In this paper, we report the observation of surface plasmon virtual probes in water by using near-field scanning optical microscope. The full-width half-maximum of the probe is as small as $\lambda_0/5.5$. Such deep-subwavelength sized plasmonic virtual probe may lead to many potential applications, such as super-resolution fluorescence optical imaging and optical manipulation.

Keywords: Surface plasmon polariton; virtual probe; deep-subwavelength.
1. Introduction

Surface plasmon polaritons (SPPs) are essential surface electromagnetic waves propagating along the interface of a metal layer and a dielectric. These particular near-field high-confinement characteristics and associated remarkable field enhancement trigger off great interests to a wide range of researchers from physicists, chemists, materials scientists and biologists.\(^1\) Renewed interest in SPPs comes from the recent advances of nanofabrication techniques, which offer the capability to tailor SPPs to subwavelength sized probes for specific applications in nano-optics, for instance, surface Raman enhanced imaging,\(^2\) high-precision fabrication,\(^3\) and super-resolution optical imaging.\(^4\)

SPP near-field probes are usually designed for generation and detection of evanescent photons in subwavelength size. A high-refractive-index prism or high-numerical-aperture oil immersion objective lens is the simplest demonstration for generating subwavelength SPP probes.\(^5\) Another most popular near-field probe is a metal-coated fiber tip with a subwavelength aperture at the end.\(^6\) Recently, isolated subwavelength energy hot spots are obtained and dynamically positioned by coherent control of nanoscale light localization in metamaterial.\(^7\) Active control of plasmonic field and eventually focusing at a freely predefined point on the surface of a nanohole array is also realized by optimizing the plasmonic phases with a digital spatial light modulator.\(^8\) The digital addressing and scanning of subdiffraction-limited SPP hot spot, not involving any mechanical motion, may make possible novel interdisciplinary applications in super-resolution imaging. However, just like all other scanning microscopes, they naturally bring about the disadvantages of the time-consuming point-by-point scanning schemes, which is difficult for fast operation and real-time imaging. Therefore, a wider-field optical imaging system with resolution beyond the diffraction limit will stir considerable interests especially in the biological fields. In this paper, we propose a versatile method to manipulate SPPs in water to form deep-subwavelength SPP standing wave virtual probes with the introduction of subwavelength-sized metallic structures. Essentially, these subdiffraction SPP virtual probes can be utilized as a wide-field fluorescence excitation profile since they contain subdiffraction-limited spatial frequencies.

2. Principles

Due to the intrinsic nature of electromagnetic wave, the wave solution of SPPs can be obtained via a straightforward solution of Maxwell’s equations with appropriate boundary conditions.\(^9\) As a result, the dispersion relation of SPP propagating along an interface of the metal and the dielectric is described as

\[
k_{\text{SPP}} = k_0 \left( \frac{\varepsilon_1(\omega) \varepsilon_2}{\varepsilon_1(\omega) + \varepsilon_2} \right)^{1/2},
\]

where the metal is described with a complex frequency-dependent permittivity function \(\varepsilon_1(\omega)\), and the dielectric permittivity \(\varepsilon_2\) is real. In optical frequency, with a negative permittivity of metal, the dispersion curve of the SPP always lies to the right of the free space photon with wave vector of \(k_0\). When the frequency \(\omega\) increases, the \(k_{\text{SPP}}\) dispersion deviates far further away from \(k_0\), which leads to larger wave vector and shorter SPP wavelength.

That means that SPPs on a plane interface cannot be excited by the free-space propagating light. Excitation of SPPs by light is possible only if the wave vector component of the exciting light can be effectively increased over its free-space value. The SPPs field can be generated by a grating when the SPP momentum matches the condition \(k_{\text{SPP}} = k_0 \sin \theta \pm mG\), where \(\theta\) is the angle of incident light, \(G = 2\pi/\Lambda\) is the grating constant and \(m = (1, 2, 3, \ldots)\). Specially, with a grating period (\(\Lambda\)) finer than the wavelength of light, the nonradiative diffraction results in an SPP evanescent field near the grating. This method is preferred to excite a particular SPP mode with a high efficiency.

In our experience, four perpendicular subwavelength slit arrays are utilized to generate SPPs propagating along the metal/dielectric interface. As a result, high-contrast SPP standing wave virtual probes can be designed by the interference of SPPs in the center area of slit structure. With their intrinsic standing wave nature, SPP probe patterns carry high spatial frequency, \(K_{\text{SPP}} = 4\pi \lambda_{\text{SPP}}\), where \(K_{\text{SPP}}\) is the spatial frequency of SPPs standing waves and \(\lambda_{\text{SPP}}\) the wavelength of SPPs.\(^10\) As such, the SPP standing wave virtual probes offer a unique opportunity as the excitation profiles in imaging to substantially include high frequency information, resulting in higher resolution.

In optical microscopy, a common method to improve the resolution is to shorten the wavelength
by filling high refractive index oil in the gap between the sample and the objective lens. With the similar technology, the spatial frequencies in SPP standing wave probes can be increased by shortening the SPP wavelength. Since the excitation condition of the SPP surface waves is very sensitive to the metal–dielectric interface condition: Small changes in the refractive index of an adjacent dielectric layer can bring dramatic changes in the dispersion characteristics of the SPP waves, the SPP wavelength can be substantially shortened by immersion in high refractive index medium without changing the optical frequency.

The most common high refractive index medium in biological application is water (1.33) and oil (1.51). With the calculation from formula (1), the effective wavelength of the water or oil immersion SPP standing wave on Ag film could be shortened by 29% or 39% respectively, compared to the case in air/Ag condition with 633 nm free-space incident light, which may effectively improve the resolution in bio-imaging applications.

