Coherent stitching of light in multilayered diffractive optical elements

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Abstract: Diffractive optical elements serve an important function in many dynamic and static optical systems. Multilayered diffractive elements offer powerful opportunity to harness both phase and amplitude modulation for benefits in diffraction efficiency and beam shaping. However, multilayered combinations have been difficult to fabricate and provide only weak diffraction for phase gratings with low refractive index contrast. Femtosecond laser writing of finely-pitched multilayer volume gratings was optimized in bulk fused silica. We identify and quantify an optimum layer-to-layer separation according to Talbot self-imaging planes and present systematic experimental validation of this new approach to enhance otherwise weakly diffracting volume gratings.

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1. Introduction

Diffractive optical elements serve in many light systems today as dynamic or static devices, for example, Spatial Light Modulators [1] or Computer Generated Holograms (CGH) [2], to serve in pulse shaping, 3D imaging, microscopy, optical tweezers, and beam shaping [1, 3, 4]. The theory and design of surface diffractive structures are well understood and practically applied at high spatial and phase resolution for a wide range of optical applications in science and industry [5]. However, these structures normally only harness phase modulation of uniform fields for the beam diffraction and therefore limit their range of application. Further, the finely structured phase elements on an open surface are highly susceptible to damage. Volume diffractive elements, such as photorefractive volume gratings [6], CGH [2] and laser direct written gratings [7], offer an attractive alternative approach that mitigates sensitivity to surface damage. Such volume elements can also capture higher diffraction orders than is not possible with surface elements or can enable new types of form-birefringence [8, 9]. Towards this end, Gerke and Piestun [10] explored the design and laser fabrication of aperiodic volume optics by considering a volumetric scattering model, but limited to only cases of very weak diffraction. Various other groups have applied femtosecond laser writing to demonstrate various types of buried phase gratings and diffractive optics [7, 11–13] with moderately high diffraction efficiency, but aimed towards larger pitch phase elements of several wavelengths in size. For more advanced cases of creating finely pitched buried diffractive optics, the combination of low refractive index contrast and wavelength-scale phase elements is constrained to unattractively low diffraction efficiency that cannot be compensated with thicker diffractive structures except in the very limited case of Littrow configuration [14].

New optical design concepts are therefore required before low contrast DOEs can be practically developed into powerful high resolution optical devices that offer high efficiency.

Various groups [15–17] have used the principle of cascading diffractive structures to accumulate stronger phase contrast and greatly improved the efficiency over a single refracting layer by effectively increasing the overall diffracting length. In this paper, we show the break down of this simple principle for weakly contrasting gratings when the period scales toward the size of optical wavelength. Rather than increasing the grating thickness, much earlier work considered the stacking of multiple planes of surface relief gratings by Malysch et al. [18] and Nordin et al. [15] or layers of acoustic phase gratings by Hargrove et al. [19] that identified theoretically a curious periodic relationship between diffraction efficiency and
separation of the layers of gratings. With high index contrast and large period gratings, relative to the incident wavelength, the stacking of such gratings for high diffraction efficiency has not been essential. We identify and confirm in optical models that this periodic layer-to-layer separation for enhancing diffraction efficiency corresponds to the positioning of grating layers precisely onto Talbot planes where the concept of self-imaging manifests in coherent summation of diffraction from each layer. Modeling shows high diffraction efficiency is available for an optimized number of ideally separated grating layers. We exploit these concepts in the context of femtosecond laser writing of weakly diffracting and finely pitched phase gratings inside fused silica glass, and definitively demonstrate for the first time the coherent and incoherent collective response of multilayer stacked gratings that respectively enhance or diminish the diffraction efficiency according to their stacking period and number of grating layers. With this new understanding, a realm of new opportunities is now available for the harnessing of weakly diffracting DOEs for high resolution applications.

