Laser direct writing of volume modified Fresnel zone plates

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Volume zone plates consisting of a number of Fresnel zone plate layers are designed and fabricated inside a fused silica by the femtosecond laser direct writing method. Light diffracted from different layers of a volume zone plate constructively interfere at the focal spot to increase diffraction efficiency. The technique is experimentally verified to be effective for both low-numerical-aperture (NA) and high-NA zone plates, resulting in a significant increase in overall diffraction efficiency. The spatial resolution of a high NA volume zone plate is 500 nm. © 2007 Optical Society of America

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1. INTRODUCTION

Recently, the use of femtosecond laser direct writing to produce optical devices inside transparent materials has been growing considerably. Tightly focused femtosecond laser pulses can induce nonlinear absorption within the focal volume and permanently modify the index of refraction of the material [1,2]. Although the mechanism responsible for refractive index increase by ultrashort laser pulses is not fully understood, researchers have fabricated various optical devices inside transparent materials with this direct writing method. These devices include waveguides [2,3], gratings [4], and diffractive optical elements (DOEs) [5–7].

In recent years, the development of fabrication techniques for DOEs has accelerated because DOEs can perform many optical functions such as lenses, gratings, optical signal processors, beam splitters, and wavelength (de)multiplexers. Fabrication of DOEs requires high-resolution techniques such as lithography. Femtosecond laser direct writing offers advantages as compared to other techniques such as volume writing, precision, speed, simplicity, and flexibility. In particular, this method of fabrication can be applied for 3D or volume DOEs without difficulties in mask changing or mask alignment.

A simple form of DOE is the Fresnel zone plate. The most attractive features of Fresnel zone plates are their compactness and capability for high resolution while maintaining high efficiency. In the absence of losses and absorptions, an ideal diffractive zone plate can offer 100% diffraction efficiency provided that the zone plate correctly modulates phase. In practice, a quantized zone plate is used as an approximation of the perfect diffractive lens and its efficiency ranges from 10% to almost 100% depending on the number of quantization levels. A higher number of phase quantization levels provides higher efficiency, yet increases complication in fabrica-

tion. A two-level Fresnel zone plate having a focal length f is constructed with a series of concentric zones whose radii are defined by [8]

$$r_n = \sqrt{n\lambda f + \left(\frac{n\lambda}{2}\right)^2} \approx \sqrt{n\lambda f},$$
 (1)

where the integer n indicates the nth Fresnel zone and λ is the operating wavelength. The working principle of the Fresnel zone plate relies on the fact that the diffraction of light from alternating zones interferes constructively at the designed focal point. It is required that the alternating zones have different transmission properties (i.e., refractive index), and the incident light is monochromatic.

Compared to other fabrication methods, the laser direct writing technique has the advantage of ease of fabrication of such a high-efficiency zone plate, eliminating complications in multistep processes. Studies of direct laser fabrication of Fresnel zone plates inside fused silica have been reported for both amplitude-type zone plates [5] by utilizing scattering damage and phase-type [6] zone plates by refractive index change induced by femtosecond laser pulses. However, all the previously studied zone plates suffered from low diffraction efficiency because of effects such as scattering from damaged regions, phase shift errors due to nonuniform index change inside fused silica, and planar fabrication. Attempts to fabricate multilevel phase zone plates by stacking of the index change filaments were also reported, yet had difficulty eliminating the phase shift errors due to fabrication [7]. The drawback of femtosecond laser direct writing is the low refractive index contrast, which is insufficient to create a single high-efficiency phase zone plate. Recently, we presented a new approach of fabricating a number of Fresnel zone plates or modified Fresnel zone plates within a volume, which we called volume zone plates, to achieve high diffraction efficiency [9]. By using a series of zone plates, efficiency is improved significantly, and this method is flexible with different laser writing conditions regardless of the individual diffraction efficiency. In that work, we fabricated volume zone plates and showed significant increases in diffraction efficiency for both amplitude-type and phase-type [9]. The highest efficiency of 59.1% was reported for a phase volume zone plate. According to the design, each volume Fresnel zone plate operates as a single diffractive optical element with a much higher diffraction efficiency than a single zone plate.

