# Direct laser writing of pyramidal plasmonic structures with apertures and asymmetric gratings towards efficient subwavelength light focusing

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Abstract: Efficient confining of photons into subwavelength scale is of great importance in both fundamental researches and engineering applications, of which one major challenge lies in the lack of effective and reliable on-chip nanofabrication techniques. Here we demonstrate the efficient subwavelength light focusing with carefully engineered pyramidal structures fabricated by direct laser writing and surface metallization. The important effects of the geometry and symmetry are investigated. Apertures with various sizes are flexibly introduced at the apex of the pyramids, the focusing spot size and center-to-sidelobe ratio of which could be improved a factor of ~4 and ~3, respectively, compared with the conical counterparts of identical size. Moreover, two pairs of asymmetric through-nanogratings are conceptually introduced onto the top end of the pyramids, showing significantly improved focusing characteristics. The studies provide a novel methodology for the design and realization of 3D plasmonic focusing with low-noise background and high energy transfer.

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#### 1. Introduction

In the past decade, efficient delivering of light energy to nanoscale objects and concentrating this energy within dimensions as small as a few nanometers, have been of the main concerns and the heart of revolutionary in optical physics and technologies [1-3]. To this aim, the utilization of surface plasmon polaritons (SPPs)-propagating electron oscillations along the interface between a metal and a dielectric medium-has been proposed as a feasible mechanism for overcoming the diffraction limits of conventional optical devices [1, 2]. To achieve the focusing, localization and consequent enhancement of optical fields in nanoscales, various SPP structures, for instance, curved hole arrays [4], plasmonic rings [5], particle arrays [6], nanoattenna [7] and tapered waveguides [8], have been theoretically proposed or experimentally demonstrated. Among them, three-dimensional (3D) light focusing with metallic probes has attracted extensive studies [9–13] due to their successful applications in sophisticated technologies such as near-field scanning optical microscopy (NSOM) [14] and tip-enhanced Raman spectroscopy [15], where the tradeoffs among coupling efficiency, spatial resolution, and the ease for operation always existed. For example, Zhang *et. al.* in USTC [16] recently achieved sub-nanometer spatial resolution during the chemical mapping of a single molecule by using a STM-controlled metallic probe under external illumination. However, the external excitation efficiency was rather low (<10<sup>-5</sup>) and the strong background noises and thermal effects have limited the working condition to a ultra-high vacuum chamber with a low temperature of 80 K [16]. In comparison, the internal excitation of 3D probes possesses obvious advantages in avoiding background noise and thermal effects [10, 17]. However, most of the 3D probes under internal excitation rely on an aperture at the apex of the probe with a limited size (e. g., ~100 nm for the NSOM probe), which cannot be further decreased due to the cutoff of the internal waveguide modes (WGMs) [18, 19] at smaller aperture and therefore inherently limited the spatial resolution of the probes.

On the other hand, recent studies showed that 3D focusing of SPPs other than waveguide modes, i.e. 3D plasmonic focusing, could help to improve the spatial resolution of the internally excited probes since SPP modes have no cutoffs [9, 18, 20, 21]. To improve the light-SPP coupling efficiency (i.e. enhancing the light throughput of the probe) of 3D plasmonic focusing, people have introduced various modulations on the surfaces of the metallic probes, such as metallic gratings [9–11] and corrugations [17]. Recently, a new generation of tapered metal-insulator-metal structure, named as the campanile probe, has been proposed and demonstrated with nearly background-free imaging resolution of  $\sim 40$  nm [22– 25]. However, these 3D tiny features arouse big challenges with traditional top-down fabrication approaches, such as electron beam lithography (EBL) and focused ion beam (FIB) related techniques, in the cases of complexities, expenses, or time-consumptions. In comparison, direct laser wiring (DLW) method, based on two-photon polymerization of photoresist, has exhibited the capability of realizing arbitrary 3D structures with ultimately sub-10-nm resolution [26, 27], which has been successfully applied in fabrication of 3D photonic crystals [28], metamaterials [29], chip-scale interconnects [30], etc. Very recently, we have demonstrated DLW as a powerful tool to fabricate a spiral taper that possesses polarization-insensitive three-dimensional (3D) plasmonic focusing properties [31].