2. Methods

2.1 Theoretical modeling

A full vector calculation of diffraction from single-layered phase gratings was made with commercial software (Grating Solver Development Company; GSolver 4.20c). Advanced Finite Difference Time Domain (FDTD) methods (Lumerical; FDTD Solutions) were used for generating near-field intensity patterns from multi-layer phase grating structures inside fused silica glass and further projected into far-field diffraction patterns to ascertain the multi-order diffraction efficiencies.

2.2 Device fabrication and characterization

The 3D optical devices were fabricated inside fused silica (Corning 7980, \( n_r = 1.46 \), 1 mm thickness) with a Yb-fiber amplified femtosecond laser (IMRA \( \mu \)Jewel D-400-VR) operating at 1MHz repetition rate and frequency doubled through an LBO crystal (Newlight Photonics) to \( \lambda = 522.5 \) nm wavelength for a pulse duration of 218 fs (FWHM). The laser was focused into the fused silica target with a microscope objective (Zeiss Plan 100 ×, \( NA = 1.25 \)) and immersion oil (Carl Zeiss Immersion glycerine 462959) to an estimated spot size of \( w = M^2(\lambda/\pi w_0) \approx 330 \) \( \mu \)m, where \( M^2 = 1.4 \). For precise positioning (± 100 nm reproducibility) and motion control (G-code programming), the focusing lens and translations stages (Aerotech ABL1000) were affixed onto a granite arch for forming linear and multi-level stacked phase gratings over a range of lateral periodicity, \( \Lambda = 0.6 \) to 5 \( \mu \)m. The refractive index contrast and grating efficiency were optimized by scanning laser tracks over a 5 to 100 nJ range of laser energies and 1 to 100 mm/s sample scan speeds, with the objective to exploit the smallest periodicity that does not wash out the diffraction efficiency.

The diffractive devices were probed with a diode-pumped solid state green laser (Lasermate GME-532-10FBP1, \( \lambda = 532 \) nm), focused to a large spot size of ~300 \( \mu \)m. The broad laser line width of 0.1 nm precluded consideration of Fabry-Perot interference effects. The diffraction efficiency, through diffractive structures of 500 \( \mu \)m × 500 \( \mu \)m area, was measured with a low-power optical detector (Newport 818-SL), filtered through a pin-hole to isolate individual diffraction orders. Other modes of observation included optical and refractive near-field (Exfo OWA9500) microscopy.

3. Weakly diffracting volume phase gratings

For traditional surface diffractive structures of binary rectangular grooves, scalar theory predicts that the diffraction efficiency can be controlled by the refractive index contrast (\( \Delta n \)) and the depth of the grating groove (\( d \)), which imparts a phase shift modulation of

\[
\phi = 2\pi (\Delta n) d / \lambda
\]
on the phase front for probe wavelength, $\lambda$, for the case when $\Lambda \gg \lambda$.

Figure 1(a-d) shows the Finite Difference Time Domain (FDTD) calculation of interference intensity pattern of light traversing through a volume phase grating of $\Lambda = 1 \mu m$ period and $d = 20 \mu m$ thickness. The different refractive index contrast gratings, $\Delta n = 0.02, 0.06, 0.016$, and 0.46, with the higher refractive index volume demarked by the hatched region in the figures, were probed from the bottom ($z = 0 \mu m$) with $\lambda = 532$ nm light at uniform incident intensity $I_0 = 1$. For each grating case, the projected combined 1st order diffraction efficiency, $\eta$, was calculated for increasing grating depths, $d$, as shown in Fig. 1(e).
The minimum grating thickness, $d_\phi = \pi$, required for maximum 1st order diffraction efficiency was also determined as anticipated by Eq. (1) and plotted in Fig. 1(f) over a broad spectrum.