In this paper, we focus on phase-type volume zone plates because of their higher efficiency, and our method is further improved by controlling the filament length for uniform zone plate thicknesses and also considering the effects of aberrations. We also characterized the refractive index changes in the material using different fabrication parameters to correctly determine the phase shift. In addition, fabrication of high numerical aperture (NA) volume zone plates is investigated because they are highly demanding in microimaging applications. The results indicate that our volume zone plate method is highly effective for laser direct writing, improving the diffraction efficiencies up to 71.5% and 7.1% for low NA and high NA zone plates, respectively. Our high NA volume zone plate has a resolving resolution on the order of 0.5 μ m. The rest of the paper is organized as follows. In Section 2, we describe the concept and design of the volume modified Fresnel zone plate. The fabrication processes are described in Section 3 in which low NA and high NA Fresnel zone plates are discussed separately because they rely on different fabrication approaches. Discussion of results and conclusions are given in Sections 4 and 5, respectively.

2. DESIGN OF A VOLUME FRESNEL ZONE PLATE

The concept of a volume Fresnel zone plate is based on the principle that diffracted light from many Fresnel zone plates can be effectively coupled together to enhance the intensity at the focal spot. An incident plane wave was considered for our design. A volume Fresnel zone plate consists of a number of layers of modified zone plates centered on the same optical axis. According to the concept, a volume modified zone plate should satisfy two conditions: focal point matching and phase matching. First, each Fresnel zone plate inside a volume has to be designed specifically according to its relative location so that all the zone plates focus light exactly at the same focal point. In addition, the diffracted light from all the zone plates must be "in phase," while the phase at the focal point can be chosen arbitrarily. In other words, the phase shifts produced by every zone plate are the same at the focal point so that the diffracted light from all the zone plates constructively interferes at the focal point.

Equation (1) is valid only when a Fresnel zone plate diffracts light in a medium of constant index of refraction. For our case, the volume zone plate operated inside a glass substrate whereas the focal point is in air. Therefore, we also took into account the phase shift by the refractive index of the glass and the refraction of light at the air-glass interface. The phase shift due to wave propa-

gation in a homogeneous medium can be calculated by [10] $\phi = 2\pi nt/\lambda$, where n is the index of refraction of the medium, and t is the propagation distance. In Fig. 1, the volume zone plate contains two layers of modified Fresnel zone plates and optical path length in air and glass are shown

In the design process, the number of layers in a volume and the spacing between zone plates need to be initially chosen. The spacing between zones is arbitrary but must be greater than the thickness of a zone plate to avoid overlapping of zone plates. Then, the radii of zone plates are generated by applying Eq. (1) with adjusted focal length, f_i , according to the axial distance from the focal point. The total optical path length was calculated by adding the traveling distance in air and glass, considering Snell's angle of refraction. Iteratively, the radii of the zones in each Fresnel zone plate are slightly adjusted to satisfy the phase matching requirement. For example, two zone boundaries (r_1 and r_2), shown in Fig. 1, are adjusted during the design by using Eqs. (2) and (3) below, such that their phase remainders are equal (b1=b2) at the end of the numerical iterations:

$$\phi_{1} = \phi_{air,1} + \phi_{glass,1} = \frac{2\pi}{\lambda} (t_{air,1} + n_{glass,1} t_{glass,1}) = 2\pi m_{1} + b_{1}, \eqno(2)$$

$$\phi_2 = \phi_{air,2} + \phi_{glass,2} = \frac{2\pi}{\lambda} (t_{air,2} + n_{glass,2} t_{glass,2}) = 2\pi m_2 + b_2. \tag{3}$$

Once all the modified zones have the same phase shift at the focal point, the volume modified Fresnel zone plate reaches the highest diffraction efficiency.

For ease of fabrication with minimum phase errors, another modification of the Fresnel zone plate is to fabricate the central rings only instead of full zones [9]. In this modification, each complete zonewidth is replaced by a single central ring in the middle of the zone, as shown in Fig. 2. With this approach, several advantages are achieved. Implementation time is greatly reduced. Since the central circle of each ring corresponds to the exact phase desired, the method is robust against small imple-

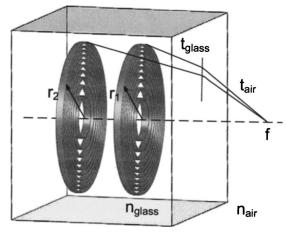


Fig. 1. Geometry used in phase computation for volume modified zone plate.

