Self-aligned grating couplers on template-stripped metal pyramids via nanostencil lithography

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We combine nanostencil lithography and template stripping to create self-aligned patterns about the apex of ultrasmooth metal pyramids with high throughput. Three-dimensional patterns such as spiral and asymmetric linear gratings, which can couple incident light into a hot spot at the tip, are presented as examples of this fabrication method. Computer simulations demonstrate that spiral and linear diffraction grating patterns are both effective at coupling light to the tip. The self-aligned stencil lithography technique can be useful for integrating plasmonic couplers with sharp metallic tips for applications such as near-field optical spectroscopy, tip-based optical trapping, plasmonic sensing, and heat-assisted magnetic recording. *Published by AIP Publishing.*
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Near-field scanning optical microscope (NSOM) systems frequently utilize metal probes possessing an aperture,¹ nanoparticle,^{2,3} or tip,^{4,5} the dimensions of which are smaller than the free-space wavelength of light used in the measurement. Such probes act as optical antennas⁶ and are capable of focusing light below the diffraction limit and obtaining optical information in the vicinity of the probe with nanometer-scale resolution. Tip-based probes have widely been used because their nanofocusing mechanism produces an intense, nanometric spot of light at the tip.^{4,7,8} To convert free-space radiation into localized near-fields at the tip, however, a non-trivial optical excitation or an asymmetrically patterned tip is often necessary. For example, to produce intense near-fields around a symmetric tip such as a tapered gold wire, either a radially polarized beam at normal incidence or a linearly polarized beam at side illumination should be used.⁹ Alternatively, the shape of the metal tip can be engineered^{10–12} to convert a normally incident linear or circularly polarized beam into surface plasmons that converge at the tip, simplifying beam shaping and optical alignment.

Among various techniques to produce metallic tips for NSOM,^{13,14} template stripping has shown promising results in terms of mass-producing sharp pyramidal tips.^{11,12,15,16} In template stripping,^{15,17,18} metal is deposited into an ultrasmooth Si mold, and an adhesive such as epoxy is applied to pull the metal out of the mold, transferring the shape and smoothness of the mold to the metal. Metal pyramids template-stripped from inverted pyramid molds formed by crystal-orientation-dependent wet etching of Si have been used as NSOM probes due to their high quality (sub-10 nm tip radius of curvature, sub-1 nm roughness) and reproducibility.¹⁶ Hot spots on template-stripped pyramidal tips have been achieved by using a radially polarized beam¹⁶ on smooth pyramids or by milling gratings around the tip such that incident light generates plasmons which focus and interfere constructively at the tip.¹¹ Fabricating grating patterns

on three-dimensional (3D) tip structures, however, is challenging, and previously has been accomplished using focused ion beam (FIB) lithography to manually define a grating-coupler pattern on the pyramid facets¹¹ or on the surface of a tapered gold wire.¹⁹ Since each pyramid must be addressed manually, the FIB alignment process is difficult and time-consuming. In this work, we improve the throughput and uniformity of patterned pyramid fabrication while maintaining much of the alignment accuracy and resolution that FIB lithography affords by combining template stripping with nanostencil lithography to integrate self-aligned gratings around the sharp pyramidal tip. Nanostencil lithography is a versatile technique to create patterns by depositing the material through a patterned stencil mask.^{20–25} We modify this approach by using the stencil mask itself to etch an inverted pyramid mold in an Si substrate and then deposit self-aligned nanometric bumps and gratings through the same mask into the mold. Our stencil-based template fabrication improves the throughput over FIB lithography while faithfully projecting complex planar patterns in the stencil mask onto all facets of the 3D metallic pyramids.

The substrate is a (100) Si wafer coated with a 40 nm-thick Si₃N₄ film. Electron-beam lithography (Vistec EBPG5000+) is used to define the patterns, which are then etched through the Si₃N₄ film via reactive ion etching. The etching parameters are as follows: 30 mT of pressure at 100 W RF power, with 12 sccm CHF₃, 42.5 sccm N₂, and 3 sccm O₂. The patterns consist of a ring split into four segments by unexposed gaps and a stencil pattern in the center of the ring (Figure 1(a)). The ring segments are used to create the pyramid mold base, while the pattern in the center is the stencil that later defines the pattern on the pyramid facets. The wafer is then placed in a potassium hydroxide (KOH) bath, which defines the pyramid molds by anisotropically etching the exposed Si along its slowly etching {111} crystal planes.²⁴ These planes join at the center of the ring segments to form the tip apex. The result is an inverted pyramid in the Si with a patterned Si₃N₄ membrane suspended above (Figure 1(b)). The unexposed regions that separate the ring