In Fig. 1(a), the grating with an air-glass interface of $\Delta n = 0.46$ results in strongly contrasting intensity fringes (0 to 7.43$I_0$) with the first peak positioned at $z = 0.7 \mu m$ that corresponds to a build up of $1.19\pi$-phase shift and a peak diffraction efficiency of $\eta = 90\%$ in Fig. 1(e)(purple). Here, a breakdown of scalar theory for such fine grating period ($\Lambda \sim 2\lambda$) leads Eq. (1) to underestimate the grating thickness for optimum diffraction efficiency [20, 21]. With decreasing refractive index contrast, the intensity contrast decreases strongly from Fig. 1(a) to 1(d) and, correspondingly, the diffraction efficiency also drops dramatically from $90\%$ (Fig. 1(e)(purple)) to a maximum of only $4.4\%$ (Fig. 1(e)(green)) for the weak contrast case regardless of the phase grating thickness. For this $\Delta n = 0.02$ case (Fig. 1(d)), the constructive interference builds up to only a moderate intensity peak of $1.7I_0$ at grating depth $z = 2.65 \mu m$. An examination of Fig. 1(d) revealed that the periodic oscillation of the intensity pattern did not correspond to $d_\phi = 21.3 \mu m$ (red arrow) as anticipated by Eq. (1). However, Chanda and Herman [22] noted that the interference pattern generated by a phasemask of period $\Lambda$, would lead to coherent combination of the diffraction orders on planes that repeated over a similar distance, $c$, identified as Talbot planes,

$$c = (\lambda/n_r)\sqrt{(1 - (\lambda/n_r \Lambda)^{-2})}$$  \hspace{1cm} (2)

where $n_r$ is the background index of refraction. Indeed, the periodic oscillation observed in Fig. 1(d) corresponds to this Talbot self-imaging separation, as indicated by the white arrow in the figure. For the gratings with high index contrast in Fig. 1(a), the oscillations within the grating length correspond more closely with the expected $2\pi$-phase modulation distance given by $d_\phi = 2\pi$ (red arrow), which is significantly shorter than the Talbot plane separation, $c$ (white arrow). As the refractive index contrast decreases (Fig. 1(a) to 1(d)), the $d_\phi = 2\pi$ (red arrow) length expands to beyond the Talbot spacing $c$ (white arrow) which limits the available diffraction efficiency.

A plot of the 1st expected intensity fringe as predicted by Eq. (2), given by $c/2$, is shown in Fig. 1(f)(black) to rise strongly with decreasing wavelength. This half-Talbot distance is independent of the index contrast ($\Delta n$) as noted by the white arrows in Fig. 1(a-d). The fundamental limit of high diffraction efficiency is governed by the transition from classical diffraction where the $\pi$-phase contrast distance, $d_\phi = \pi$, is found to exceed the half-Talbot plane distance, $c/2$. The fall off in diffraction efficiency occurs when the Talbot self-imaging oscillates on distances shorter than required for generating high contrast fringes on spacing $d_\phi = \pi$, namely, when $c/2 < d_\phi = \pi$. In this way, the grating thickness for the 1st diffraction peak follows the $d_\phi = \pi$ condition, increasing linearly with wavelength in Fig. 1(f) for all values of index contrast, until intercepting the $c/2$ line (black). For the case of $\Delta n = 0.02$, this juncture at 269 nm wavelength turns over the optimum grating thickness to follow the $c/2$ curve towards longer wavelength as shown by the dashed green curve in Fig. 1(f). In the present case of 532-nm probe wavelength, the half-Talbot self-imaging length of $c/2 = 2.65 \mu m$ (Fig. 1(f)(black)) is 4-fold shorter than the $d_\phi = \pi = 10.6 \mu m$ distance (Fig. 1(f)(green)) required for $\pi$-phase shift, and insufficient phase shift of $0.25\pi$ is accumulated that limits the diffraction efficiency to peak at $\eta = 4.4\%$ as shown in Fig. 1(e)(green). For high refractive index contrast of $\Delta n = 0.46$, the intensity in Fig. 1(a) oscillates on $d_\phi = 0.7 \mu m$, as the accumulation of maximum phase modulation occurs well before the Talbot self-imaging distance for all permitted diffracting wavelengths ($\lambda < 1 \mu m$) shown in Fig. 1(f)(purple). Figure 1(a) also reveals finer fringe patterns (i.e. $\Delta z \sim 200 \text{nm}$) inside the Talbot self-imaging distance $c = 5.3 \mu m$ that arises from interference of the $m = 0$ and $\pm 1$ with the $m = \pm 2$ diffraction orders that undergo total internal reflection at the glass-air interfaces. Continuous diffraction along the propagation length of the phase grating leads the 2nd order beams to cause a longer range modulation of the peak intensity and the 1st order diffraction efficiency that is apparent particularly for the
high index contrast case of $\Delta n = 0.46$ case on distance $\Delta z = 7.3 \mu m$ in Fig. 1(a) and Fig. 1(e)(purple).