Here we show that metallic tapers in pyramidal shapes with apertures or asymmetric gratings exhibit advantages of high-throughput and hold the potentials towards efficient subwavelength light focusing. By using DLW followed by a metal deposition process, pyramidal metallic probes with apertures of various sizes are directly fabricated. It is found that the internal WGMs in the pyramidal structures are cut off at smaller aperture size than the conical structures, indicating that taper probes with pyramidal shape possess higher optical throughput than common conical tapers of identical size. Moreover, asymmetric nanogratings are conceptually introduced on the two opposite surfaces of the pyramidal structures, which greatly improve the focusing quality. The studies provide a novel methodology of design, realization, and application of 3D plasmonic focusing structures with low-noise background and high throughput.



Fig. 1. Calculated electric field intensity distributions of the tapers in the x-z plane in steady states. The internally incident light is polarized along x-direction with wavelength  $\lambda = 800$  nm. (a) Conical taper without aperture. (b) Conical taper with an aperture diameter of D = 120 nm. (c) Pyramidal taper without aperture. (d) Pyramidal taper with an aperture width of D = 120 nm. The white lines indicate the boundaries of 70-nm-thick Au film and the dark line denotes the inner boundary of the 360-nm-thick dielectrics. The horizontal white arrows indicate the position where the waveguide modes are cut off (defined as the position where the normalized light intensity is less than 1%). All image sizes:  $3 \times 6 \mu m^2$ . In calculations, the dielectric constant data of gold (or silver) are taken from [32].

#### 2. Three-dimensional hollow pyramidal plasmonic structures with apertures

To compare the transmission properties of conical and pyramidal tapers under internal excitation, Fig. 1 plots the simulated results of two conical and pyramidal tapers without and with apertures. It can be seen that for the tapers without apertures [Figs. 1(a) and 1(c)] and under internal excitation, the internal WGMs (indicated by dark red color) are cut off at a certain critical diameter (or width)  $D_c$ , while SPP modes coupled at the outer surfaces of the metal films can propagate towards the apex of the taper. Due to the symmetry of the structures, the electric fields of the excited SPPs have opposite polarities on the opposite sides of the taper, which cancel out each other when SPPs arrive at the apex [19, 31]. Therefore, no light can form a "focus" at the apex in both tapers [Figs. 1(a) and 1(c)].

An important finding from Figs. 1(a) and 1(c) is that the WGMs in conical taper are cut off at a larger radius than those in the pyramidal taper, i.e.  $D_{c-conical}>D_{c-pyramid}$ , as noted by the arrows in Figs. 1(a) ( $D_{c-conical} = 150$  nm) and Fig. 1(c) ( $D_{c-pyramid} = 80$  nm). Therefore, when the two tapers are opened with apertures of an intermediate size  $D_{inter}$ , for example  $D_{inter} = 120$  nm as shown in Figs. 1(b) and 1(d), the WGMs will still be stopped by the conical taper but can transmit through the pyramidal taper since  $D_{c-conical}>D_{inter}>D_{c-pyramid}$ . This means that for tapers with the same aperture size (determining the spatial resolution of the taper probe), tapers with pyramidal shape possess higher optical throughput than common conical tapers. For example, in Figs. 1(b) and 1(d), the calculated light intensity transmitted through the aperture of the pyramidal structure [Fig. 1(d)] is about 80 times of that of the conical structure [Fig. 1(b)]. In other words, under the condition of obtaining the same transmission efficiency, the pyramidal tapers will have an aperture size much smaller than the conical tapers, and thus have higher spatial resolution if used as a probe. Moreover, the square pyramidal shape is also preferred by the wet chemical etching of Si with KOH aqueous solutions [11].