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FIG. 1. Fabrication process diagram for patterned metal pyramids. (a) The stencil pattern and features to define the pyramid are simultaneously etched into the nitride (top-view). (b) After KOH etching, an inverted pyramid is created in the Si leaving the nitride stencil suspended over it. (c) Oxide is deposited on substrate through the stencil, allowing the pattern to be transferred into the mold. (d) The suspended nitride stencil is removed. (e) Au is deposited into the mold. (f) Then the pyramid is template stripped from the mold, producing a patterned Au pyramid.

segments bridge the patterned membrane to the rest of the Si_3N_4 film. SiO_2 is then directionally evaporated onto the substrate (Varian 3118 Thermal Evaporation System). The Si_3N_4 stencil defines the pattern of the SiO_2 in the mold (Figure 1(c)). The sample is then milled using an argon-ion milling system with the sample surface inclined at a 5° angle from parallel to the beam. This removes the top SiO_2 layer, while leaving the patterned SiO_2 at the bottom of the molds relatively untouched. The Si_3N_4 is then etched away in a phosphoric acid (H_3PO_4) bath (Figure 1(d)). The ion-milling step is necessary because the phosphoric acid is not selective enough between Si_3N_4 and evaporated SiO_2 to undercut the SiO_2 and lift it off of the substrate before all of the SiO_2 is etched away. Finally, gold is deposited (CHA SEC-600 electron-beam evaporator) onto the substrate (Figure 1(e)) and is template-stripped using optical adhesive (NOA 61, Norland Products, Inc.) as a backing layer to yield an array of patterned pyramids (Figure 1(f)).

Using this method, patterns can be easily transferred from the stencil and integrated into the pyramid. Scanning electron micrographs (SEMs) of a pyramid containing a spiral pattern are shown in Figure 2. The suspended stencil over the etched pyramid can be seen in Figure 2(a). The spiral shape in the center will define the pattern. The layer of oxide evaporated through the stencil can be seen clearly after the stencil is removed in Figure 2(b). Since the features which define the position of the pyramid and those defining the stencil are made in a single step, the patterns are self-aligned to the pyramid. One confounding factor for placement accuracy of the oxide patterns is that the primary direction of the evaporation may not be exactly normal to the substrate interface and will

FIG. 2. SEMs of the patterned pyramids at various stages in the fabrication process. (a) Nitride membrane with a spiral pattern suspended over an etched pyramid. (b) After the oxide is deposited and the stencil is removed, a spiral-shaped oxide pattern is left inside the pyramid mold. (c) Top view of a template-stripped, patterned gold pyramid and (d) a side view of a nominally identical pyramid. (e) and (f) show the stencil and resulting patterned pyramid from using a linear diffraction grating design. All scale bars are $1\ \mu\text{m}$.

therefore offset the deposited patterns from the center of the mold. By comparing the actual placement of the oxide in several pyramid templates to the nominal position of the patterns, the average alignment error was found to be $32 \pm 6\ \text{nm}$ along a pyramidal facet. Additionally, as the distance from the membrane to the bottom of the pit is several microns, the deposited oxide will blur relative to the pattern in the membrane. For the $5\ \mu\text{m}$ base-width pyramids used in this work, the line width of the deposited oxide patterns blurred from the nominal design of $150\ \text{nm}$ to an average of $220\ \text{nm}$ in the template. The line height of the transferred patterns is less than the $200\ \text{nm}$ of SiO_2 which was deposited and initially defined the patterns, but this is to be expected since both the ion mill and the phosphoric acid bath have a nonzero etch rate of the patterned, evaporated oxide in the mold. The SEMs show that the groove depth is in excess of $50\ \text{nm}$, which is sufficient for generating plasmons to focus light at the tip. After the metal deposition and template stripping, the gold pyramid can be seen with the integrated spiral pattern (Figures 2(c) and 2(d)). It is also easy to make different patterns. As an example, a center-offset line grating pattern was made around the tip of a pyramid and is shown in Figures 2(e) and 2(f). The yield of patterned pyramid structures from this process is

approximately 95%, with only one or a few structures out of seventy-two failing to template-strip properly in the last step.

Through slight modifications to the process, it is also possible to produce metal nanoparticles in the pyramid molds. After the stencil is formed and suspended above the pyramid, Au is evaporated into the mold, rather than SiO₂. The stencil membrane is then removed and yields gold nanoparticles with shapes defined by the stencil, transferred onto the 3D facet of the pyramid. Some examples of Au nanoparticle structures can be seen in Figure 3. After these structures are made in the mold, they can be template stripped out and result in Au nanoparticles embedded in a dielectric pyramid.