3. Experimental Results

As depicted in Fig. 1, four perpendicular one-dimensional (1D) slit arrays are patterned on the 100 nm Ag film by focus ion beam (FIB) to convert free-space light into SPP waves propagating along the metal surface. The pitch of the slit array structure is designed as 450 nm in order to match the SPP momentum in-water under normally incident illumination with the excitation wavelength of 633 nm. In order to sustain the uniformity intensity of SPP interference fringes, separation between two parallel slit arrays is set as 6 µm, which is smaller than the propagation length $l_{SPP}$ (8.4 µm) of SPPs, calculated with formula: $l_{SPP} = 1/(2k_{SPP}^*)$, where $k_{SPP}^*$ is the imaginary part of $k_{SPP}$.

The characterization of SPPs in the water condition is realized with an NSOM system, which comprises a near-field detector and works in collection mode. The schematic configuration is shown in Fig. 2. The SPP waves have a typical decaying length in the perpendicular direction to the metal surface in the order of hundreds of nanometers. An aluminum-coated glass fiber is dipped into the SPP field to couple the evanescent electromagnetic waves to the aperture, where SPPs are converted into propagating modes and guided toward a detector.

The NSOM equipment in our experiment is a commercial system based on shearing force mechanism. In the system, the optical fiber attached to quartz tuning fork was excited for oscillations at the quartz resonance frequency (33 KHz) with certain initial amplitude. In common cases, when approaching to the metal surface less than 10 nm, the fiber probe oscillation amplitude decreases to reach the preset threshold value. With that distance, the feedback system was triggered off to maintain the fiber scanning of sample surface at this threshold condition. In our experiment, when the fiber with tuning fork approaches the air/water interface even far away from the metal surface, due to the surface tension force of water, the amplitude of the fiber probe dramatically decreases to below the pre-set threshold, which fails the feedback control. Practically, in order to circumvent this issue and
reduce the disturbance to fiber oscillation from the water–air interface, the water is dropped on the metal surface to immerse the structures and fiber tip, only after the fiber probe is landing to 20 µm above the sample surface. Then the tune fork is excited for oscillations with reasonable amplitude. The feedback control procedure is essential for detection of SPP fields in water-immersion condition, showing reasonable stability and safe operation of the NSOM tips.

The distribution of SPPs intensity on the plane of less than 10 nm above the structure surfaces are shown in Fig. 3. Since the SPP waves can only be excited by p-polarized incident light, when with the x-polarized light normal incident on the slit array structure, SPPs modes are excited by the left and right vertical slit array couplers, corresponding to wave vectors of +k_{SPP} and −k_{SPP} in both directions. Since the laser beam is normally incident on the sample, the SPP fields excited by each slit array are equal in phase and amplitude and counter-propagate toward the center area of structure to generate 1D SPP standing wave probes, as shown in Fig. 3(a).

The period of SPP standing wave probes at the center area is measured to be 230 ± 5 nm in average. The FWHM of fringes is equal to 115 ± 5 nm, corresponding to λ₀/5.5. Similarly, y-polarized light is applied to generate 1D standing SPP probes along horizontal direction with the same FWHM of interference fringes as the vertical one, as shown in Fig. 3(b). When the linearly polarized light is illuminated 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, while the other 50% of incident light is p-polarized for y-direction slit arrays. Therefore, as shown in Fig. 3(c), dot array shaped 2D SPP virtual probes are generated in the central area of slit array structure, due to the overlapping of both amplitude and phase of the four perpendicular propagating SPP waves. The period of SPP virtual probe arrays equals to 325 ± 5 nm along diagonal direction of four slit arrays. And the FWHM is 157 ± 5 nm, or λ₀/4. The characterization results agree well with the theoretical calculation of FWHM of 1D or 2D SPP standing wave probes (λ_{SPP}/4 or √2λ_{SPP}/4, respectively).

4. Discussions
A wider-field optical imaging system with a resolution beyond the diffraction limit will stir considerable
interests especially in the biological research. In order to image fluorescence objects of the subdiffraction feature size, the illumination fields must be modulated and contain high spatial frequency.

In this paper, subwavelength slit arrays are studied to excite SPPs on the metal surface. In the design, the edges of slit can be thought as a line of SPP point source. By patterning SPP couplers with such sharp edges at the metal film, high-contrast standing wave virtual probes are obtained by interference of SPPs in the center area of slit structure with the FWHM of \( \lambda_0/5.5 \). Moreover, the FWHM of SPP standing wave probes is determined by both the excitation wavelength and the available high effective-refractive-index \( (n_{SPP}) \) of the metal/dielectric interface. Typically, \( n_{SPP} \) is 1.05 for an Ag/air interface and 1.7 for an Ag/oil interface with 532 nm incident light. Therefore, there is possibility of significant reduction of the FWHM SPP standing wave probes to 80 nm by optimizing the design of slit arrays to meet SPP dispersion relation at Ag/oil interface with green laser excitation.

Furthermore, the deep-subwavelength SPP probes can also be dynamically tailored to phase shift as the excitation profiles of super-resolution wide-field fluorescence imaging. The profile gives a 2D lateral resolution enhancement in wide-field high-resolution fluorescence imaging. Although a super-resolution imaging may be achieved with well-known stimulated emission depletion (STED), and stochastic optical reconstruction microscopy (STORM), our approaches provide the possibility of highly compact, potentially integrated architectures within much smaller volumes. Additionally, with the surface wave properties of SPPs, the applicability of SPP virtual probes in wide-field fluorescence imaging applications is confined to the study of surface phenomena, such as observation of F-actin cytoskeleton in cell. Moreover, the imaging targets located only in the center area of slit arrays could be resolved and the samples under test are required to be fixed in the dielectric medium during the imaging process.

References