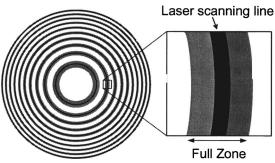


Fig. 2. (a) "Full" Fresnel zone plate showing central rings in the middle of each zone. (b) Modified zone plate with central rings (thickness determined by laser writing).

mentation errors [11]. Hence, the advantages of central rings replacing full zones are especially amplified in the implementation of volume zone plates. In addition, the full zone is not suitable for line by line scanning because the refractive index change is not uniform within the irradiated region such that it is difficult to create a uniform index change across the full zone. Although the individual diffraction efficiency of the central-ring zone plate decreases, the overall efficiency of the volume zone plate will increase because the central rings minimize the interfering effect of different zone plates [9]. The full zone is not suitable for volume diffraction because the strong interaction of each zone plate causes light to diffract multiple times and focus at different spots.

According to the above concept and modifications, the volume zone plate should coherently focus light at the single spot with minimum phase mismatching. By assuming each zone plate has low efficiency, the light diffracted from each modified Fresnel zone plate could be considered to be a small percentage of the total beam. The rest of the main beam transmits and is diffracted by the next layers. As the number of layers increases, the coherent interaction between the zone plates allows the diffraction efficiency to increase. Nevertheless, when the number of layers increases beyond a certain number, other effects such as phase shift error and fabrication error could accumulate and eventually diminish the diffraction efficiency of the volume zone plate.

3. EXPERIMENTS

In the experiments, a fused silica sample optically polished on all surfaces was fixed on a computer-controlled x-y-z air bearing stage that has a translational accuracy of 200 nm. The sample was irradiated by 90 fs pulses delivered by a Ti:sapphire amplified laser system at a central wavelength of 800 nm and 1 kHz repetition rate. The laser beam was attenuated before being focused inside the bulk of fused silica. An electronic shutter was connected to the computer controller, allowing the laser exposure and the stage movement to be synchronized. The fabrication process was monitored in real time via a CCD camera imaging on a TV screen.

In femtosecond laser direct writing, both the change in index of refraction and the shape of the filament where change of index of refraction occurs are sensitive to laser processing parameters such as laser frequency, pulse en-

ergy, scanning speed, focusing conditions, and focusing depth from the surface. For volume Fresnel zone plate fabrication, the processing parameters need to be adjusted when fabricating different layers because the focusing depth has influence on the change in the index of refraction caused by spherical aberration. The confocal distance is a function of self-focusing and spherical aberration. As reported by Sun et al. [12], the spherical aberration induced by air-glass interface extends the focal region toward the laser propagating direction. For deep focusing, this interface spherical aberration significantly increases the length and distorts the shape of the filament induced by femtosecond laser pulses. At the same laser writing condition, we experimentally found that the length of region with index change was relatively longer at the deeper position from the surface depending on the focusing condition. For example, using a $5 \times$ objective lens (NA=0.15), the filament length was 100 μ m at 0.5 mm below the surface compared to the filament length of $250 \,\mu\mathrm{m}$ at $2 \,\mathrm{mm}$ below the surface. This discrepancy in length was undesirable for the volume zone plate because we need consistent thickness of each zone plate to determine the influence of the number of layers used in the volume zone plate. To obtain a constant filament length for all layers, we applied a different laser energy for a different depth of fabrication. The laser energy and writing conditions required for a specific depth were experimentally predetermined for the above reasons.

In what follows, we will first describe characterization of the change in index of refraction caused by different laser parameters. Since low and high NA zone plates are fabricated using different methods, they will be described separately in Subsections 3.A and 3.B.

A. Measurement of Refractive Index Change

The $5\times$ and $100\times$ objective lenses were used to fabricate low and high NA volume zone plates, respectively. An estimation of refractive index changes due to femtosecond pulses focused by $5\times$ and $100\times$ lenses were necessary to correctly predict the phase shift by fabricated volume zone plates. Usually, the refractive index change due to femtosecond laser pulses is not uniform along the filament length, and it is difficult to measure such a small index change directly. However, the amount of refractive index change can be estimated by fabrication of gratings and measuring their diffraction efficiencies. Based on a Kogelnik's coupled wave theory [13], the index change can be estimated by

$$\Delta n = \frac{\lambda \cos \theta_B}{\pi t} \sin^{-1} \sqrt{n}, \qquad (4)$$

where θ_B is the Bragg incident angle, t is the thickness of the grating (filament length), and η is the first-order diffraction efficiency. According to Solymar and Crook [14], the coupled wave theory by Kogelnik offers good accuracy when dealing with thick grating (Q>10). In our case, the low NA volume zone plates were fabricated with much longer index filament than that of the high NA volume zone plates. The index filament lengths were 120 and 8 μ m for low and high NA, respectively. Therefore, the corresponding fabricated gratings give the value for Q

=65 for low NA volume zone plates and Q=11 for high NA volume zone plates. The grating pitches were 5 and 2 μ m for respective low and high NA volume zone plates, and the efficiencies of gratings were measured with He–Ne laser (632.8 nm). As a result, it is valid to estimate the refractive index increase by using the coupled wave theory.

