 215 ± 5 nm. The contrast of 2D SP standing wave pattern is higher than that of 1D ones by around 12%. This is mainly due to the higher intensity of SP interference field excited by a larger number of slits for diagonal polarization illumination than those of vertical or horizontal polarization illumination. These results well agree with the simulation result by using the FDTD calculation as shown in Fig. 5.2.

In our experiments, the grating pitch of SP coupler is designed to excite SPs at 633 nm. A He-Ne laser is utilized as the light source. The gain bandwidth of the He-Ne laser is dominated by Doppler broadening rather than pressure broadening due to the low gas pressure, and is thus quite narrow: only about 1.5 GHz full width for the 633 nm transition (a wavelength range of about 0.002 nm at a central wavelength of 633nm) [125]. When the SPs are excited by He-Ne laser, they have the same bandwidth as illumination source. Therefore, we can expect that there is less than 1%

contrast of SP interference patterns affected by the bandwidth of the He-Ne laser light, which can be neglected.

As mentioned in section 2.2.3, the field amplitude of the SPs decreases exponentially as $\exp(-|k_{i,z}||z|)$, normal to the surface. In our experiment, it is characterized by using NSOM scanning at several different constant heights above the structure. The data is fit by exponential decay curve, as shown in Fig. 5.5. The penetration depth in the air medium at which the SP field falls to $1/e$, is 245 ± 5 nm for silver with 633 nm excitation wavelength. This value defines the fluorescence excitation area using SP interference patterns as the excitation profiles.

Fig. 5. 5 SP penetration depth in the air medium.

The subwavelength-sized standing-wave SP pattern stems from the fact that the excitation light grid has a period of $\lambda_0/2n_{SP}$, where n_{SP} is the effective-refractive-index of the metal/dielectric interface where SP existed. Fundamentally, the FWHM of SP standing wave fringe is determined by the excitation wavelength and the available high effective-refractive-index of the metal/dielectric interface. Typically

n_{SP} equals to 1.05 for an Ag/air interface and 1.7 for an Ag/oil interface with 532 nm incident light. Therefore, it is possible to significantly reduce the pitch of 1D SP standing wave fringes to 80 nm by optimizing the design of slit arrays to meet SP dispersion relation at Ag/oil interface with green laser excitation. These standing waves carrying high frequency can be used as the excitation profiles to realize wide-field super-resolution fluorescence imaging.

Compared to the standing wave pattern formed by free laser beam interference, an obvious advantage of SP standing wave interference is the higher resolution, due to n_{SP} , as our discussion in previous part. In addition, in order to achieve 1D or 2D standing wave interference patterns with conventional optical system, multiple laser beams and complicated controlling optics are need to precise direct the light. In our case, the SP interference is simply realized by normally illuminating a subwavelength-sized metallic structure with the controlled polarization of excitation light. Moreover, the subwavelength-sized SP interference device is flat in nature, thus can be integrated in a chipset system for applications in wide-field on-chip imaging and sensing.

5.4 Chapter summary

In this chapter, we have demonstrated that subwavelength-sized slit array structure on metal film can excite SPs propagating along the metal/dielectric interface. The counter-propagating SP waves excited by slit array couplers interfere each other at the center area of structures. By varying the polarization of illumination light, high contrast 1D or 2D standing wave patterns are generated. The SP standing waves, carrying high spatial frequency information, confined to several hundreds nanometer

in the dielectric medium, and strongly enhanced by constructive interference, are our interests to act as excitation profiles for investigation of a wide-field high-resolution fluorescence microscopy. Moreover, the advantage of subwavelength-sized plasmonic interferometer is that the structure can simply be written on a chip in nanometer scale, thus integration of such an element in microscopy system is possible to realize lab-on-a-chip imaging. In the following chapter, we will utilize these properties in novel microscopy techniques and describe the engineering approaches in building the imaging system.

My contribution on the generating SP standing wave has been published in Opt. Express 18, 2662 (2010).

CHAPTER 6

WIDE-FIELD **HIGH-RESOLUTION** **FLUORESCENCE IMAGING WITH** **SURFACE PLASMON STANDING WAVE** **EXCITATION**

6.1 Chapter introduction

Biological diagnosis and medicine research always require dynamic imaging of biological interaction processes with high-resolution. Therefore, far-field optical fluorescence microscopy remains as the most widely used tool due to the fluorescence staining of interested bio-sample's components. Unfortunately, like all forms of optical tools, it suffers from the fundamental limitation by the light diffraction. During the last decades, there have been extensive researches on super-resolution optical microscopy ranging from of linear methods to nonlinear methods. However, the complex imaging system or scanning nature limit their applications in low-cost on-chip biological sample detection.

