Research article

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Multilevel phase supercritical lens fabricated by synergistic optical lithography

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Abstract: The advent of planar metalenses, including the super-oscillatory lens (SOL) and the supercritical lens (SCL) with distinctive interference properties, has profoundly impacted on the long-lasting perception of the far-field optical diffraction limit. In spite of its conspicuous success in achieving marvelously small focal spots, the planar metalens still faces tough design and fabrication challenges to realize high focusing efficiency. In this work, we demonstrated a dual-mode laser fabrication technique based on two-photon polymerization for realizing the multilevel phase SCL with focusing efficiency spiking. Synergistically controlling two types of movement trajectory, which is implemented with the piezo stage and the scanning galvo mirror, enables the fabrication of complicated structures with sub-diffraction-limit feature size. By utilizing such advantage, SCLs with discretized multilevel phase configurations are explicitly patterned. The experimental characterization results have shown that a four-level phase SCL can focus light into a sub-diffraction-limit spot with the lateral size of $0.41 \lambda/\text{NA}$ (NA is the numerical aperture), while achieve the focal spot intensity and the energy concentration ratio in the focal region 7.2 times and 3

times that of the traditional binary amplitude-type SCL with the same optimization conditions, respectively. Our results may release the application obstacles for the sub-diffraction-limit planar metalens and enable major advances in the fields from label-free optical super-resolution imaging to high precision laser fabrication.

Keywords: supercritical lens; sub-diffraction-limit focusing; two photon-optimization optical lithography; multilevel phase; high efficiency.

1 Introduction

Due to the capability of achieving sub-diffraction-limit light field modulation beyond the Rayleigh criterion, planar diffraction metalenses have been heavily appreciated since the theory of optical superoscillation was proposed [1–5]. As their typical representatives, the super-oscillatory lens (SOL) [6–10] and the super-critical lens (SCL) [11–14] have been intensively investigated in the past few years. Through delicately controlling the interference effect, band-limit functions can oscillate much faster than its highest Fourier components in a certain region, leading to a sub-diffraction-limit focal spot in the far field and further inspiring versatile application potentials, such as far field optical nanoimaging [6, 12], nanometrology [15], nanofabrication and big-data optical storage [7], etc. Nevertheless, most of the previous sub-diffraction-limit planar metalenses have been demonstrated with binary amplitude configuration [6, 12, 16], leading to inefficient focusing with low contrast. Although binary phase configuration can boost the efficiency to some extent [17–20], its intrinsic symmetry characteristic drains a large portion of light energy in the interference process from the higher order diffraction and the secondary focusing, which severely jeopardizes the focusing capability of planar metalenses.

To eliminate these obstacles, discretizing the binary configuration into multilevel phase configurations is supposed to be the most effective scheme [21–25]. The new configuration brings in, however, unprecedented

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conundrums for precisely realizing nerve-wracking structural complexity in practice. The sub-diffraction-limit feature size required by the planar metalens inevitably surpasses the fabrication capability of the conventional mask-less direct laser printing technique, the resolution of which is restricted to hundreds of nanometers. Accordingly, nonlinear optical lithography techniques based on two photon polymerization (TPP) offer a new way to address this problem for tremendous superiorities in fabricating deep subwavelength features with unique three-dimensional (3D) fabrication capability [26–30]. The recent development of optical super-resolution laser fabrication even improves the fabrication resolution up to 9 nm, which allows for more precisely modulating the phase accumulation for light traveling through a lens [29]. Nevertheless, current nonlinear optical lithography techniques still have some major difficulties yet to be settled for fabricating specially designed multilevel phase planar metalenses [31–34]. The structural feature sizes in lateral directions and longitudinal directions are commonly correlated due to the joint influence from the aspect ratio of diffraction-limit focal spot and the photopolymerization threshold level, which excludes the realization of the multilevel phase planar metalenses.

