Two-photon polymerization of microstructures by a non-diffraction multifoci pattern generated from a superposed Bessel beam

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In this Letter, superposed Bessel beams (SBBs) are realized by alternatively imprinting holograms of opposite-order Bessel beams along the radial direction on a spatial light modulator. The propagation invariance and non-rotation properties of SBBs are theoretically predicted and experimentally demonstrated. The focusing property of SBBs with a high numerical aperture (NA) objective is investigated with the Debye vectorial diffraction theory. Near the focal plane, a circularly distributed multiple foci pattern is achieved. The multiple foci generated from SBBs are adopted in a two-photon fabrication system, and micropattern fabrication by a single exposure is demonstrated. Facile fabrication of three-dimensional microstructures with SBBs is realized by dynamically controlling the number of focal spots, and the diameter and rotation of the focal pattern. © 2017 Optical Society of America

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The femtosecond two-photon polymerization (TPP) technique, due to its simplicity, down to sub-100 nanometer resolution [1], and intrinsic three-dimensional (3D) processing ability, is widely used for the high-precision fabrication of complex functional 3D micro- and nanodevices [2–4]. A conventional method is to move the focus of a Gaussian beam within materials to build micro- and nanostructures point by point [5]. To improve the micromachining efficiency and flexibility, microlens arrays [6], diffractive optical elements [7], and multibeam interference [8] are introduced for parallel processing. However, the positions of the foci are fixed by the optical design of these optical elements. It is a flexible and efficient method to fabricate complicated microstructures through spatial laser beam shaping by using a spatial light modulator (SLM) [9–12], which provides the possibility of designing beam shapes that are optimized for specific applications. Up to now, many kinds of shaped beams, including a multiple beam spot array [9], spatially shifted vortex beams [10], and double-helix beams [11] have been explored for the rapid fabrication of functional structures.

Bessel beams, among these engineered specific beams, have drawn broad attentions due to their interesting properties such as propagation invariance, “self-healing,” and the possession of orbital angular momentum. Bessel beams have been used in the area of optical trapping and manipulation [13], high-aspect-ratio nanochannel drilling [14], silicon and glass processing [15], and surface structuring [16]. It has become clear that the generation of specific Bessel beams can enable the parallel processing, the enhancement of the processing speed and flexibility, and the creation of the specially required focal pattern in microfabrication.

In this Letter, superposed Bessel beams (SBBs) are realized by using a SLM without any other optical elements. In contrast with other SBB generation methods [17,18], the optical system is much simpler, and the parameter controlling of the SBB is much more convenient. The propagation invariance and non-rotation of SBB is demonstrated. The focusing property of a SBB with a high NA objective is theoretically investigated with the Debye vectorial diffraction theory. The focused SBB is used for efficient TPP, and the experiment results confirm that a multifoci pattern can be fabricated by a single exposure. Furthermore, the flexible fabrication of 3D microstructures by dynamically controlling the multifoci pattern is realized. Our novel laser fabrication process enhances the efficiency and flexibility of microfabrication.

Bessel beams have an intensity profile that approximates Bessel functions, and high-order Bessel beams exhibit a non-diffraction dark core on the beam axis. Bessel beams can be generated by a SLM imprinted with holograms [19] that have the transmission function as below:

\[ T_n(r, \theta) = \exp(i n \theta) \exp(-i 2 \pi r / r_0), \]

where \( r \) and \( \theta \) are transverse and polar coordinates, \( r_0 \) is an adjustable constant parameter, and \( n \) denotes the \( n \)-th-order.
Bessel beam. Since the display of the SLM is constituted by pixels, we rewrite \( r = \left( i^2 + j^2 \right)^{1/2} d \), where \( i \) and \( j \) are integers indicating each pixel of the SLM, and \( d \) is the pixel pitch. Figures 1(a) and 1(b) show the holograms corresponding to the 3rd- and -3rd-Bessel beams.

The superposition of Bessel beams can be realized by two methods. The first method is simply adding the transmission function of \( T_\pm(r, \theta) \) and \( T_\mp(r, \theta) \). The second method is to imprint the holograms of the \( \pm n \)-th-order Bessel beam alternately along the radial direction, which is chosen in this Letter to ensure that the generated patterns do not rotate as they propagate. The transmission function for superposed \( \pm n \)-th-order Bessel beam is

\[
T_{\pm n}(r, \theta) = \exp(-i2\pi r / r_0) \exp(i(-1)^{n/2} n \theta) \exp(i\phi).
\]  

These holograms are composed of concentric rings with continuous azimuth modulation from 0 to \( 2\pi \), and \( \Delta \) is the width of each ring. The factor \( \phi \) determines the rotating angle of the holograms. The handedness of the azimuth modulation is determined by the radial coordinate \( r \), and every two neighboring rings have opposite modulation directions (handedness). Figure 1(c) is the result superposed by \( T_3 \) and \( T_{-3} \).