SPs, arising from their strong localization and nonradiative nature, have been shown to be a versatile approach for applications in high-resolution fluorescence imaging. Previous section 1.1.2 has reviewed some SP patterned illumination microscopes [49, 53], which benefit from planarity design and high-resolution with great promise for on-chip biological imaging. However, they are presently only realized in numerical experiments due to high quality requirements of substrate preparation or illumination control. Therefore, we propose an alternative simple approach to realize high-resolution wide-field microscopy by using subwavelength-sized plasmonic structures for generation and modulation of SP standing wave patterns as the excitation profiles. Compared to previously discussed SP patterned illumination microscopes [49, 53], this approach has several desirable advantages. Firstly, subwavelength-sized slit arrays can be designed to match SP momentum in high frequency without limitation of incident angle, which will result in high resolution in fluorescence imaging. Secondly, two directional SP standing waves are generated by changing polarization direction of illumination beams, serving as the fluorescence excitation profiles. No sample rotation or stage rotation is needed for lateral resolution enhancement in two dimensions. Thirdly, with the normal incident of beam illumination requirement and the flat design of plasmonic structures, our approach offers a high level of miniaturization for potential applications in integrated lab-on-chip super-resolution microscopy.

In this chapter, surface plasmon standing wave fluorescence microscopy (SP-SWFM) is demonstrated. With this approach, the subwavelength slit arrays are embedded in a thin silver film to generate SP standing wave interference patterns. These fringes carrying high spatial frequency information serve as excitation profiles to excite the nanoscale fluorescence objects. The super-resolved fluorescence density

distribution is reconstructed from a weighted sum of a series of fluorescence images with differently phase-shifted SP standing wave illumination. The flat configuration, wide-field and non-invasive nature makes this approach suitable for real time analyzing the fine details of bio-samples in many bio-chip applications.

6.2 Image reconstruction of surface plasmon standing wave fluorescence microscopy

6.2.1 Description of the illumination configuration

The theoretical basis for resolution enhancement of surface plasmon standing wave fluorescence microscopy is based on the formation of an excitation field containing super-diffraction limited spatial frequency components. Once this excitation field is translatable experimentally, the high frequency information of objects can be extracted from a set of diffraction limited images where the excitation fields contain different displacement vectors [126].

Chapter 5 has shown the study on the excitation of SPs by subwavelength wide slit arrays embedded in Ag film. The two counter-propagating SP waves interfere to form 1D or 2D SP standing wave patterns. These SP fringes carrying high spatial frequency information can be exploited to realize super-resolution SP standing wave fluorescence imaging. In order to retract the high frequency information of objects from fluorescence images, a series of SP standing wave patterns with different phase-shift are required to act as the excitation profiles. Some previous researches have demonstrated that the phase of SP field can be dynamically tailored by adjusting the excitation light beam [118, 124, 127]. In our proposed slit array

structure, with normal incidence of Gaussian beam, the SP waves from slit arrays carry the equal initial phases. However, by manipulating the phase profile of the incident beam, it is possible to shift the SP standing wave interference pattern.

Assuming there is an initial phase difference (φ) between the two counter-propagating SPs, which is introduced by manipulation of incident beam. The SP interference pattern intensity (I_{SP-x}) distribution can be calculated by evaluating

$$\begin{aligned} I_{SP-x} &= \left\langle \left[E_1 \cos(k_{sp}x - \omega t) + E_2 \cos[k_{sp}(d - x) - \omega t - \varphi] \right]^2 \right\rangle \\ &= I_{SP0} \left[1 + \cos \left(2k_{sp} \left(x - \frac{d}{2} \right) - \varphi \right) \right] \end{aligned} \quad (6.1)$$

where d is the distance between two counter-propagating SPs. That implies that one can control the pattern shift of SP standing wave by introducing phase difference (φ) between the two counter-propagating SP waves.

In experiment, we design binary phase plate to modulate the initial phase of SPs excited by slit array couplers. The binary phase plate is made of rectangular-shaped photo resist SU-8 (MicroChem 2005) with uniform thickness. It is inserted into optical path of illumination light corresponding to one of slit arrays in order to shift the initial phase of the SP fields excited by the same slit array coupler. The phase difference ($\Delta\varphi$) introduced by binary phase plate can be got from

$$\Delta\varphi = \frac{2\pi(n_{SU8} - 1)d}{\lambda_0} \quad (6.2)$$

where $n_{SU8} = 1.59$ is the dielectric constant of SU-8, d is the thickness of binary phase plate, and λ_0 is the wavelength of incident beam. Therefore, the thickness of 350nm can bring with $2\pi/3$ phase difference with the incident beam of 632.8 nm wavelength.

The binary phase plate can easily be fabricated using an UV lithographic method. SU-8 photoresist is firstly 1:4 diluted by SU-8 thinner. Then it is spin-coated on a glass substrate at a speed of 2050 rpm to get uniform thickness of 350 nm,

undergoes pre-bake at 65°C for 90 sec, left to cool for 2 mins, subjected to UV exposure (MJB3 Karl Suss Mask Aligner) under a patterned mask with 14 mW/cm² dose for 20 sec. After that, the film is post-baked at 65°C for 90 sec for cross-linking of the exposed regions. The film is then developed in SU-8 developer solvent for 60 sec to yield a rectangular-shaped (10 mm × 10 mm) binary phase plate.