In this work, we demonstrate a focusing efficiency spiking of multilevel phase SCLs fabricated by a dual-mode two-photon polymerization optical lithography technique. Through synergistically controlling the movement trajectory of a piezo stage and galvo mirrors, the correlation of the feature size between lateral directions and longitudinal directions can be broken. The width and the thickness of the patterned line can be flexibly manipulated. As a proof-of-principle demonstration, a multilevel phase supercritical lens capable of efficiently focusing light into sub-diffraction-limit region with high intensity contrast has been experimentally demonstrated (Figure 1A). Such lens consisting of varied belt widths and thicknesses can be conveniently fabricated by our synergistic dual-mode optical lithography technique (Figure 1B). The theoretical and experimental characterization has shown that the multilevel phase SCL could achieve the focusing intensity of 7.2 times that of (Figure 1C) its counterpart for the traditional binary amplitude-type SCL. Our nanofabrication approach leads to the application possibility of the nonlinear optical lithography technique in the field of the fabrication of optical integrated circuits, and the demonstrated multilevel phase supercritical lens with high focusing efficiency and intensity contrast can greatly thrust the planar metalens forward in the extended range from scientific researches to practical applications.

Figure 1: Schematic of the sub-diffraction-limit focusing with high efficiency from multilevel phase supercritical lens. (A) Schematic of the binary amplitude-type lens (I), binary phase-type lens (II) and four-level phase-type lens (III) with the same diameter and the numerical aperture, and the corresponding far field transmitted light intensity distributions along the optical path. (B) Sideview of the binary amplitude SCL (I), the binary phase SCL (II), and the four-level phase SCL which is constructed by decomposing the binary phase pattern (II) into the gradient four-level phase pattern in lateral direction. (C) Comparison of the lateral size and the normalized intensity of the focal spots between different types of SCL. Line profile distribution of the focal spot intensity of three types SCL. The FWHMs of $0.42 \lambda/\text{NA}$, $0.42 \lambda/\text{NA}$ and $0.40 \lambda/\text{NA}$ are found for the binary amplitude SCL, the binary phase SCL, and the four-level phase SCL, as shown with the blue, green and red lines, respectively. And the four-level phase SCLs can realize increased focal spot intensities of $1.8\times$ and $7.2\times$ that of its binary phase-type and binary amplitude-type counterparts, respectively.

2 Synergistical dual-mode optical lithography

For the experimental realization of the discretized multilevel phase SCL, completely controlling the lateral and longitudinal feature size of each lens part is a necessary prerequisite. The position and the width of each concentric belt in the planar SCL determine the spatial frequency components that constructively interfere with each other in the focal region. The thickness of the dielectric belts corresponds to the phase accumulation depth as the light beam passes through. Two photon polymerization-based maskless optical beam lithography (OBL) technique can elaborately provide sub-100-nm

resolution in the far field with low cost. The movement trajectory in the fabrication could be controlled either by the galvo mirror dithering or by the piezostage scanning. For the traditional direct laser writing technique, the line as being achieved is subject to elliptical cross-sections, as the focal depth of the focused Gaussian beam is commonly found to be twice or more as large as the lateral size in the beam waist, which is well known as the fabrication elongation in the direct laser writing (DLW) process [32, 35, 36]. It intrinsically prevents us from achieving optical devices with large degrees of flexibility and freedom. Although several techniques have been proposed to conquer this restriction in the past years [37–40], thoroughly solving this problem has not been reported yet.

The essential cause for the elongation effect is that both of the lateral resolution and the longitudinal resolution of the OBL technique are indistinguishably determined by the same set of fabrication parameters, such as the laser intensity distribution in the focal region and the scanning speed of the optical laser beam. One effective way to overcome this obstacle is to separate the influence on the lateral resolution from that on the longitudinal resolution. In order to achieve this target, we developed a dual-mode optical lithography technique, as shown schematically in Figure 2A. The trajectory of the fabrication is synergistically controlled through the galvo mirror dithering and the nanostage scanning. During the fabrication, while the outline contours of the structure are sketched by scanning the focal spot with the nanostage, the internal fine features along the horizontal and the longitudinal directions are traced out by the means of the galvo mirror dithering and the laser intensity in the focal region, separately. Such synergistic dual mode optical lithography technique provides us with an ideal solution to realize the multilevel phase SCL which consists of a series of concentric belts with precisely designed thicknesses and widths. We can scan the nanostage following the trajectory equation of a circle with the radius of R_n , and in the meantime, manipulate the galvo mirror dithering with a radius of r_n following the circling path simultaneously. When the dithering rate of the galvo mirrors matches with the scanning speed of nanostage, a string of circles can assemble to a belt with the width determined by the galvo mirror dithering radius of r_n , as shown in the inset of Figure 2A. Therefore, we can adjust the width of each belt by only changing the radius of the galvo mirror dithering. Meanwhile, the thickness of the belt is explicitly maintained by fixing the power of the fabrication laser beam and the scanning speed of the nanostage.