We fabricated phase gratings inside fused silica using the same conditions as with the volume zone plates. The diffraction efficiencies of gratings were measured at the He–Ne laser wavelength (632.8 nm). The estimation of the refractive index increases were 0.16×10^{-3} for the laser processing conditions used for fabricating low NA volume zone plates and 1.8×10^{-3} for the laser processing conditions used for fabricating high NA volume zone plates.

B. Low-Numerical-Aperture Volume Zone Plates

Phase-type volume Fresnel zone plates with a focal length of 20 mm and a diameter of 0.8 mm (NA=0.02) were fabricated. Each layer of Fresnel zone plate consisted of 20 zones (10 central rings). First, we characterized a single layer phase zone plate and found that the pulse energy of $10-15 \mu J$ and $10 \mu m/s$ laser scanning speed were the optimum conditions for generating a phase zone plate when using a $5\times$ objective lens (NA=0.15). At 0.5 mm below the surface, the change of the index of refraction caused by each pulse was elongated over a length, which is determined by the Rayleigh range, resulting in a 120 μ m long filament of refractive index change. The filament lengths were predetermined for the specific pulse energies and depths of fabrication from the silica surface. Single layer zone plates were fabricated using central rings instead of "full zones" as discussed previously to reduce the fabrication time. The width of each ring depended on laser writing conditions: focusing lens and depth of fabrication. Using a 5× objective lens, the linewidth of the refractive index change was $\sim 3 \, \mu \text{m}$. With this approach, the fabrication time of a modified phase zone plate was 40 min.

According to our estimation of index increase, the phase shift of He–Ne laser passing through a single layer zone plate was 0.08π . Thus, the combination of refractive index change and the filament length of single layer zone plate were not enough to achieve a π phase shift, which is required for a phase reversal zone plate to achieve high diffraction efficiency. Further increase in phase shift can be obtained by either increasing the refractive index change or extending the filament length. Here, we use the volume zone plate method as an alternative to increase the diffraction efficiency.

The volume zone plates were fabricated layer by layer starting with the deepest layer from the surface facing the laser. Each layer was carefully placed according to the design equations to assure that all the layers would couple light together at the focus with constructive interference. The deeper layers were fabricated with less energy to maintain a constant thickness of each zone plate. The separation distance between the layers was approximately 200 μm , namely, about twice the thickness of each zone plate, and the zone plate closest to the surface was fabricated at 500 μm beneath the silica surface. The low

NA volume zone plates containing up to ten layers of Fresnel zone plates were fabricated to determine their focusing performance.

C. High-Numerical-Aperture Volume Zone Plates

We also fabricated high NA volume zone plates that can diffract light at a large converging angle at a short focal length. A Fresnel zone plate with high NA offers a highresolution focal spot, and it has potential in various optical applications. For such a short focal length zone plate, the Fresnel zones (rings) are densely spaced, and therefore high-resolution fabrication is needed. The closest separation distance between the adjacent zones becomes smaller as the diameter of the zone plate increases according to Eq. (1). Theoretically, the resolution of a high NA Fresnel zone plate is determined by the smallest zone width that can be produced by the fabrication method. The resolution limit is given by [15] $d=0.61\lambda/NA$ $\approx 1.22 \Delta r_N$ where Δr_N is the width of the outermost zone. In addition, aberrations such as spherical aberration become noticeable with the high NA Fresnel zone plate when the number of zones (transparent and opaque) are larger than $N=\sqrt{2f/\lambda}$ [15]. Therefore, our high NA zone plates could be affected by spherical aberration.