Fig. 6. 1 NSOM image of phase-shifted SP standing wave pattern

Figure 6.1 shows the NSOM image of the SP standing wave pattern shift by adding binary phase plate in the illumination optical path. The SP standing waves clearly show a lateral shift of 1/3 pitch fringe, corresponding to $2\pi/3$ phase shift.

6.2.2 Description of the image reconstruction algorithm

We begin our discussions with imaging reconstruction algorithm in one dimension. With subwavelength-sized slit array coupling geometry, a SP standing wave interference pattern can be generated laterally across a sample. The SP field then excites the fluorescent stains to form an image which is collected by traditional microscope. Because the excitation profile is spatially modulated by the high frequency SP standing waves, the image function (I_x) can be expressed as

$$I_x = \{o \cdot [1 + \cos(K_{sp}x + \phi)]\} \otimes p \quad (6.3)$$

where o is object function, $K_{SP}=4\pi/\lambda_{SP}$, ϕ is an unknown phase that specifies the position of the SP standing waves relative to the object, and the p is the PSF of conventional microscope.

Previous section has shown that the SP standing wave fields are translatable, and the placement of fringes can be experimentally controlled. Therefore, the image function (I_x) can always synthesize a new image function (I'_x) as

$$I'_x = o \otimes \{(1 + \cos(K_{SP}x + \phi)) \cdot p\} \quad (6.4)$$

where $(1 + \cos(K_{SP}x + \phi)) \cdot p$ can be seen as the effective PSF of new image containing super-diffraction limited frequency components originating from SP standing waves. Moreover, the new image can be computationally reconstructed by a weighted sum of intermediate image functions (I_x) with higher resolution than the intermediate ones.

In order to extract the high spatial frequency information in one dimension, at least three intermediate fluorescence images with phase-shifted SP standing wave illumination are necessary [128]. In this analysis, three intermediate images (I_{x0} , I_{x1} , I_{x2}) with phase-shift step (ϕ) are expressed as:

$$I_{x0} = \{o \cdot [1 + \cos(K_{SP}x + \phi)]\} \otimes p \quad (6.5)$$

$$I_{x1} = \{o \cdot [1 + \cos(K_{SP}x + \phi - \phi)]\} \otimes p \quad (6.6)$$

$$I_{x2} = \{o \cdot [1 + \cos(K_{SP}x + \phi + \phi)]\} \otimes p \quad (6.7)$$

Utilizing the identity

$\{\exp[\pm i(K_{SP}x + \phi)] \cdot o\} \otimes P = \exp[\mp i(K_{SP}x + \phi)] \{o \otimes \exp[\mp i(K_{SP}x + \phi)] \cdot P\}$, the image functions can be written as

$$\begin{aligned}
I_{x0} &= o \otimes p + \frac{1}{2} \exp[i(K_{sp}x + \phi)] \{o \otimes \exp(-iK_{sp}x) \cdot p\} \\
&+ \frac{1}{2} \exp[-i(K_{sp}x + \phi)] \{o \otimes \exp(iK_{sp}x) \cdot p\}
\end{aligned} \tag{6.8}$$

$$\begin{aligned}
I_{x1} &= o \otimes p + \frac{1}{2} \exp[i(K_{sp}x + \phi - \varphi)] \{o \otimes \exp(-iK_{sp}x) \cdot p\} \\
&+ \frac{1}{2} \exp[-i(K_{sp}x + \phi - \varphi)] \{o \otimes \exp(iK_{sp}x) \cdot p\}
\end{aligned} \tag{6.9}$$

$$\begin{aligned}
I_{x2} &= o \otimes p + \frac{1}{2} \exp[i(K_{sp}x + \phi + \varphi)] \{o \otimes \exp(-iK_{sp}x) \cdot p\} \\
&+ \frac{1}{2} \exp[-i(K_{sp}x + \phi + \varphi)] \{o \otimes \exp(iK_{sp}x) \cdot p\}
\end{aligned} \tag{6.10}$$

Substituting Eq. (6.8), (6.9) and (6.10) into Eq. (6.4), the composited image can be rewritten as

$$I'_x = f_0 I_{x0} + f_1 I_{x1} + f_2 I_{x2} \tag{6.11}$$

where f_0, f_1, f_2 are weighting function as

$$\begin{aligned}
f_0 &= \frac{\cos \varphi}{\cos \varphi - 1} + \frac{1}{(1 - \cos \varphi)} \cos(K_{sp}x - \phi) \\
f_1 &= \frac{1}{2(1 - \cos \varphi)} + \frac{\sqrt{2(1 - \cos \varphi)}}{2 \sin \theta |(\cos \varphi - 1)|} \cos(K_{sp}x - \phi - \delta) \\
f_2 &= \frac{1}{2(1 - \cos \varphi)} + \frac{\sqrt{2(1 - \cos \varphi)}}{2 \sin \theta |(\cos \varphi - 1)|} \cos(K_{sp}x - \phi + \delta) \\
\tan \delta &= -\frac{(\cos \varphi - 1)}{\sin \varphi}
\end{aligned} \tag{6.12}$$