For building up the fabrication system, a femto-second (fs) laser beam with 800 nm wavelength is used in our system to initiate the two photons polymerization (TPP) process. (See Figure S1 in Supporting Information for more details.) After systematic investigation, the power of the fabrication laser beam of 36 mW and the nanostage scanning speed of 40 $\mu\text{m/s}$ are found as the optimized parameters to match with the belt thickness for accumulation phase modulation depth of 0.5π for light wavelength at 633 nm, as described in Figure S2–S5 in the supporting materials. When the dithering radius of the galvo mirrors varies from zero to 0.55 μm , the corresponding belt widths are tuned from 180 nm to 1.3 μm , as shown in Figure 2B and Figure S6. As the power of the fs laser beam and the movement speed of the piezo-stage are fixed at the same condition for all cases, the fabricated belts can keep an invariable thickness at around 350 nm close to the theoretical value of 316 nm, as represented by pink cubes shown in Figure 2C. The relationship between the width of the belts and the trajectory radius of the galvo mirror dithering is approximately linearly dependent, as shown by the green dotted line in Figure 2B, which demonstrates that the belt width can fit perfectly between the design and the fabrication while the thickness is stably maintained. The scanning electron microscopy (SEM) images for four representative belt widths of $W_o = 180 \pm 30$ nm, $W_m = 435 \pm 30$ nm, $W_n = 770 \pm 50$ nm and $W_b = 1200 \pm 50$ nm are shown in sub-panels I–IV in Figure 2B, respectively, which correspond to different dithering radiuses of the galvo mirrors. As long as a single layer of belt precisely adds phase accumulation of 0.5π vertically, we can readily realize other values of cumulative phase addition with a step of 0.5π through layer by layer stacking. (The details for the thickness controlling and the corresponding phase modulation can be found in the Figure S7 in Supporting Information.)

3 Binary phase supercritical lens

To validate the capability of flexibly controlling the width and the thickness of the belts, a binary phase SCL, which is composed of a series of concentric belts with the width of 1.2 μm and the thickness of 633 nm to satisfy the particular phase modulation depth of π , is experimentally realized. The synergistic dual-mode optical lithography approach proposed here can be easily used to fabricate the binary phase SCL. As depicted in Figure 2C, the belt width of 1.2 μm can be simply achieved by a 0.5 μm dithering radius

Figure 2: Synergistic dual-mode optical lithography technique.

(A) Schematic for the fabrication system. The SCL is patterned by the synergistic movement of the piezo stage and a pair of scanning galvo mirrors. As shown in the inset figure, the structure is sketched out by the trajectory of the piezo stage, and the internal structural feature is controlled by the oscillation of the galvo mirrors. R_n is the radius of the n_{th} belt of the SCL following the trajectory of piezo stage movement, and r_n is the oscillating radius of the galvo mirrors which governs the width of the n_{th} belt (W), where the $R_{n,in}$ and the $R_{n,out}$ stand for the inner radius and the outer radius of the belt, respectively. (B) Experimentally measured feature sizes of the fabricated belts versus the oscillating radii of the galvo mirrors. The widths of the belts can be precisely controlled by the r_n . Sub-panels of I–IV show the SEM images of the belts fabricated with $r_n = 0$ nm, 80 nm, 320 nm, and 500 nm, corresponding to $W_0 = 180 \pm 30$ nm, $W_m = 435 \pm 30$ nm, $W_n = 770 \pm 50$ nm, $W_b = 1200 \pm 50$ nm, respectively. As the power of the fs laser beam and the piezo-stage movement speed are fixed at 30 mW and 40 $\mu\text{m/s}$ for all cases, respectively, the thickness of the belts as fabricated are kept in an invariable value of around 350 nm, as represented by pink cubes shown in (C). Scale bars: 1.2 μm .

for the galvo mirrors. The phase modulation depth of π can be achieved by stacking two layers of belts for a total thickness of 633 nm. The SEM images of the SCL as fabricated and the related characterization of sub-diffraction focusing properties are presented in Figure 3. The measured belt width shown in Figure 3A, B is 1200 ± 50 nm, which is in accordance with the designed value. As shown in the optical characterization results, the measured

intensity distribution at the focal plane of $z = 63.3$ μm is in good agreement with the theoretical plots (Figure 3C, D). The lateral size of focal spot in experimental is in sub-diffraction-limit scale with the full width at half maximum (FWHM) of $(0.45 \pm 0.03) \lambda/\text{NA}$, close to the simulated results of $0.42 \lambda/\text{NA}$ (Figure 3E), which suggests the dual mode optical lithography technique is of excellent fabrication accuracy.