The optical field of SBBs behind the SLM can be obtained according to Fresnel diffraction integration:

\[
U(x, y, z) = \frac{\exp(i k z)}{i z} \int_{x_0, y_0} U(x_0, y_0) T_{\pm n}(x_0, y_0) \times \exp \left[ \frac{i k (x-x_0)^2 + (y-y_0)^2}{2z} \right] \, dx_0 \, dy_0.
\]  

\[ \text{(3)} \]

where \((x, y, z)\) is the Cartesian coordinate with the origin at the center of the SLM, \((x_0, y_0)\) is the coordinate on the SLM, \( U(x_0, y_0) = \exp(-i(x_0^2 + y_0^2) / w_0^2) \) is the electrical field of the incident laser beam, and \( w_0 \) is the waist radius. Theoretical calculation indicates that the intensity profile of a pure Bessel beam is a circular beam pattern [inset of Figs. 1(a) and 1(b)]. For SBBs, a laser beam is divided equally along the circumferential direction, and a petal-like pattern is produced [inset of Fig. 1(c)]. The petal number is decided by the order of SBBs. For a superposition of \( n \) \( \pm \) \( n \), the final petal number is \( 2n \). Using the \( 3 \pm (3) \) superposition as an example, generated SBB, as viewed in the far field, is shown in Figs. 1(d)–1(i). Figure 1(d) is the theoretically calculated intensity distribution along the beam propagation direction. The intensity profiles in the transverse direction, at distances of 1.2, 1.6, 2.0, and 2.4 m from the SLM, are also calculated [Figs. 1(e)–1(h)] and experimentally measured [Figs. 1(i)–1(l)]. The theoretical prediction is in excellent agreement with the experimental measurement. It is calculated and observed that the intensity profile remains almost unchanged in a long distance of 2.0 m [Figs. 1(d)–1(l)], which demonstrates the propagation-invariant property of SBBs.

For TPP or other microfabrication processes, the SBBs need to be tightly focused with a high NA objective. The focusing property of SBBs is investigated with the Debye vectorial diffraction theory [9,11]. The field distribution at the focal region is derived according to the Debye vectorial diffraction integral:

\[
E_2(x_2, y_2, z_2) = -\frac{iC}{\lambda} \int_0^{\alpha} \int_0^{2\pi} \sin \theta_2 \sqrt{\cos \theta_2} P(\theta_2, \varphi_2)
\times \exp[i k n_0 (x_2 \cos \theta_2 + x_2 \sin \theta_2 \cos \varphi_2
+ y_2 \sin \theta_2 \sin \varphi_2)] d\theta_2 d\varphi_2,
\]  

\[ \text{(4)} \]

where \((x_2, y_2, z_2)\) is the Cartesian coordinate with the origin at the center of the exit pupil of the objective; \( C \) is a constant, \( \lambda \) is 800 nm, the wavelength of incident light; \( n_0 \) is 1.518, the refractive index of immersion medium; \( \alpha \) is the maximum focusing angle of the objective lens and can be calculated according to the formula \( \alpha = \arcsin(NA / n_0) \); \( NA \) is the numerical aperture of the objective lens, and \( NA = 1.35 \) in this Letter; \( \theta_2 \) represents the focusing angle of objective lens, and \( \varphi_2 \) is the azimuthal angle of object plane. \( P(\theta_2, \varphi_2) \) indicates the polarization state of the EM field in the focal region. For incidence with linear polarization at X direction, \( P(\theta_2, \varphi_2) \) can be rewritten as

\[
P(\theta_2, \varphi_2) = \left[ 1 + (\cos \theta_2 - 1) \cos \varphi_2 \right] i + \left[ (\cos \theta_2 - 1) \cos \varphi_2 \sin \varphi_2 \right] j - (\sin \theta_2 \cos \varphi_2) k.
\]  

\[ \text{(5)} \]

According to Eqs. (4) and (5), the intensity distributions at the focal region of \( 3 \pm (-3) \) [Figs. 2(a) and 2(b)] and \( 10 \pm (-10) \) [Figs. 2(c) and 2(d)] SBBs are calculated. The generated intensity distributions at the focal plane are 6 and 20 foci uniformly arranged in a circular manner. The intensity distribution in the XZ direction shows [Figs. 2(b) and 2(d)] that most of laser energy distributes near the focal plane with a depth of focus of 2.8 \( \mu \)m. However, a bit of laser energy distributes about 10 \( \mu \)m above the focal plane. We find that this part of intensity distribution is negligible when \( n > 10 \). As \( n \) decreases, this part of intensity distribution becomes more and more pronounced.
The diffraction-limited features of the foci in the focal plane [Fig. 2(e)] and at a location of 15 μm away from the focal plane [Fig. 2(f)] are checked by plotting the intensity distribution along the X direction (marked in a white line). The full widths at half-maximum of the focal spots are 0.44 and 0.54 μm, and the radii of the intensity pattern are 11.5 μm and 11.9 μm, respectively, for 3 + (−3) and 10 + (−10) SBBs. In contrast, the intensity distribution at a location of 15 μm from the focal plane has a lower intensity and a smaller radius.