The set of weighting function completely determines the algorithm for reconstructing a high-resolution composite image. From the periodicity of SP standing wave fringes, $\varphi=2\pi/3$ is chosen to get the equal span phase-shift vectors for the highest signal-to-noise ratio [129]. Therefore, the weighting factor is obtained as

$$\begin{aligned}f_0 &= \frac{1}{3} + \frac{2}{3} \cos(K_{sp}x - \phi); \\f_1 &= \frac{1}{3} + \frac{2}{3} \cos(K_{sp}x - \phi - \frac{2}{3}\pi) \\f_2 &= \frac{1}{3} + \frac{2}{3} \cos(K_{sp}x - \phi + \frac{2}{3}\pi)\end{aligned}\tag{6.13}$$

1D reconstructed high-resolution image is given by

$$\begin{aligned}I_x' &= \frac{1}{3} [1 + 2 \cos(K_{sp}x - \phi + \alpha)] I_{x0} \\&+ \frac{1}{3} \left[1 + 2 \cos\left(K_{sp}x - \phi + \alpha - \frac{2}{3}\pi\right) \right] I_{x1} \\&+ \frac{1}{3} \left[1 + 2 \cos\left(K_{sp}x - \phi + \alpha + \frac{2}{3}\pi\right) \right] I_{x2}\end{aligned}\tag{6.15}$$

where α is the arbitrary phase to be chosen for optimum resolution.

In order to realize two-dimensional enhanced SP standing wave fluorescence image, six intermediate images taken by x and y directional phase-shifted standing wave illumination are required, where the perpendicular one-dimensional SP standing wave patterns are simply generated by rotating the polarization of the incident beams. With this approach, total six intermediate fluorescence images (I_{x0} , I_{x1} , I_{x2} , I_{y0} , I_{y1} , I_{y2}), corresponding to $2\pi/3$ phase-shifted SP standing wave excitation ($\phi_x, \phi_x - 2\pi/3, \phi_x + 2\pi/3, \phi_y, \phi_y - 2\pi/3, \phi_y + 2\pi/3$), are obtained. The two-dimensional enhanced image (I_{xy}) is given by

$$\begin{aligned}I_{xy}' &= \frac{1}{6} [1 + 2 \cos(K_{sp}x - \phi_x + \alpha_x)] I_{x0} \\&+ \frac{1}{6} \left[1 + 2 \cos\left(K_{sp}x - \phi_x + \alpha_x - \frac{2}{3}\pi\right) \right] I_{x1} \\&+ \frac{1}{6} \left[1 + 2 \cos\left(K_{sp}x - \phi_x + \alpha_x + \frac{2}{3}\pi\right) \right] I_{x2} \\&+ \frac{1}{6} [1 + 2 \cos(K_{sp}y - \phi_y + \alpha_y)] I_{y0} \\&+ \frac{1}{6} \left[1 + 2 \cos\left(K_{sp}y - \phi_y + \alpha_y - \frac{2}{3}\pi\right) \right] I_{y1} \\&+ \frac{1}{6} \left[1 + 2 \cos\left(K_{sp}y - \phi_y + \alpha_y + \frac{2}{3}\pi\right) \right] I_{y2}\end{aligned}\tag{6.16}$$

where a_x and a_y are the arbitrary phase to be chosen for optimum resolution. ϕ_x and ϕ_y are unknown phases that specify the position of x and y directional 1D SP standing waves relative to the objects, respectively.

6.3 Enhancement of lateral resolution in fluorescence microscopy with surface plasmon standing wave excitation

6.3.1 Simulation results and discussions

In our imaging simulation model, we consider one layer of fluorescence beads of 20 nm in diameter randomly deposited in the central area of slit array structure. Once the intermediate fluorescence images are obtained by successive phase modulation of the SP standing waves excitation profiles in two dimensions, according to Eq. (6.15) and Eq. (6.16), 1D and 2D super-resolved fluorescence images are reconstructed by the weighted sum of a series of these phase-shifted illumination intermediate images to obtain the final super-diffraction limited image, respectively.

Similar to other high-resolution technique based on interference, there exists sidelobes artifact in the fluorescence images [130, 131]. This artifact can be easily removed by linear deconvolution during post image processing [132].

Fig. 6. 2 Simulation result of SP-SWFM. (a1)-(b1) 1D enhanced SP-SWF image along vertical direction (a1) and horizontal direction (b1). (c1) 2D enhanced SP-SWF image. (a2)-(c2) Cross sections of PSF profiles at a selected region of interest from (a1) -(c1).