Figure 3: Binary phase-type SCL.

(A, B) The SEM images of the binary phase-type SCL as fabricated with its related sectional zoom-in view. The binary phase SCL with a diameter of $197\ \mu\text{m}$ is consisted of 40 concentric belts with the width equaling to $1.2\ \mu\text{m}$. Scale bars for (A) and (B) are $20\ \mu\text{m}$ and $3.6\ \mu\text{m}$. Through precisely controlling the oscillating radius of the galvo mirrors, the belt width (W) of the SCL could be controlled at $1200 \pm 50\ \text{nm}$. (C, D) Simulated and measured intensity distributions of the focal spot in the focal plane. Scale bar: $500\ \text{nm}$. (E) The intensity profiles of the focal spot along the dashed line, which shows sub-diffraction-limit focusing property and the agreement between the simulation data and the experimental results.

4 Multilevel phase supercritical lens

The most significant advantage of our technique is the ability to pattern a complicated structure with varied features sizes, which ideally satisfies the requirement of constructing discretized multilevel phase SCLs with different belt widths. To demonstrate reliable focusing efficiency improvement, the design of the four-level phase SCL shares the same structural parameters with the binary phase SCL as demonstrated in Figure 3, except that the phase modulation values of $0, \pi$ in binary configuration are replaced with $0, \pi/2, \pi, 3\pi/2$ for the configuration of four-level phase SCL. The detailed design is described in Figure S9. The width of each sub-belt is diverse for different supercells, when the binary phase SCL design is transformed for the four-level phase SCL, given distances between each belt in the binary phase SCL are non-equal, as the detailed parameters shown in Table S1 in the Supporting Information. The different sub-belt widths can be accomplished by different galvo mirror dithering radiuses, while different belt thicknesses which refer to the discretized four-level phase patterns can be achieved by multilayer stacking. The patterning process is shown in Figure 4A, and the white dotted lines in the figure indicate the boundary of each layer. The SEM image of an isolated supercell with four-level phase configuration is shown in Figure 4A (iv), where different colors correspond to different phase modulation depths.

Figure 4 (B, C) are the topview SEM images of a four-level phase SCL and its sectional zoom-in views.

The sideview SEM image presented in Figure S10 in the Supplementary Materials has clearly shown the four-step configuration with different thicknesses as designed. The P_n represents the period of each supercell. The simulated light intensity distribution and the corresponding measured values at the focal plane of 100λ ($\sim 63.3\ \mu\text{m}$) away from the SCL are depicted in Figure 4D, E. The intensity profile of the focal spot obtained along the dashed line across the center of focal spot is shown in Figure 4F, which shows a good fit between the simulation and the experimentally measured result. The FWHM of the focal spot for the experimentally measured value of $(0.41 \pm 0.015)\ \lambda/NA$ is in line with the simulated value of $0.40\ \lambda/NA$. The slight discrepancy may reside in unavoidable fabrication errors and inaccurate characterization.

Unlike the focusing performance of conventional Fresnel zone plate (FZP), which comes from in-phase construction interference of all zone belts at the desired focal region, the SCL inevitably introduces significant multiple secondary focusing to break the diffraction limit and obtain an unusual small hotspot. The multiple secondary focusing effect and the high-order diffraction property cause not only a wastage of illumination energy but also huge damage to the contrast of the intensity distribution on the focal plane. Differing from the binary phase SCL, the four-level phase SCL discussed involves more phase level values in the interference process, and thus, the multiple secondary focusing effect can be significantly suppressed. As shown in Figure 5A–D, both of the intensity distributions of the binary phase SCL and the four-level

Figure 4: Multi-level phase SCL.

