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Quasi-3D plasmonic coupling scheme for near-field optical lithography and imaging

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Near-field optical imaging and lithography rely on achieving both high resolution and efficient coupling. Particularly conventional near-field scanning optical microscopy (NSOM) suffers from the trade-off between resolution and efficiency. Planar plasmonic lens schemes can partially solve this issue utilizing plasmonic resonances, but the performance is not robust over a large range of sample materials. In this work we show a novel quasi-3D plasmonic scheme to focus light into the extreme subwavelength region in the near field with an efficiency orders higher than NSOM. The superb performance comes from the strong coupling between the localized mode with an off-plane E-field component and the sample being processed. Our scheme can efficiently focus light to a spot with a diameter down to 1/20 of its wavelength, and the coupling efficiency can be as high as 10%. Theoretically, we demonstrate that the FWHM of the focus spot can be 7 nm with an enhancement of about 800 at the UV region. The focusing performance is constantly good over a large variety of materials and the illumination and collection imaging scheme has been demonstrated by simulation. An example design of this quasi-3D coupling scheme is fabricated and its imaging performance is characterized by the apertureless optical near-field measurement. The high coupling efficiency at extreme subwavelength resolution of this quasi-3D coupling scheme opens the door to many applications, such as optical lithography, nanoscale imaging, heat-assisted magnetic recording, plasmon-enhanced Raman spectroscopy, etc. © 2015 Optical Society of America

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Coupling the propagating electromagnetic waves into the extreme subwavelength scale is important for many applications, including optical imaging, nanolithography, high-density optical and/or magnetic data storage, biosensing, nanospectroscopy, and molecular trapping [1–4]. The near-field scanning optical

microscopy (NSOM) has been widely used in optical imaging beyond the diffraction limit and great efforts have been devoted to achieving high resolution and sensitivity. By working in the optical near field, people can utilize the large wave vector components in the evanescent waves created by a subwavelength aperture to selectively excite and probe the features far smaller than the far-field optical diffraction limit in the superresolution lithography and imaging studies. In general, the ultimate resolution of NSOM is mainly limited by the skin depth of the opaque film to the order of a few tens of nanometers. And their major technical obstacle has been the trade-off between spatial confinement and energy transmission efficiency. The typical efficiency for optical power transmission through a NSOM probe is of the order of 10^{-5} or lower for an extreme subwavelength size opening, limited by the large wave vector mismatch between the aperture and propagating light [5–7]. The low energy efficiency limits the NSOM to scientific studies and cannot meet the practical needs for high-throughput lithography and imaging applications. Efficient light focusing has been realized by utilizing the large wave vectors of surface plasmon polaritons (SPPs). Various aperture- and antenna-based plasmonic structures have been proposed to enhance the subwavelength light spots in NSOM [8–10]. Despite the successes in achieving highly enhanced and confined light spots, the confinement of light is usually affected by the optical properties of the sample being scanned because their resonance is usually detuned by the change of sample properties at their close proximity. Another widely used approach is to use nanoscale objects, such as nanoparticles or surface apertures, to localize and produce an intense electromagnetic hotspot for lithography, material processing, and imaging. This approach has its advantages over aperture- and antenna-based plasmonic structures as to achieve high spatial confinement and this mechanism has been employed in the apertureless and scattering-type NSOM (aNSOM and sNSOM) [3,11–13]. However, in a typical aNSOM system, the optical energy is delivered by directly coupling the incident propagating light with the nanoscale particles or apertures. So only a small fraction of photons (usually 10^{-5}) can reach the optical localization site with a diameter of the order of nanometers, leading to order-of-magnitudes stronger background noises than the signal. Many techniques have been

employed to suppress the background noise and enhance the signal-to-noise ratio (SNR) [14,15]. As a result, the aNSOM techniques usually yield to low throughputs and complicated systems. Recently, many researchers have been working on improving both the resolution and throughput by developing new nanoscale plasmonic structures for NSOM and aNSOM applications [16,17].