Fig. 2. Characterizations of pyramidal structures with apertures. (a, b) SEM images of a hollow pyramidal structure with an aperture size of D = 300 nm. (c) Side-view SEM images of the pyramidal structure after cutting by FIB. (d) Crosscut intensity (normalized) and false-color microscope images of the "focuses" of the fabricated conical and pyramidal tapers under internal excitation with aperture size of D = 260 nm. The images are measured with an Olympus microscope (BX51) equipped with a  $100 \times$  objective (NA = 0.9) and a CCD camera. (e) Top-view SEM images of pyramidal structures with various apertures. From left to right: D = 40, 140, 220, 240, 260, 300, and 440 nm. (f-g) False-color microscope images of the "focuses" of the structures in Fig. 2(e) with 70-n-thick and 200-nm-thick gold layer, respectively, under internal excitation. Images in Fig. 2(f) were measured in wavelength region of 400-800 nm with unpolarized white light. Images in Figs. 2(d) and 2(g) were measured in wavelength region of 650-750 nm under x-polarized excitation. Microscope image sizes:  $5 \times 5 \mu m^2$ .

To demonstrate the important feature of the pyramidal tapers, metallic hollow pyramidal structures are fabricated by using the DLW and a metal deposition process. The DLW we used is based on 2PP technique employed by a commercial fabrication system (Photonic Professional, Nanoscribe GmbH) [31]. In fabrication, a 780 nm femtosecond laser beam (with pulse width ~120 fs and repetition rate ~80 MHz) is focused into a negative photo-resist (IP-L) by a high numerical aperture (NA) oil-immersion objective  $(100 \times , NA = 1.4, Zeiss)$ . The polymerized elliptical voxel, as the replica of the laser focus after polymerization, is about 360 nm in width and 1000 nm in height [31]. For the fabrication of pyramidal shapes, the laser focus is scanned layer-by-layer with square routes in each layer, while between neighboring layers, the height is increased by dz and the width is decreased by dx = 40 nm (in our following studies, dz/dx = 4 is used for tapers with apertures to achieve high spatial resolutions and dz/dx = 2 is used for the pyramids with nanogratings to achieve high transmission). The fabrication time can be controlled by setting the scanning speed of the piezo stage and it takes less than 30 minutes to fabricate a pyramidal structure around 50 µm in height, which is much faster than conventional EBL and FIB related techniques. After the DLW process, a 70-nm-thick gold film is deposited onto the structures through magnetron sputtering. Towards applications of different kinds, the hollow structures can be fabricated on cover glass, metallic film, copper grids, etc. In the case of fabrication of hollow pyramidal structures on cover glass, a hollow square frame is fabricated as the basis [31], whose width, height, and thickness are 26, 10, and 4  $\mu$ m, respectively, with a hole of 4  $\times$  4  $\times$  6  $\mu$ m<sup>3</sup> in each side.

#241502 © 2015 OSA With the arbitrary 3D fabrication capability of the DLW method, pyramidal tapers with various apertures can be directly fabricated by simply terminating the writing programs of the pyramidal structures at certain height (similarly as the fabrication of conical structures with various apertures in [31]). One advantage of this direct fabrication process is that it can bypass the FIB milling and avoid any pollution accompanied by  $Ga^{3+}$  ion implantations. As shown in the SEM images of Figs. 2(a)-2(c) and 2(e), apertures with sizes down to sub-100 nm are realized. In principle, by carefully optimizing the design parameters, subwavelength apertures with arbitrary sizes could be directly fabricated, holding potentials for the design of NSOM probes with a pyramidal shape.