The simulation results in Fig. 6.2 show that 153 nm FWHM of the PSF of one-dimensional and 176 nm for two-dimensional SP-SWF image can be achieved. The PSF for two-dimensional SP-SWF image is larger than one-dimensional one due to the symmetrical PSF with reduced sidebands from summation of two directional scanning [126]. However, the improvements of two-dimensional resolution are more useful in practical imaging applications. Compared to one-dimensional SP-SWF image in Fig. 6.2 (a) and Fig. 6.2 (b), a two-dimensional SP-SWF image in Fig. 6.2 (c) enables to resolve more particles. With this approach, the lateral resolution of SP-SWFM mostly depends on the SP standing wave fringe period rather than the wavelength of illumination light.

6.3.2 Experimental setup

Fig. 6. 3 Schematic diagram of the SP-SWFM.

The schematic setup of the proposed SP-SWFM is shown in Fig. 6.3. An inverted microscope with a high NA objective (NA=1.42) is used as the basic configuration in the experimental setup. The SP standing waves are generated by focusing the incident laser beam (633nm) at the back focal plane of the objective lens to uniformly illuminate the specimen/metal interface. The fluorescence image is collected by the same objective lens, and subsequently captured by a charge-coupled device (CCD) camera. A notch filter ($\lambda = 632.8$ nm, Laser line blocking > 6 OD, 50% Notch Bandwidth = 25 nm) is utilized to block the illumination light into CCD. A half wave plate is rotated to generate perpendicular one-dimensional SP standing wave patterns in the center area of slit array structure. The binary phase plate of rectangular shaped SU8 photo resist with uniform thickness of 350 nm is inserted into optical path corresponding to one of slit arrays to shift the initial phase of the SPs excited from that slit array coupler. With this approach, the SP standing waves shifts 1/3 pitch of fringe. Practically, the position of binary phase plate is changed in order to modulate one-dimensional SP standing wave patterns shift in two respective directions for obtaining intermediate images to reconstruct two-dimensional high-resolution fluorescence imaging.

6.3.3 Experimental results and discussions

With the permittivity of Ag film $\epsilon_m = -15.87 + 1.07i$ at 633 nm, the theoretical calculation of SP propagating length is equal to 21 μm . Therefore, the center area of the slit array structure is designed as 18 $\mu\text{m} \times 18 \mu\text{m}$, in order to get large area of SP standing wave illumination fields for wide-field imaging. The subwavelength-sized silver structure is fabricated by a standard electron beam lithography process followed by thermal evaporation and lift-off steps. NSOM (NT-MDT) is applied to characterize

the SP intensity distributions with an aluminum-coated fiber tip of 100 nm diameter aperture.

Figure 6.4 (a) and (b) shows the SP standing wave patterns with different polarization illumination indicated by the white arrows. The 1D SP standing waves along two perpendicular directions offer an unique opportunity to realize two-dimensional resolution improvement. The FWHM of SP standing wave fringe equals to 155 ± 5 nm. The intensity contrast of the SP fringes is larger than 0.68 in the middle of the slit array structure, which is sufficiently high for fluorescence imaging purpose. Due to defect of the half wave plate, less than 5% illumination light of vertical polarization illuminates on the structure to generate SPs propagating along the vertical direction, which modulates 1D SP standing wave fringes in dotted shaped, as shown in Fig 6.2 (a1).

Fig. 6. 4 NSOM images of SP standing wave interference patterns in the center area of slit array whose size is designed as $18 \mu\text{m} \times 18 \mu\text{m}$. The white arrows indicate the polarization along vertical (a1) and horizontal direction (b1). (a2) and (b2) are cross sections of electrical field intensity distributions corresponding to (a1) and (b1).

Fluorescence polystyrene microspheres (diameter 20 nm, peak emission at 645 nm, Molecular Probes) are deposited on the Ag film surface as the subdiffraction objects. When the fluorescence beads near the metal surface are excited by SP standing waves, the fluorescence emissions couple with the SPs on the metal surface and enter the transparent substrate at a sharply defined angle, which is the same to the SPs angle for the emission wavelength. It was estimated that 40%-70% of the total emission enters the substrate at the plasmon angle and can thus be directed towards a detector [133]. Due to nearly isotropic emission of fluorescence bead, a uniform and

isotropic cone of emission is observed, which results in doughnut-shaped PSF in SP fluorescence image. Background emission from fluorophores far away from the silver surface will not be detected because they are not coupling with the SP. This unique surface plasmon-coupled emission (SPCE) phenomena in SP fluorescence imaging enjoys several advantages such as reduction of detective volume due to the fluorophore quenching on the metal surface, and better suppression of background noise [48].