Figure 1(g) shows the minimum refractive index contrast in glass gratings ($n_r$) before oscillation on Talbot self-imaging planes will curtail diffraction efficiency for cases of 400, 532, and 800 nm wavelength. For the visible spectrum ($\lambda > 400$ nm), Talbot oscillation restricts the choice of index contrast to $\Delta n > 0.27$ for generating efficient gratings with 1 $\mu m$ period. One can see that with longer period gratings, such as $A = 3 \mu m$, much lower index contrast of $\Delta n = 0.003$ will result in efficient gratings that are optimized on the traditional $-\pi$-phase modulation condition.

Intuitively, a reconstruction of the phase gratings to follow this Talbot self-imaging oscillation along the propagation direction, $z$, may push light coherently into the desired diffracted order and result in stronger intensity contrast with higher diffraction efficiencies. Indeed, Hargrove et al. [19] noted, without physical insight, that gratings separated by periodic zones of uniform dielectric resulted in strong enhancement of diffraction efficiency when the layer to layer grating separation was a multiple of $c_r$ given by:

$$c_r = 2(n_r^3/\lambda).$$  \hspace{1cm} (3)

We identify Eq. (3) as an approximated version of the Talbot plane separation (Eq. (2)) with the assumption that the grating period is much larger than the incident wavelength ($A \gg \lambda$).

A similar expectation for coherent combination of 1st order diffraction beams is proposed here for multi-layered gratings constructed by segmenting the volume grating such as in Fig. 2(a) into thin phase grating layers separated with centre-to-centre spacing of $c_r$ or multiples thereof. Figure 2(b) is an example of 8 layers of gratings, each of $d = 7 \mu m$ thickness, with hashed regions corresponding to high index zones that are separated by $2c = 10.6 \mu m$ to coincide with the predicted Talbot planes. As the light travels through the multiple layers, the intensity is seen to build into a deeper modulation contrast with each layer, peaking to a maximum of $3.6I_0$ at $z = 16c = 84.8 \mu m$ (8 layers) that is more than double the maximum intensity contrast found at the Talbot positions $z = c/2, 3c/2, 5c/2...$ for the uniform phase grating in Fig. 2(a). By extending further the phase grating in Fig. 2(b) to 15 layers, there is a reversal of this building intensity contrast together with diminishing first order diffraction efficiency to zero at 15 layers as seen in Fig. 2(c). Therefore, the intensity contrast oscillates with increasing Talbot layers, but are enveloped by the trend such as seen in Fig. 1 for continuous phase gratings that yield oscillating maxima (8 layers = $16c$) and minima (15 layers = $30c$). The finer scaled periodic intensity modulations (i.e. $\sim 1 \mu m$) in Fig. 2(b) and 2(c) also arise from weak 2nd order diffracted beams present only inside the glass substrate.