To experimentally verify the theoretical prediction, a microfabrication system containing phase modulation is implemented, as schematically illustrated in Fig. 3(a). A collimated linearly polarized femtosecond laser beam with a wavelength of 800 nm (Chameleon Ultra, Coherent GmbH) illuminates on an a SLM (Pluto NIR II, Holoeye) with 1920 × 1080 pixels of pitch 8 μm. Calculated holograms (inset) are encoded on the SLM, and generated SBBs are relayed to the back aperture of a high-NA objective lens (60×, SLR, and generated SBBs are relayed to the back aperture of a high-NA objective lens (60×, Olympus). A uniform circular foci pattern is generated at the focal plane of the objective (inset). A multiple spot pattern is prototyped by TPP with a single exposure of a pure 10th-order Bessel beam, 1 + (−1), 2 + (−2), 3 + (−3), 4 + (−4) and 5 + (−5) SBBs, respectively. The insets are intensity profile observed at the focal plane of objective. All the circular multipole patterns are fabricated by a single exposure, with an exposure time of 100 ms at laser power of 90 mW. The diameters of the circular patterns are ~11.5 μm, which show exact agreement with the simulation results [Fig. 2(e)]. The fabricated microstructures are symmetrical based on the symmetrical pattern of the superposition of Bessel beams. It should be noted that the laser energy distributes about 10 μm above the focal plane, as illustrated in Figs. 2(b) and 2(d), and two-photon polymerization may arise when the fabrication power exceeds a threshold.

In addition, a remarkable feature of this approach is that the number of focal spots, the diameter, and the rotating angle of the multipurpose pattern can be flexibly manipulated by controlling the imprinted holograms without any change of the optical path. The number of focal spots is exclusively determined by the order of SBBs, n. The diameter of multiple spot patterns is decided by both the phase factor r₀ and n, according to a theoretical calculation. r₀ plays a dominant role in controlling the diameter, while n affects the diameter slightly. In our experiments, when r₀ varies from 420 to 9600 μm, the diameter of the multipurpose pattern varies from 12.3 to 0.86 μm and fits well with the theoretical predictions of 12.7 and 1.1 μm. The diameter can be controlled in a much larger range if the limitation of the superposition of SBBs.

Fig. 3. Single exposure of multiple spot patterns by focused SBBs. (a) Illustration of the microfabrication system. The arrow indicates the polarization direction of the laser beam. The insets are holograms imprinted to a SLM and intensity profile measured at the focal plane of the objective. (b)–(g) SEM images of multipole patterns fabricated by the single exposure of a pure 10th-order Bessel beam, 1 + (−1), 2 + (−2), 3 + (−3), 4 + (−4) and 5 + (−5) SBBs, respectively. The insets are intensity profile captured with a CCD. The scale bars are 3 μm.

Fig. 2. Intensity distributions of focused SBBs with a high NA objective. (a) and (b) are the intensity distributions at XY and XZ planes of focused 3 + (−3) SBB. (c) and (d) are the intensity distributions of focused 10 + (−10) SBB. (e) and (f) are intensity cross-sectional plots at the focal plane and 15 μm away from the focal plane. The blue line corresponds to the 3 + (−3) SBB, and the brown line corresponds to the 10 + (−10) SBB.
and generate SBBs by designing the holograms. When the holograms are dynamically switched on the SLM, the generated SBB and its focal pattern composed of multisots will also achieve dynamic motion synchronously. When the diameter and rotating angle of the multifoci pattern change according to a certain rule simultaneously, some special focal trace can be realized. For instance, Fig. 4(a) shows a designed trace composed of six focal spots, of which the diameter decreases from $R_1$ to $R_2$ and, finally, to $R_3$. At the same time, the focal spots rotate clockwise by an angle of $\gamma$. Finally, the focal spots move along the black arrow forming a flower-like pattern. In the TPP processing, 52 holograms are dynamically displayed on a SLM with a refresh rate of 10 Hz. The laser power is controlled to be 50 mW. A flower-like microstructure with six petals [Fig. 4(b)] is fabricated. The number of foci can also be controlled [Fig. 4(c)]. When $r_0$ is changed linearly in Figs. 4(b)−4(c), the diameter of the multifoci decreases faster as the foci close to the center. Thus, the outer side is exposed for a longer period and, finally, exhibits a higher structure than the inner side. Furthermore, by moving the foci pattern along the $z$ direction, together with the dynamic control of the focal pattern, the microtubes with slits, which have important applications in the research into the flexible and fast TPP fabrication technique, and the fabricated microtubes with slits are suitable for the number of important applications, such as artificial blood vessels and biomechanical research of blood cells.

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![Fig. 4. 3D microstructures fabricated by dynamically controlling the focal pattern. (a) Illustration of the trace of multiple foci for flower-like microstructure fabrication. (b) and (c) are SEM images of flower-like microstructures with six petals and 10 petals, respectively. The scale bars are 5 μm. (d) Illustration of the fabrication process of a microtube with slits. (e) and (f) are SEM images of microtubes with slits. The scale bars are 10 μm.](image)