The reconstruction procedure of extended-resolution images with SP-SWF in two dimensions is shown in Fig 6.5. Owing to the unique doughnut-shape PSF of SP-SWF, as shown in Fig. 6.4 (a), there exists a central dip in the PSF, and overall FWHM becomes widened [48]. A nonlinear deconvolution algorithm named by Richardson-Lucy with SPCE PSF kernel is applied to convert the original doughnut-shaped PSF into single-peaked PSF, as shown in Fig. 6.5 (b), subsequently followed by the application of the SP-SWFM algorithm to get high-resolution reconstructed images. According to Eq. 6.15 and Eq. 6.16, three or six fluorescence images with phase shifting of $\phi_x, \phi_x - 2\pi/3, \phi_x + 2\pi/3, \phi_y, \phi_y - 2\pi/3$ and $\phi_y + 2\pi/3$ are weighted sum to generate 1D or 2D resolution enhanced image, respectively. Figure 6.5 (c)-(e) displays the PSF of 1D and 2D enhanced resolution images, respectively. Finally, the side band of 2D PSF is reduced by using conv2 filter mode in Matlab program, as shown in Fig. 6.5 (f).

Fig. 6. 5 The reconstruction procedure of extended-resolution images with SP-SWF in two dimensions (a1) The doughnut-shape SPCE rings in the intermediate image. (b1) The single-peaked spot converted from doughnut-shape SPCE rings by convolution. (c1) The PSF of 1D enhanced resolution SP-SWF image along x direction. (d1) The PSF of 1D enhanced resolution SP-SWFM image along y direction. (e1) The PSF of 2D enhanced resolution SP-SWF image along diagonal direction. (f1) The PSF of 2D enhanced resolution SP-SWF image with linear deconvolution to remove the side band. (a2)-(f2) Cross sections of PSF profiles corresponding to (a1)-(f1).

According to Eq. (6.16), 2D enhanced resolution fluorescence image is obtained by superimposing six intermediate images with phase-shifted SP standing wave illumination.

Fig. 6. 6 The experimental result of SP-SWFM compared with high NA fluorescence microscopy. (a1) Experimental results of standard high NA fluorescence images. (b1) Experimental results of the reconstructed two-dimensional enhanced resolution SP-SWF image. (a2) and (b2) Cross sections of PSF profiles at a selected region of interest from (a1) and (b1).

From experimental result shown in Fig. 6.6 (b2), the FWHM of PSF in 2D enhanced resolution SP-SWF image is approximately 172 ± 5 nm ($0.28\lambda_{SP}$) which is in good agreement with the simulation result in Fig. 6.2 (c). Compared with the standard

high NA (1.42) fluorescence images in Fig. 6.6 (a), the resolution of SP-SWF images achieves a twofold improvement and lower background noise due to the high contrast of SP standing waves excitation profiles and unique SPCE phenomenon in SP involved fluorescence imaging.

In our proposed SP based high-resolution fluorescence microscope, the fluorescence emission strongly depends on the distance separating fluorophore and metal. For large separations, fluorescence is not affected, while for very small separations, the fluorescence tends to quench at the metallic surface [134, 135]. Therefore, the excitation of fluorophores by SPs occurs for intermediate metal-fluorophores separations. With it, the range of applicability of SP based fluorescence imaging can be recognized. This technique will find applications primarily in the study of surface phenomena, such as in cell membranes. Moreover, no mechanical contact with the specimen is required because the SP itself acts as the probing mechanism. This would permit a high fidelity in soft biological samples' images.

One major advantage of the proposed SP-SWFM lies in the fact that there is no requirement to build a complex optical-controlled system to generate standing wave interference patterns as the fluorescence excitation profiles. The exploration of SP standing wave fluorescence imaging is directly realized by only replacing the cover glass in the conventional microscopy with the metal grating structured cover slip. Recently, the advances in nano-imprinting suggest that a quick and parallel production of nanostructures is feasible to replace the time-consuming EBL fabrication process [136]. With this advance, batch fabrication of metal grating structured film is possible with low cost.

Since the image acquired through a microscope is given by convoluting the excitation pattern with the PSF of the microscope, in our case, resolution of the

reconstructed fluorescence image is determined by the effective PSF which equals to the convolution of standing wave and PSF of microscope. Therefore, the resolution of reconstructed image is fundamentally determined by the FWHM of SP standing wave fringes. Further reducing the fringe pitch is feasible by changing the SP dielectric condition such as immersing metal grating in high refractive index oil to decrease the wavelength of SP wave. Since the k_{SP} may be extremely large up to $100 \times k_0$, the theoretical resolution of fluorescence image with such SP standing wave excitation profiles may reach up to a scale of a few nanometers. However, in reality, the Ohmic losses limit the propagation length of the short-wavelength SPs, and thus severely limit the attainable resolution.