4. Modeling diffraction from multi-layered phase gratings

The coherent enhancement of diffraction efficiency when satisfying the Talbot separation condition in Eq. (2) was examined by calculating the far-field diffraction efficiency from two layers of identical phase gratings inside 1-mm thick fused silica. The grating layers each have periodicity, $A = 1 \mu m$, base refractive index, $n_r = 1.46$, refractive index contrast, $\Delta n = 0.025$, and grating thickness, $d = 7 \mu m$, and were probed with incident wavelength, $\lambda = 532$ nm.
Fig. 2. A plane wave ($\lambda = 532$ nm) incident from the bottom ($z = 0$) generates intensity interference patterns through (a) a thick continuous phase grating of $20 \mu$m thickness, (b) 8 layers of $7 \mu$m thick gratings, and (c) 15 layers of $7 \mu$m thick gratings, each with $1 \mu$m longitudinal period and $\Delta n = 0.02$ refractive index contrast. Colour scale bar represents 0 to 3.6 intensity normalized to an incident intensity, $I_0 = 1$. The hatched zones indicate the position of the higher refractive index volumes. Light is incident from the bottom.

The combined 1st order diffraction efficiency generated for increasing separation, $l$, between the grating layers yielded the sinusoidal-like modulation from 0% to 13.6% efficiency in Fig. 3(a)(solid red line), with the maxima aligned with grating separations matched precisely to the Talbot planes at multiples of $c = 5.3 \mu$m, $2c = 10.6 \mu$m, $3c = 15.9 \mu$m, ... Correspondingly, the first order beams from each grating layer are interfering destructively for intermediate layer separations $3c/2$, $5c/2$, ..., attesting to the powerful coherent Talbot alignment effects present in multi-layered gratings. To further scale up the efficiency of such weak phase gratings, additional grating layers were considered with ideal $2c$ grating layer separation. In Fig. 3(b)(red), the theoretical combined 1st order diffraction efficiency is found to build to a peak of ~95% for 8 layers and trough at ~15 layers as anticipated by the intensity pattern trends presented in Fig. 2(b) and 2(c).

5. Femtosecond laser direct writing of volume phase gratings

New direct-writing techniques afforded by femtosecond lasers [7, 23–26] permit formation of very small refractive index structures that can be arbitrarily spaced on optical wavelength dimensions inside bulk transparent glasses. Such laser writing presents a unique opportunity here to systematically study the Talbot effects and the predicted enhancement of diffraction efficiency for multilayered diffractive structures.
Fig. 3. Calculated (red triangle) and measured (blue square) efficiency of both first order diffracted beams ($\lambda = 532$ nm, $A = 1$ $\mu$m, $\Delta n = 0.025$) for (a) 2 layers of 7 $\mu$m thick gratings with increasing layer-to-layer separation and (b) increasing number of layers, with ideal layer-to-layer separation, $2c$. Solid lines in (a) are sinusoidal fits to the data.

Figure 4(a) shows a microscope image end view of the index modification formed by a single laser track for the exposure conditions in the methods section. Both positive and negative index changes are observed across a vertical length of $\sim 10$ $\mu$m due to self-focusing and defocusing effects. Only a phase contrast modification was generated at these laser exposure conditions, which lie below the threshold for void formation [27] or filamentation [7]. The modification track as shown in Fig. 4(a) were assembled into single layered phased gratings, and optimized to provide a moderate value of $\eta = 5.2\%$ for the combined 1st order diffraction measured at the high resolution limit of $A = 1$ $\mu$m period. Refractive near-field imaging provided an estimate of $\Delta n = +0.010$ for the peak refractive index contrast for the single track. However, an optical resolution limited to $\sim 0.5$ $\mu$m precluded a reliable assessment of the finely structure refractive index profile consisting of multiple positive and negative zones seen in Fig. 4(a). Hence, a uniform phase grating representation of rectangular bars of $0.5 \times 7$ $\mu$m$^2$ and $\Delta n = +0.025$ was used to accurately represent the 1st order diffraction efficiency measured for 1 $\mu$m period laser written gratings. Figure 4(b) shows a 3D visualization of a multilayered phase grating structure formed by stacking together horizontal arrays of laser modification tracks. Diffraction from laser written grating structures ($A = 1$ $\mu$m, $d = 7$ $\mu$m), such as is shown for the 10 layer case in Fig. 4(c), was used to verify the theoretical predictions in Fig. 3(b) and confirm the coherent principles of Talbot plane alignment.