The fabrication parameters were determined experimentally to achieve the smallest linewidth of refractive index change inside fused silica. This linewidth determined our limit in fabrication resolution. The linewidth of $\sim 1 \,\mu \text{m}$ could be produced using a 100× objective lens (NA=0.80), a laser scanning speed of 10 μ m/s and a pulse energy of $0.1 \mu J$. First, we fabricated single layer Fresnel zone plates having NA ranging from 0.66 to 0.91 and a focal length of $60-130 \mu m$. The diameters of these high NA zone plates were approximately $100 \mu m$. The designs of all the high NA zone plates were limited by 1 µm minimum zone separation distance at the outermost zones. Similar to the low NA case, we fabricated zone plates with central-ring zones instead of full zones. Central zones were also more crucial in this case to minimize phase errors because the zones were much narrower in high NA zone plates, and writing a full zone with an exact width was extremely difficult. Central-ring high NA zone plates contained 50-90 rings depending on their designed NA. The fabrication time for each high NA zone plate was 40-60 min.

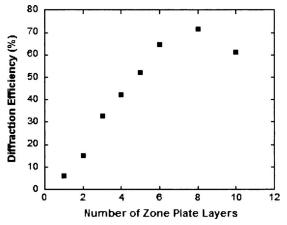


Fig. 3. Diffraction efficiency as a function of the number of modified phase zone plates in a volume.

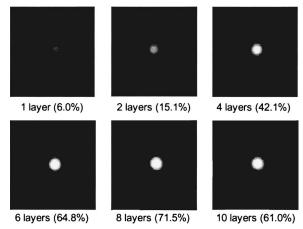


Fig. 4. Focal spots of low NA volume phase zone plates with central rings.

For a high NA volume zone plate, the spacing between layers had to be as close as possible because a small change in focal length causes the NA number to decrease significantly. The NA number of the volume zone plate was defined by using the longest focal length of the generated zone plates. Volume zone plates with NA=0.84 were fabricated up to five layers to observe an increase in diffraction efficiency.

4. RESULTS AND DISCUSSION

A. Low-Numerical-Aperture Zone Plates

The fabricated volume Fresnel zone plates were tested for their diffraction efficiencies using a He–Ne laser at a wavelength of 632.8 nm. The method for measuring the diffraction efficiency has been described previously [9]. Our single layer phase zone plates had an efficiency of 5.3%. As expected, this low efficiency was due to the small change of the index of refraction that was insufficient to induce a π phase shift. By adding more layers to build a volume phase zone plate, the highest efficiency of a volume zone plate with central rings was improved to 71.5% with eight layers of phase zone plates. This efficiency was higher than the previously published result [9] even though the number of layers was the same because of a better control of filament length and index increase. Figure 3 shows the measured efficiencies of volume Fresnel

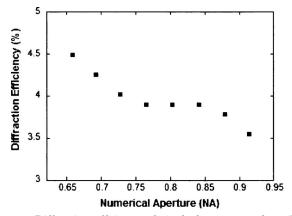


Fig. 5. Diffraction efficiency of single layer zone plate (NA = 0.66-0.91).

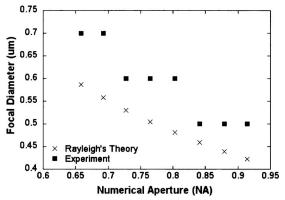


Fig. 6. Experimental spatial resolutions compared with the Rayleigh criterion of single layer high NA zone plates. zone plates as a function of the number of layers of zone plates. This result implied that the volume method was more effective than simply stacking the zone plates to achieve the π phase shift because the efficiency exceeded the maximum possible of 40% by a phase-reversal zone plate. The increase in diffraction efficiency of the volume zone plate can be explained in terms of coherent addition of light from different zone plates [8]. For example, the intensity due to two zone plates can be written as $I=I_1+I_2$ $+2\sqrt{I_1I_2\cos\delta}$ where δ is the phase shift between the two. In our case, we specified zero phase shift so that diffracted intensity is as large as possible. However, the diffraction efficiency could not be simply predicted by only the coherent addition of beams because there are other effects involved, such as multiple diffractions from one layer to the next layers, which required a rigorous analy-

Without a model simulation, it is hard to confirm that the possible maximum efficiency is 71.5%. The coherent addition of diffracted waves allows the volume zone plates to achieve relatively high efficiencies compared to binary zone plates. These volume zone plates can be further optimized by changing the laser scanning speed to fabricate a zone plate with different efficiency at each layer. The maximum efficiency is believed to be a function of such processing. The imperfection in manufacturing also causes a decrease in efficiency, which is hard to estimate in the simulation. Further investigation requires more careful study of refractive index change and numerical simulation of light diffraction from a volume zone plate. Also, the efficiency is expected to be a wavelength dependent property.