Here, we report a new optical coupling scheme, named quasi-3D plasmonic coupling, which is capable to obtain more efficient optical coupling reaching the sub-10 nm region for the purposes of high-throughput lithography and imaging. In this new scheme, we utilize two sets of phase-mismatched SPP gratings to generate standing waves with preferable off-plane E-field components at the center. The obtained SPPs are then converted into localized resonant modes to excite a nanoscale pin structure. Importantly, the scheme can be optimized for efficient nanoscale energy delivery to a broad range of sample materials, which makes it a useful tool for applications in imaging, lithography, sensing, and data storage. Compared with other schemes that directly excite nanoscale tip or pole structures using propagating lights [18,19], the quasi-3D plasmonic focusing can provide optical coupling efficiencies reaching 10%, which is a few orders of magnitude higher. A novel type of plasmonic nanofocusing structure, called the push-pin plasmonic lens (PPL), is shown here as an example. We experimentally studied the performance of the PPL device with an aNSOM setup. Compared with the conventional aNSOM scheme, our numerical study also shows that the PPL can reversely collect the scattered near-field waves by the sample surface. Meanwhile, the collected near-field waves are converted back into far-field propagating light for imaging purposes without the needs of high numerical aperture optics. The achieved SNR is a few orders of magnitude higher than the conventional aNSOM or sNSOM scheme. In addition, the quasi-3D devices feature their planar thin-film structures and can be easily fabricated into a large array. It has the potential to enable the development of parallel quasi-3D plasmonic systems with high throughput.

A variety of thin-film-based plasmonic nanofocusing structures have been intensively studied in the literature to concentrate free-space lights into highly confined optical spots, such as bull's eye, C-shaped and H-shaped apertures, and the bowtie antennas [16,20–22]. During the imaging or lithography operations, the samples are placed in the optical near field of these plasmonic nanofocusing structures and processed by the fringing field near the metallic gap [23]. An extension of these plasmonic nanofocusing structures, named plasmonic lenses, are constructed by placing one of these apertures or antennas at the center of a set of concentric through-slit ring gratings as SPP couplers. Fine resolutions reaching the extreme subwavelength scale have been demonstrated using these plasmonic lenses in laboratory studies [24]. However, all of these nanofocusing structures still need improvements before entering imaging or lithography applications. Despite their enhanced transmission efficiency compared with a pin-hole aperture, the nanofocusing structures are limited to illumination-mode, which means the light can only be coupled from the far field to the near-field hot spot but not the other way round. They can only deliver a small amount of the optical energy to the sample as fringing field, which still leads to a trade-off between the resolution and energy throughput. To make matters worse, the desired resonances of plasmonic nanofocusing structures

are usually detuned by the permittivity changes of the sample surface.

Instead of exciting a localized gap resonance horizontally, our novel quasi-3D plasmonic coupling scheme excites the direct resonance between the plasmonic structure and the sample vertically. The high coupling efficiency allows us to create a reversible coupling from the local interaction between the plasmonic structure and the sample to the free-space backpropagating light, which can be used for reflecting mode near-field imaging and sensing. Figure 1(a) shows the oblique view of a typical quasi-3D plasmonic nanofocusing structure designed for working with a plane wave linearly polarized along the symmetry axis of the lens, named here PPL. The PPL has two sets of mismatched half-ring slit gratings on both sides of the pin. Each set of the slit gratings has the same period as the SPP wavelength (λ_{spp}) except that the first ring of the right-hand set is placed at $1\lambda_{\text{spp}}$ distance away from the pin while the left-hand side at $0.5\lambda_{\text{spp}}$. Upon the forward plane wave excitation, each slit can generate a localized SPP resonance and further excite the propagating SPP along the metallic surface. The two propagating SPPs launched by the mismatched gratings have a relative phase shift of π at the center, which allows us to construct SPP interference at the center and generate a standing wave with strong E-field along the off-plane direction. At this location, the lightning pole effect of the center pin further converts the background standing wave SPPs into a localized SPP mode with a strong off-plane E-field resonance between the push pin and the object being processed underneath. The localized mode forms a direct interaction between the collective resonance of the plasmonic nanofocusing structure and the local sample surface underneath the focus spot. This collective structure resonance can be converted back into backward propagating light and detected by measuring the intensity of the reflected light at the far field. By rastering the PPL over the sample surface, we can correspondingly construct an optical image of the material surface. Figure 1(b) shows the coupling scheme between the propagating light and the local mode with a strong off-plane E-field.