When a nonlinear dependence of the fluorescence emission rate on the illumination intensity is considered, the fluorescence microscope is in principle capable of unlimited resolution [28, 137]. The classical resolution limit specifies a maximum spatial frequency $k_0 = 2\text{NA}/\lambda_{\text{em}}$ that can be observed through the microscope, where λ_{em} is the observation wavelength. SP-SWFM extends the resolution beyond the cutoff by moving high spatial frequency information into the observable region with a phase shift method. Thus, to maximize the resolution, it is desirable for the illumination to contain as high spatial frequencies as possible. If the fluorescence emission rate can be made to depend nonlinearly on the illumination intensity, such a nonlinearity would cause the effective illumination pattern to contain harmonics with spatial frequencies that are multiples of the spatial frequency contained in the illumination. Thus, a nonlinearity in theory can produce an infinite number of harmonics, which correspond to theoretically infinite resolution. In our proposed SP-SWFM, if the sample is illuminated by a sinusoidal SP standing wave pattern with a peak intensity that is near or above saturated emission threshold, then

the pattern of emission rate per fluorephore takes on a nonsinusoidal shape, which contains the desired series of harmonics with higher spatial frequency than the illumination pattern itself, thereby to increase its resolution-enhancement power. However, the resolution in practice will be finite due to signal-to-noise ratio and photostability.

6.4 Chapter summary

In this chapter, with the spatial modulation of SP standing wave patterns, we demonstrated a wide-field SP fluorescence microscopy with a resolution beyond diffraction limit. The reason that SP standing wave illumination can provide resolution extension is that it can encode high-spatial resolution information into the observed data through phase-shifted method. With this approach, six images with phase-shifted SP standing wave illumination along two perpendicular directions are superimposed to obtain wide-field fluorescence image with subdiffraction resolution. Both numerical studies and experimental results show that the resolution of two-dimensional reconstructed SP-SWF image can approach $0.28\lambda_{sp}$. It is improved nearly by a factor of two in comparison with normal high-NA optical image. The wide-field nature make this approach to have significantly faster imaging speed than the point-by-point scanning approaches. The subwavelength-sized plasmonic device benefits from grating coupler to offer a high level of miniaturization for potential applications in integrated lab-on-chip super-resolution microscopy in the future.

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CHAPTER 7

SUMMARY AND RECOMMENDATIONS

7.1 Thesis summary

The main theme of this thesis was to investigate on-chip plasmonic modulation by subwavelength-sized surface structures, for applications in subdiffraction resolution fluorescence imaging. The designed plasmonic devices are fabricated by electron beam lithography and the SP field distributions are characterized by near-field scanning optical microscopy, respectively.

To begin, we designed a subwavelength-sized slit array to convert free space light into SP waves propagating along the Au/dielectric interface. The slit array coupler configuration provides much flexibility in realization of on-chip plasmonic device. The phase modulation of SPs is studied as a function of geometric length, width and thickness of surface relief dielectric structure. The numerical simulations by using finite-domain-time-difference (FDTD) methodology indicate that the SPs are analogous to conventional free-space light waves in terms of phase modulation by dielectric geometric sizes and refractive indices. Therefore, a modified effective-refractive-index model is proposed with the consideration of SP losses to provide an intuitive method to describe how SP phase modulation is affected by dimensional sizes of dielectric rectangular structures. Specifically, one-dimensional dielectric

Fresnel zone plate is designed by employing the proposed model. The SP waves coming from all Fresnel zones are in phase synchrony and therefore totally constructive interfere to produce a focal point beyond the diffraction limit, if as a virtual probe, which may help improve the performance of SP based on-chip in-plane microscopy.

Subsequently, we demonstrated an alternative approach to modulate SPs to standing waves by using subwavelength-sized metallic structures. Four perpendicular subwavelength-sized slit arrays are designed to be embedded in the Ag film to excite and modulate counter-propagating SP waves to interfere. With the change of the incident beam's polarization, 1D or 2D SP standing wave patterns are generated in the center area of slit array structure with a high signal-to-noise ratio. The SP standing wave patterns carrying the high frequency information allow reconstruction of super-resolution images for resolving nanoscale objectives. In essence, this technique relies on the formation of a set of phase-shifted excitation fields which contain subdiffraction limited spatial-frequency components. In our case, the SP standing waves is phase-shifted by modulation of initial phase of illumination light beam, with incepting a binary phase plate into the optical path. From near-field scanning image, a uniform thickness of 350 nm SU8 binary plate is observed to introduce $2\pi/3$ phase-shift of SP fringe.

Last but not least, SP standing wave fluorescence microscopy (SP-SWFM) is demonstrated to implement wide-field fluorescence images with resolution beyond the diffraction limited. This approach theoretically brings nanoscale resolution to a wide-field microscopy by using high frequency SP standing waves illumination. In SP-SWFM, the excitation light consists of a set of phase-shifted SP standing wave patterns. The rapid spatial modulation of excitation profiles encodes high-resolution

information of objectives into the observed fluorescence images. The weighted sum of six intermediate fluorescence images illuminated by phase-shifted 1D SP standing waves, finally reconstructs a high-resolution fluorescence image. Simulation and experimental results show that the lateral resolution is enhanced to 172 nm ($0.28\lambda_{SP}$) in two dimensions, which is near a factor of two improvement in comparison to normal high-NA (1.42) optical image.