The photogeneration of plasmonic hot spots using circularly polarized light has been investigated by many groups.^{26–28} The polarization state of circularly polarized light can be decomposed into the sum of its radial and azimuthal polarization components $\hat{E}_r$ and $\hat{E}_\phi$ ²⁹

$$\hat{E}_{RHC} = \frac{1}{\sqrt{2}} e^{i\varphi} (\hat{E}_r + i\hat{E}_\phi),$$

where the factor $e^{i\varphi}$ is known as the geometric phase. Only the radial component of the polarization is a transverse magnetic (TM) wave relative to the patterned grooves on the pyramid, so only that component will launch surface plasmons towards the tip. The plasmonic field contribution at the tip from a ring structure a uniform distance away from the tip will sum to 0 due to the spatial $e^{i\varphi}$ phase term. In order to gain field enhancement at the tip, the geometry of the plasmonic structure must be tuned, so that the launched plasmons have an $e^{-i\varphi}$ phase offset. An Archimedean spiral with opposite

chirality to the incident circularly polarized beam provides such a phase difference, causing the plasmons at the tip to interfere constructively and form a hot spot (Figure 4(a)). Conversely, when the spiral and light are of the same handedness, the plasmons will defocus at the tip (Figure 4(b)).

Finite-difference time-domain (FDTD) modeling was performed using Lumerical FDTD Solutions to gauge the effectiveness of linear and spiral grating patterns at focusing light to the tip. For both patterns, the optically thick gold pyramid was oriented with the 5 μm base parallel to the xy-plane on an epoxy substrate. Illumination was from above using a planar pulse centered in the near-infrared. The dielectric function of gold used in the simulation was fitted to experimental data for template-stripped Au.³⁰ Time-averaged field maps were acquired in the xz-plane, which bisects the pyramid. The Archimedean spiral groove on the pyramid facets can be described in polar coordinates by $r = (1 + \frac{\theta - \pi}{2\pi})600$ nm for right-handed spirals. The linear grating is offset from the tip by 360 nm in one direction and 480 nm in the other, with subsequent grooves offset by 240 nm. Perfectly matched layers were set as the boundary conditions in all dimensions. As expected, both left-handed circular and transverse magnetic (TM) polarization focused plasmons at the tip of the spiral and linear grating pyramids, respectively, while the complementary polarizations defocused light at the tips (Figure 4). While full optimization of 3D grating patterns for plasmonic nanofocusing is left for future work, our fabrication method makes it possible to readily transfer 2D patterns from a stencil mask onto 3D pyramid facets, opening up additional possibilities to design optical antennas.

FIG. 3. SEM images of three gold structures in pyramid molds. (a) A bowtie-shaped nanoantennas, (b) x-shaped nanogaps, and (c) a single gap on the edge of the mold apex. All scale bars are 500 nm.

FIG. 4. Simulated maps of the magnitude of the z-component of the electric field present of patterned gold pyramids when illuminated from above. In the top two panels, both pyramids possess a right-handed spiral groove structure. They are illuminated with (a) left-hand circularly polarized light producing a clear hot spot at the tip and with (b) right-hand circularly polarized light, exhibiting destructive interference of the electric field at the tip. The bottom two panels are from pyramids with an asymmetric linear diffraction grating illuminated with (c) linear polarization parallel to the x-plane (TM), producing a hot spot, and with (d) polarization perpendicular to the x-plane that does not produce significant enhancement. All tips have a 30 nm radius of curvature, and scale bars in all insets are 50 nm.

In conclusion, we have combined stencil lithography and template stripping to fabricate patterned metal pyramids with self-aligned nanometric gratings. By using a single patterning step to define both the pyramid and the gratings, they are self-aligned to each other and allow for large arrays to be fabricated in parallel. The resulting structures could be useful in many applications. For example, it is possible to couple incident light to the pyramid apex without the need for a radial polarizer that requires nanometric beam alignment. Simulations predict that both the spiral and asymmetric linear grating structures presented in this work are capable of efficiently coupling light to the tip. It is advantageous to pattern metal pyramids using stencil lithography, because the patterns are self-aligned to the sharp pyramidal tip as part of the mold fabrication process. As a result, the throughput is improved over what is achievable using FIB lithography, and the repeatability of patterns is largely maintained. Because these patterned Si molds are reusable and possess smooth facets, this stencil-based template fabrication method can further improve the utility of template stripping for applications in near-field optics and plasmonics.

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