By aligning the localized SPP mode vertically with the centered pin structure, this quasi-3D scheme can provide an ultimate resolution down to the extreme subwavelength scale with high coupling efficiency. Figure 2(a) shows a FWHM spot size of about 17 nm is achieved with a peak enhancement of about 2500 times of incidence under 355 nm illumination. A silicon

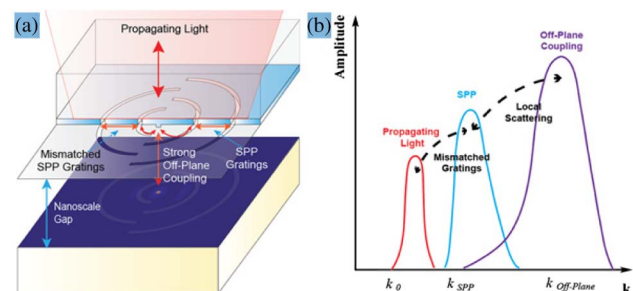


Fig. 1. (a) The 3D oblique view of push-pin plasmonic design. (b) Coupling scheme in wave vector space. k_0 is the propagating light wave vector. k_{spp} is the wave vector of the propagating surface plasmon polariton. $k_{\text{Off-Plane}}$ is the wave vector of the strong off-plane coupling mode.

wafer is placed 5 nm away from the top surface of the pin. The light intensity profile is obtained at the middle between the pin and the silicon wafer. In the above PPL design, aluminum is chosen as the PPL material for its low loss at 355 nm and sapphire is chosen as the substrate material where the PPL structure is placed on. The width of the grating slits is 40 nm and the radii of the left and right set of gratings are 140 and 380, 260 and 500 nm, respectively. The pin structure of 10 nm tall and 10 nm in diameter is placed at the center of the lens. When a pin structure of 5 nm tall and 5 nm in diameter is used as shown in Fig. 2(d), a FWHM spot size of 7 nm is achieved with a peak enhancement of about 800 times of incidence.

Compared with conventional designs, the nanofocusing of the quasi-3D scheme has a consistently good performance over a wide range of sample materials. Figure 2(b) shows the obtained intensity enhancement factors for a broad range of samples, including metallic, magnetic, polymer, semiconductor, and dielectric materials. All of the spot sizes are around 20 nm, which is in the extreme subwavelength region. As a comparison, Fig. 2(c) shows the corresponding performances of a bull's eye plasmonic lens [Fig. 2(c) inset] with and without another metallic object placed at its proximity. The illustrated bull's eye PPL is made of 80 nm thick Al film as lens materials upon plane wave excitations at 355 nm wavelength with linear polarization along the horizontal direction for our comparison study. As shown in Fig. 2(c), an 80 nm FWHM spot has been achieved at 20 nm (1/4 of focal spot diameter) away from the lens surface with an enhanced field intensity of about 10 times of incident, but when another silicon wafer is placed at 30 nm