Because of a wide-field excitation, SP-SWFM can rapidly acquire data with a high signal-to-noise ratio. The integration of the excitation profiles with fluorescence imaging procedure on the metal surface provides much feasibility for instrument miniaturization. Although wide-field super-resolution SP fluorescence microscopy is in its infancy, it is still a significant and powerful technology in applications of lab-on-a-chip bio-imaging. Future research in these directions could prove to be decisive.

7.2 Recommendations

There are still much works that can be done to further optimize our results, as well as to explore potential applications in a wide range. The following are some specific challenges and improvement to be made for the future research work.

(a) *Investigation of further resolution improvements*

In optical fluorescence microscopy, a smaller PSF of illumination pattern will always produce a higher resolution image. As previously stated, the PSF size is determined by the in-plane wavelength of SP. Increasing the frequency of SP excitation light or the refractive index of dielectric within the close vicinity of the metal surface can result in the decrease of in-plane wavelength of SPs. However, this phenomenon has limitations on its spatial resolution. It is therefore necessary to add more ideas beyond

plasmonic effects to reach even better spatial resolution. A combination of SP with nonlinear properties of fluorescence can potentially help to get high spatial frequency information with the additional advantage of side-lobe suppression [28, 137]. For example, when a nonlinear dependence of the fluorescence emission rate on the illumination intensity is considered, it would cause the effective illumination pattern to contain harmonics with spatial frequencies that are multiples of the spatial frequency contained in the illumination. Thus, fluorescence nonlinearity in theory can produce an infinite number of harmonics, which leads to theoretically infinite resolution in structure illumination microscopy. Further, with utilizing the fluorescence nonlinear property of stimulated emission depletion, it also can significantly reduce the PSF of fluorescence image by introducing second illumination beam into SP-SWFM [24, 25]. Specifically, parallelized STED microscopy of doughnut or line array excitation profiles will be possible. This STED-SP-SWFM is an extremely powerful technique for the wide-field super-resolution fluorescence imaging.

(b) Integration of plasmonic microscope on a chip

Some modifications could be employed in order to integrate all the elements of the plasmonic microscope instrument on a chip. For example, more planar geometries of SP-SWFM are possible by replacing the imaging collection system composed of objective, tube lens and CCD, with integration of high-resolution opto-electronic sensor array elements, such as a complementary metal-oxide-semiconductor (CMOS), on the top of metal grating structure for direct collection of fluorescence images.

Illumination source can also be replaced by a semiconductor laser of smaller size to make the whole design truly compact.

(c) *Applications in biological sample imaging*

The primary focus of the work done in this thesis thus far has been on the proof on concept in resolution enhancement of microscope by using subdiffraction SP standing wave excitation patterns. The SP based structure illumination microscope is suitable for in vivo biological imaging applications because it utilizes visible light (low energy photon) while not requiring any special fluorochrome, and it is a highly parallelized scanning technique which can potentially achieve high-imaging speed. Due to the limited penetration depth of SPs in dielectric medium, the proposed SP-SWFM will find its applications primarily in the study of surface phenomena of biological samples. Hence, the adoption of our configuration will allow the observation on dynamic processes of live cells, such as video imaging of tubulin and kinesin dynamics in living *Drosophila* S2 cells with high-resolution [138, 139]. A variety of subwavelength-sized plasmonic devices to tailor SPs should also enable many interesting studies of cell behaviors in real time.

(d) *Explore the potential applications in nanolithography*

We experimentally demonstrated the generation of subwavelength-sized SP spot and standing wave patterns, which can also be dynamically tailored by adjusting the polarization and the initial phase of the excitation light beam. With a subdiffraction resolution, these SP interference patterns will have profound potential applications in large area lithography [140,

141]. It is a promising approach to enhance the fabrication resolution up to nanoscale by using high frequency illumination light source, such as deep UV ArF excimer laser.

AUTHOR'S PUBLICATIONS

Journal publications:

1. **Q. Wang**, J. Bu, P. S Tan, G. H. Yuan, J. H. Teng, H. Wang and X.-C. Yuan, "Subwavelength-sized plasmonic structures for wide-field optical microscopic imaging with super-resolution" **Plasmonics** DOI 10.1007/s11468-011-9324-2 (2012)
2. **Q. Wang**, J. Bu, and X.-C. Yuan, "High-resolution 2D plasmonic fan-out realized by subwavelength slit arrays," **Opt. Express** 18, 2662 (2010).
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6. P. S. Tan, X.-C. Yuan, G. H. Yuan and **Q. Wang** "High-resolution wide-field wide-field standing-wave surface plasmon resonance fluorescence microscopy with optical vortices" **Appl. Phys. Lett.** 97, 241109 (2010)

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1. **Q. Wang**, J. Bu, P. S Tan and X.-C. Yuan "Two dimensional standing wave surface plasmon fluorescence imaging by subwavelength slit arrays" SPIE NanoScience+Engineering conference (2011).
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3. **Q. Wang**, X.-C. Yuan and J. Bu "Modulation of surface plasmon polaritons (SPPs) for super-resolved imaging" OSA-COS Applications of Optical Metamaterials (2009).

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