Figure 2(d) shows the focusing properties of a conical and pyramidal taper with the same aperture size (with diameter/width D = 260 nm) fabricated and measured under the same condition. It can be seen that the conical structure shows strong sidelobes around the apex of the taper, the FWHM (full width at the half maximum) of the crosscut curve is ~2 µm. In comparison, the pyramidal structure exhibits intensified focusing spot with FWHM of ~500 nm (corresponding to 0.67 $\lambda$ , which is also limited by the imaging resolution of the microscope objective with NA = 0.9) and its center-to-sidelobe ratio is enhanced by a factor of ~3. It should be mentioned that due to the repolymerization effects of neighbored zones [26] near the small apertures, the aperture shapes of the pyramidal structures are not strictly square. The smaller the aperture width, the more circular the aperture shape, as shown in Fig. 2(e). This experimental degradation in aperture shape weakens the advantages of pyramidal structures over conical structures, making the results not as remarkable as those calculated in Fig. 1. Nevertheless, the improvements in focusing quality in Fig. 2(d) clearly demonstrate that the taper probe with a pyramidal shape processes potential advantages of high throughput, high resolution and high center-to-sidelobe ratio.

As already illustrated in Fig. 1(c) and 1(d), the WGMs that are cut off at certain position of the pyramidal taper can transmit through the apertures when increasing the aperture sizes. This is clearly observed in Figs. 2(e) and 2(f) that under small apertures only scattered SPPs can be seen due to the cutoff of WGMs and when the aperture size is increased beyond the critical dimension, the WGMs start to transmit through the aperture and both light from WGMs and SPPs can be imaged. The critical aperture size of the pyramidal taper measured here is around 110 nm (in half width). This is 30% smaller than that of conical tapers in [31] (~160 nm), the trend of which is consistent with the simulation results in Fig. 1. It should be mentioned that in Fig. 2(f), strong background noise is observed, which is caused by the radiation losses of the SPPs as illustrated in Figs. 1c and 1d. This kind of noise can be readily avoided by simply increasing the thickness of the deposited Au layers. For example in Fig. 2(g), when the Au thickness increases from 70 to 200 nm, the background noise nearly disappears.

## 3. Symmetry-broken hollow pyramids with through gratings

It has been shown that the introduction of asymmetric gratings or corrugations could help to increase the efficiency of external plasmonic focusing [11, 33]. However, the proposed fabrication process was complicated and the external coupling scheme may induce strong background noises and thermal effects. Here we show that asymmetric nanogratings can be readily introduced into our internally illuminated pyramidal structures. To this end, FDTD simulations were firstly conducted to test the mechanism. As shown in Figs. 3, due to its symmetric geometry, the pyramidal structure with symmetric gratings does not show any focusing effect near the apex due to the cancelling effects of SPPs that couple through the gratings and propagate towards the apex. In comparison, the asymmetric gratings in Fig. 3(b) break the structural geometry and shift the phase of the coupled SPPs, which therefore results in the subwavelength focusing of SPPs at the apex of the pyramidal taper in Fig. 3(d) [with FWHM of ~21 nm in Fig. 3(f)]. Moreover, the through slits of the nanogratings allow for the efficient coupling between WGMs and SPPs, which dramatically enhances the intensity of light focused at the apex by ~29 times.

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Fig. 3. Simulation results of pyramidal structures with through nanogratings. (a, b), Schematic view of the hollow pyramids with symmetric and asymmetric gratings through the two opposite surfaces of the pyramids under dz/dx = 2. (c-f), Simulated E-field intensity distribution in the x-z plane (y = 0) and x-y plane (10 nm top from the apex) for the Au-coated pyramidal taper with (c, e) symmetric and (d, f) asymmetric gratings (periodicity dg = 400nm), respectively, under internal cexitation by x-polarized light with  $\lambda = 700$  nm. The insets in Figs. 3(c) and 3(d) show the enlarged pictures of corresponding mages at y = 0 under the same scale. The FWHM of the focus spot in Fig. 3(f) is ~21 nm under simulative grid size of 8 nm. Slit distance: ds = 400 nm. Slit width: 100 nm. Image sizes: (c) and (d) are in  $4 \times 4 \mu m^2$ ; (e) and (f) are in  $1 \times 1 \mu m^2$ .