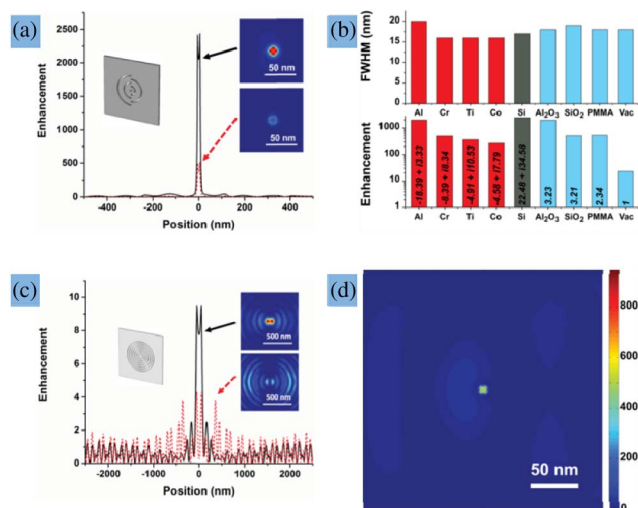


Fig. 2. (a) Light intensity profile at a distance of 2.5 nm from the 10-nm-diameter pin's top surface with (black solid line) and without (red dashed line) silicon at its proximity (5 nm away from the top surface of the pin). (b) Intensity enhancement factors and FWHM spot sizes for a broad range of samples, including metallic, semiconductor, and dielectric materials. 355 nm wavelength is used and the complex dielectric constants are listed in the histogram. (c) Performances of the bull's eye PL with (red dashed line) and without (black solid line) another silicon wafer placed at its proximity. The background material for the bull's eye PL is quartz with dielectric constant 3.21. There are 15 concentric rings and only part of them are shown in the inset. (d) Light intensity profile at a distance of 2 nm from the 5-nm-diameter-pin's top surface with Al sample at its proximity (2.5 nm away from the top surface of the pin).

away from the bull's eye structure, the previously achieved tightly focused field at 20 nm distance no longer exists. Instead, fringing fields appear at the edges of the center aperture and outer gratings. Compared with earlier plasmonic lenses, the PPL is suitable to work with different objects at its proximity with efficient optical focusing and coupling. All simulated intensity profiles shown in Fig. 2 and in the following figures are calculated using commercial electromagnetic wave software (CST Microwave Studio) and the results are normalized to the incident plane wave intensity.

Because of the high coupling efficiency, the PPL structure can also work in the reverse direction by converting the strongly localized field between the pin and the sample into backward radiation. We can get the local sample surface information underneath the pin by detecting the backward radiation in the far field. For imaging application, we studied an example PPL design at an optimized working wavelength of 633 nm.

As illustrated in Fig. 3(a), we experimentally examined the imaging performance of the quasi-3D coupling scheme using an sNSOM platform. This arrangement is equivalent to using a PPL as a scanning probe in sNSOM to map a nanoscale object, which is a flat-tip silicon probe (triangular shape) in this scenario. The experiment was conducted in tapping mode at the tapping height in the range of 5–10 nm. During the experiment, the PPL was illuminated from the backside at the wavelength of 633 nm. And the scattered light was collected using a high-sensitivity photodetector and analyzed by a lock-in amplifier to form an optical image of the silicon tip. By rastering the silicon probe over the PPL surface, we can map the optical image of the silicon probe. Figure 3(b) shows a scanning electron microscope (SEM) image of the PPL fabricated using standard microfabrication techniques. An 80 nm thick gold film was used as the lens material. The width of the grating slits is 50 nm and the radii of the left and right set of gratings are modified according to the SPP wavelength at gold surface as 205 and 565, 385 and 745 nm, respectively. The pin has a