The images at the focal spots of the low NA volume zone plates are shown in Fig. 4. The spot size was minimally affected by aberrations, and the focal spot produced by the eight layer phase volume zone plate was $\sim\!20~\mu\mathrm{m}$ at full width at half maximum (FWHM), which agreed with the theoretical value of 19.3 $\mu\mathrm{m}$. The volume phase zone plates had a significant increase in diffraction efficiency when having eight layers. However, ten layers or more of zone plates started to show a decrease in efficiency. This decrease can be explained by three possible reasons: (1) interaction of light passing through many layers causes phase and focal position error at the focal point, (2) light absorption or scattering becomes noticeable with many phase layers, and (3) accumulated fabrication errors.

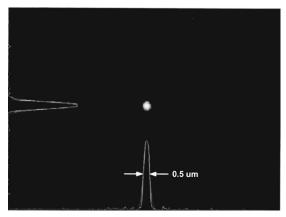


Fig. 7. Focal spot of high NA zone plate (NA=0.91, f=60 μ m).

B. High Numerical Aperture Zone Plates

For the high NA zone plate, the diffraction efficiency is low, which causes relatively large measurement uncertainties. The results of single layer high NA zone plates are shown in Fig. 5, together with the estimated measurement uncertainties. For a single layer zone plate of NA =0.66-0.91, the diffraction efficiency trended to be slightly higher for a lower NA zone plate. The possible reason is that the higher NA zone plates were partially overlapped at the outer zones, resulting in more phase shift error. The single layer high NA zone plates had efficiencies of 4.5% to 3.5% for NA from 0.66 to 0.91. The lower diffraction efficiency as compared to a low NA volume zone plate is mainly due to the much shorter filament length produced by a high NA objective lens.

In the study of high NA zone plates, we were especially interested in reducing the focal spot size or improving the spatial resolution of the zone plate since these fabricated zone plates are often intended for high-resolution applications. Our single layer high NA zone plates could produce a spot as small as 0.5 μ m (FWHM) for a zone plate with an NA=0.91. Figure 6 compares our experimental resolution of high NA zone plates with the Rayleigh criterion. The focal spots of our zone plates were larger than the predicted values, possibly because of aberrations and overlapped zones. The overlapped zones reduced the effective diameter of the zone plate and lowered the effective NA. On the other hand, the smallest spot size for high NA zone plate still showed good Gaussian profile as shown in Fig. 7.

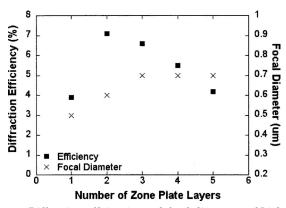


Fig. 8. Diffraction efficiencies and focal diameters of high NA volume zone plates (NA=0.8).

The diffraction efficiencies and the corresponding focal spot sizes of volume zone plates with NA=0.84 having up to five layers are shown in Fig. 8. The maximum efficiency of 7.1% was obtained with a two-layer zone plate, and the focal spot size increased with the number of layers. With two layers, the diffraction efficiency was maximized while the total phase shift was only 0.06π . This phase shift could be increased with more layers but there were other factors that diminished efficiency as the number of layers increased. Therefore, we used a high magnification microscope to observe the filament induced by a 100× objective, and found a dim spot on top of the filament. This dim spot indicates optical damage produced by a tightly focused femtosecond pulse. Therefore, the high NA zone plate produced by a 100× objective might not be purely phase-type and can absorb light. Absorption could be a significant cause for efficiency reduction when the number of layers increases. Accumulation of fabrication errors can limit the high-resolution performance when the number of layers increases.

5. CONCLUSIONS

We designed and fabricated low and high NA volume Fresnel zone plates inside a bulk of fused silica by femtosecond laser direct writing. A new technique for modified Fresnel zone plates in which only the central ring of each zone was irradiated by a laser was used to greatly reduce fabrication time and phase errors. Efficiency as high as 71.5% was obtained for a low NA volume zone plate consisting of eight layers. High NA zone plates were also successfully fabricated. Compared to low NA zone plates, high NA zone plates had much shorter filament length and absorbed light, resulting in lower diffraction efficiencies. Nevertheless, volume zone plates could offer much higher efficiency than single layer zone plates with spatial resolution of 500 nm. More rigorous design, optimization, and improved fabrication techniques are needed to further increase the performance, especially for high NA volume zone plates.

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