(A) Schematic representation of the fabrication procedure of the four-level phase SCL supercell. The sub-belts with different thicknesses for phase modulation of $\pi/2$, π , and $3\pi/2$ are discretely divided into four layers in design, and then fabricated by optical lithography technique step by step (I–III). Top view of the four-level phase units is shown in (IV), in which different colors represent different thicknesses for implementing phase additions. Scale bar: $1.5\ \mu\text{m}$. (B, C) The SEM images of the four-level phase-type SCL and its sectional zoom-in view. Scale bars for (B) and (C) are $20\ \mu\text{m}$ and $2\ \mu\text{m}$, respectively. P_n represents the width of the n_{th} supercell. The sub-belts are created by equally dividing the P_n into four belts with the same width and gradually increasing phase modulation depth. (D, E) The simulation and the measured result of the focal spot intensity distribution in the focal plane. Scale bar: $500\ \text{nm}$. (F) Line profiles of the intensity distribution of the focal spot obtained in (D) and (E), which show sub-diffraction-limit focusing property and the agreement between of the simulation data and the experimental results.

phase SCL can achieve sub-diffraction-limited focusing at a distance of $100\ \lambda$ away from lens plane. However, the results have shown that the four-level phase SCL remarkably suppresses the high-order diffraction and the multiple secondary focusing, which bundles more light energy into the hotspot region and boosts the focal spot intensity much higher than its binary configuration counterpart. The intensity contrast of the focal spot can be evaluated by the energy concentration ratio inside the hotspots, as proposed by Ref. [41]. Figure 5E shows the simulated light intensity distributions and the corresponding experimentally measured results along the longitudinal direction for the binary amplitude SCL, the binary phase SCL and the four-level phase SCL, respectively. The simulated energy concentration ratio inside the hotspots is found to be 50.48% for the four-level phase SCL, which is 3 times that of its binary phase counterpart with the value of 18.39%. In experiments, the energy concentration ratios for the binary phase configuration and the four-level

phase configuration are 19.24% and 40.03%, respectively. The higher energy concentration ratio explicitly results in higher focusing efficiency and increased spot intensity under the same illumination condition. As shown in Figure 5F, the focusing intensity of the four-level phase SCL is 1.8 times and 7.2 times that of the binary phase SCL and binary amplitude SCL, respectively, which makes it more feasible in practical applications.

5 Conclusion and discussion

In summary, we proposed a synergistic dual-mode optical lithography approach for precisely fabricating multilevel phase SCL with high focusing efficiency. The feature size in the lateral directions and the longitudinal directions can be controlled independently, subverting the restrictions imposed by the elliptical cross-section of the optical

Figure 5: Comparisons between the binary amplitude/phase and the four-level phase-type SCL.

(A–D) Simulated (A, C) and experimentally measured (B, D) focal spot intensity distributions in the propagation plane (XOZ plane) for the binary phase-type (A, B) SCL and the four-level phase-type (C, D) SCL, respectively. (E) Line profiles of the transmitted light intensity distribution along the optical axis for three types of SCL. The curves plotted with three different colors represent the simulated intensity distribution along the longitudinal direction for the binary amplitude SCL (purple line), the binary phase SCL (blue line) and the four-level phase type SCL (pink line), respectively. The two triangle-dot diagrams represent the experimentally measured results of the binary phase SCL and the four-level phase type SCL, respectively. The related focusing quality of the binary amplitude SCL is only performed with simulation in the diagram for comparison. (F) Comparison of the focal spot intensities for the binary amplitude SCL, the binary phase SCL and the four-level phase-type SCL, which shows that multi-level phase SCL can achieve remarkably high intensity than that for the traditional binary amplitude-type SCL and the binary phase-type SCL.

intensity field of the focused Gaussian beam. Combining with the sub-diffraction-limit fabrication capability of two-photon polymerization direct laser writing, we successfully demonstrated a four-level phase SCL which exhibits much higher energy utilization efficiency and provides higher contrast for the sub-diffraction-limit focusing. The numerical and experimentally measured results show that the multilevel phase SCL can not only achieve a sub-diffraction-limit focal spot with lateral size of $0.40 \lambda/\text{NA}$ (0.67 times Airy unit), but also exhibit 7.2 times higher focusing intensity compared with its binary amplitude counterpart. Besides, the four-level phase SCL can significantly suppress the high-order diffraction and the

multi-secondary focusing effect, which increases energy concentration ratio inside the focusing hotspots by 3 times in comparison with the traditional binary configuration. It should be noted that, although merely manifests its ability in fabricating SCLs with cylindrical symmetry in this work, the synergistic optical lithography technique we proposed here is applicable for printing arbitrary structures without symmetry limitation. Our technique opens a route for fabricating unprecedented sub-diffraction-limit planar nanophotonic devices, and the demonstrated multilevel SCL can serve as an ideal way for practical applications such as label free super-resolution imaging, high density optical storage, and high precision optical manipulation.

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