Fig. 4. Experimental results of pyramidal structures with through nanogratings. (a-c) SEM images of the pyramidal tapers with (a) symmetric gratings (SG) and (b, c) asymmetric gratings (ASG), respectively. (d) False-color microscope images of the "focuses" of the fabricated pyramidal structures with SG and ASG, respectively, measured in wavelength region of 650-750 nm with light polarized along x-direction. Image size:  $10 \times 10 \ \mu\text{m}^2$ . (e) Normalized crosscut intensity of the image in Fig. 4(d) in the x-direction through the "focuses". The ASG structure shows improved focusing quality with linewidth reduced by a factor of two. Structural parameters:  $dg = 400 \ \text{nm}$ ,  $dx = 400 \ \text{nm}$ , dz/dx = 2. To avoid the apex of the pyramids, i. e.,  $ds = 1.4 \ \text{µm}$ .

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For experimental demonstration, the pyramidal tapers with through nanogratings are directly fabricated by simply terminating the writing programs in the slit areas by using DLW, similarly as the fabrication of pyramidal structures with apertures. As shown in Figs. 4(a)-4(c), pyramidal structures with symmetric gratings (SG) and asymmetric gratings (ASG) through the two opposite surfaces of the pyramids are readily fabricated. Figure 4(d) shows the false-color optical microscope images of the "focuses" of the fabricated pyramidal structures with SG and ASG, respectively. It can be clearly seen that due to the cancelling of the SPPs at the apex of the symmetric pyramidal structures [Fig. 3(c)], the "focuses" SG structures are blurred. In comparisons, the ASG structure shows improved focusing quality with higher intensity and the FWHM of its plot in Fig. 4(e) is compressed by a factor of  $\sim 2$ . We'd like to point out that the improvement in focusing quality is not as large as the calculated results as shown in Figs. 3(e)-3(f), which is caused by two main factors. First, the fabricated structures are not as sharp as the theoretical designs in Figs. 3(a) and 3(b). Second, the far-field characterization cannot reflect the complete information in the near field. Nevertheless, the optical information in near field and far field are always correlated for certain type of structures, and qualitatively, the near-field simulations and far-field measurements are reasonably well in consistence. These preliminary results clearly demonstrate the feasibility to enhance the throughput and improve the focusing quality of the pyramidal structures by introducing asymmetric gratings with the DLW fabrication process.

## 4. Conclusion

In conclusion, by using FDTD method and DLW fabrication process, we have demonstrated the fabrication of hollow pyramidal plasmonic structures towards efficient subwavelength light focusing. Pyramidal metallic probes with apertures of different sizes were directly fabricated by using DLW, which showed smaller WGM-cutoff widths and improved focusing properties compared with the commonly employed conical taper probes. This gives one an important clue that for the aperture tapers with the same size (spatial resolution), taper probes with pyramidal shape possess higher optical throughput than common conical tapers. Moreover, asymmetric nanogratings were conceptually introduced on two opposite surfaces of the pyramidal structures by using the DLW process, which greatly improved both the focusing quality.

It should be mentioned that in this work, we mainly focused on the geometry correlated optical properties and comparisons between pyramidal and conical tapers with apertures, as well as the effects of asymmetric nanogratings, without intentionally pushing the fabrication resolution of DLW towards its limit. Consequently, the sharpness of the preliminarily fabricated tip (with tip radius of ~100 nm) is still not competitive with the reported TERS tips [9, 16]. However, the DLW technique has been valued for its far-field and arbitrary 3D characteristics, and it has been proved that by utilizing STED-assisted DLW [34] or the photoinhibition strategy [27], the DLW fabrication resolution could be significantly improved with a feature size down to ~10 nm. Moreover, the DLW-fabricated structures may also provide arbitrary 3D templates for finer secondary modifications with focused-ion-beam milling [9, 22] or for probes with novel designs [35]. Nevertheless, our studies provide a novel methodology and platform of design, realization, and application of 3D plasmonic focusing structures with low-noise background and high throughput.

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