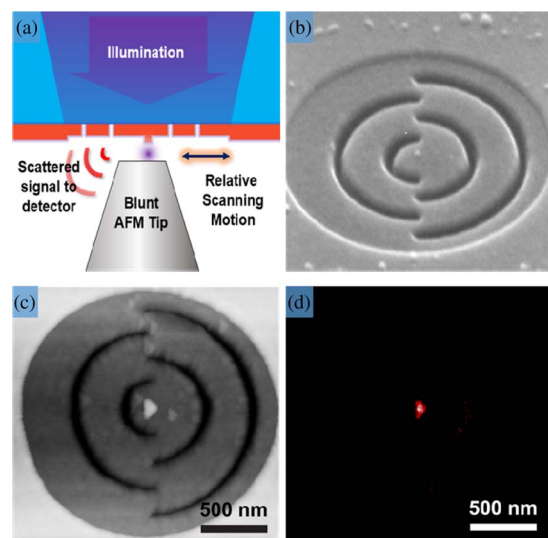


Fig. 3. (a) Imaging performance examined by apertureless NSOM. (b) Oblique view of the fabricated gold PPL structure SEM image. (c) AFM measurement of the PPL. The triangle shape reflects the top surface of the blunt tip. (d) NSOM measurement of PPL. The triangle hot spot reflects the shape of the NSOM tip.

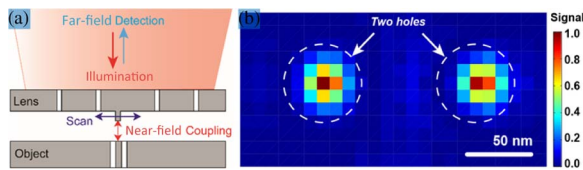


Fig. 4. Simulation result of near-field optical image using a PPL with 48 nm focusing spot size. (a) The PPL is illuminated from the backside and scans over a gold sample which has two 60 nm diameter holes with a 120 nm center-to-center distance. (b) The normalized near-field image obtained at 1 m away from the sample using interference.

diameter of 40 nm and a height of 20 nm and is placed 10 nm away from another gold surface. According to the simulation results, a peak enhancement factor of 2000 times can be achieved at the distance of 5 nm away from the pin. At the location of 10 nm away from the pin, the enhancement factor is about 800. Figure 3(c) shows the atomic force microscope (AFM) image of the PPL using the same blunt silicon tip. From this figure, we can clearly see the triangle shape of the top surface of the silicon tip. Figure 3(d) shows the sNSOM measurement result of the triangle silicon probe tip. Because of the finite size of the PPL pin, the mapped topography and the optical field are both affected by the pin-sample convolution. The optical image of the silicon tip is measured to have an overall size of 58 nm (along x) by 105 nm (along y), and the radius of curvature at the corner of the tip image indicates the potential of achieving an optical imaging resolution better than 10 nm.

We also numerically examined the imaging performance of the quasi-3D coupling scheme in the illumination-collection NSOM mode, i.e., using the PPL for both illumination and collection, as shown in Fig. 4(a). During the PPL's illumination-collection mode operation, the focused optical energy underneath the center pin strongly interacts with the sample and is coupled back by the PPL into far-field scattering light. This scattering light contains the local surface information and can propagate backward into propagating light because of the reciprocity of electromagnetic waves. By detecting the backscattered light, we can reconstruct the near-field optical image. Compared with sNSOM, this scheme also allows us to examine opaque samples without the need of high numerical aperture objective lenses and complicated signal processing techniques. This advantage opens up the opportunity of parallelize PPL structures to achieve high imaging throughput. Figure 4(b) shows an example of the near-field optical image obtained using a PPL with a center pin of 40 nm in diameter and 20 nm in height. The sample in the simulation is made of gold film which has two 60 nm diameter holes at a 120 nm center-to-center distance. A reference beam is used to interfere with the far field to suppress the background. The detection signal is defined as the normalized E-field amplitude detected at 1 m away in the normal direction. As shown in Fig. 4(b), the PPL can clearly distinguish the two pin-holes in the sample where the two peaks correspond to the centers of the two pin-holes. The ratio of the signal to the reflected light is about 0.1 percent in E-field amplitude.

In this Letter, we introduce a novel quasi-3D scheme to efficiently create an extreme subwavelength hot spot down to sub-10 nm. The high energy efficiency comes from the efficient coupling between the localized SPP having a large off-plane E-field component with the mismatched gratings. This also leads to a robust optical performance over a wide range of materials, which is crucial for superresolution optical imaging and sensing. The simulation and experiment results show its potential applications in optical lithography, nanoscale imaging, heat-assisted magnetic recording, etc.,

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