Figure 3(a)(blue) shows the diffraction efficiencies measured for double layer phase gratings with various layer-to-layer separations over the range of $l = 7$ to 29 $\mu$m. The experimental data follow the general trend of the modeling results with periodic cycles that peak at separations of $l = 11, 16, 21,$ and $27$ $\mu$m, which align closely with the expected Talbot plane multiples at $2c = 10.6, 3c = 5.9, 4c = 21.2,$ and $5c = 26.5$ $\mu$m separations.
The matching of the simulated diffraction efficiency (red triangles) to experimental values as seen in Fig. 3(a) was found for an index contrast of $\Delta n \approx 0.025$ across an inferred grating thickness of 7 μm. This strong refractive index contrast exceeds the $\Delta n = 0.015$ value reported for waveguides written with similar exposure conditions [28] and may suggest that the positive index contrast zone is longer than the 7 μm inferred here or that other factors such as the formation of weak negative index zones between the laser tracks are also contributing to the phase contrast. Further, a fortuitous alignment of the negative and positive refractive index zones seen in Fig. 4(a) to near the half-Talbot plane separation distance, $c/2 = 2.65$ μm, would also contribute a strong in-phase enhancement similar to the $c$ alignment of positive phase gratings considered in Fig. 2(b).

For the same laser writing conditions as used in Fig. 3(a), multi-layered phase gratings were written at the optimal separation of two Talbot planes ($l = 2c = 10.6$ μm), yielding the measured combined 1st order diffraction efficiencies plotted in Fig. 3(b)(blue). Figure 4(d) shows the enhanced 1st order diffraction generated by the 10 layered grating over the single layer grating. The efficiency rises strongly from 5.2% of the single layer to a peak of ~35% at 8 layers (Fig. 3(b)(blue)), which falls short of the 95% value expected theoretically. The efficiency falls off only gently to ~26% at 15 layers, where a null was expected theoretically.

Several factors may underlie the discrepancy between experimental and theoretical data for high number of grating layers (>3). The multiple negative and positive zones of refractive index change as observed in the single track of Fig. 4(a) suggest more complex diffraction phenomena underlie the optical propagation than accounted for by the uniform rectangular
contrast phase grating model applied to our FDTD analysis in Fig. 3. This simplified representation of the single layer phase grating was found to accurately illustrate the coherent layering effect on Talbot planes for the two-layer phase grating data presented in Fig. 3(a), but may become a factor for the efficiency discrepancy in Fig. 3(b) at four or more grating layers.

Alternatively, we have observed the overall stress generated in glass to increase dramatically as large volumes of laser tracks were woven into small volumes, possibly limiting the net index contrast obtainable as the overall glass structure saturates to a maximum obtainable refractive index contrast. Thus, the intermediate layers in many layered grating structures may contribute lower overall refractive index contrast and grating efficiency than that available from the top and bottom layers. To bypass this fundamental limit of forming strongly contrasting grating structures, a less dense arrangement with vertical layer-to-layer separations of \( l = 3c \) or \( 4c \) may be considered, but could not be tested here due to the limited working distance of the present objective lens.

An assessment of the fabricated grating optical losses yielded a 1.5% Fresnel reflection loss from the combination of two air-glass boundaries together with single-layer grating scattering losses of \( \sim 1\% \) attributed to submicron graininess in the laser formed grating lines. However, such losses could only account for a quarter of the efficiency discrepancy in Fig. 3(b). Alternatively, the stitching accuracy of the grating layers both laterally and vertically, with motion stage positioning precision of \( \pm 100 \text{ nm} \), may cause blazing effects as well as randomization of the diffraction efficiency that may reduce the peak of each 1st order diffraction efficiency by no more than 20% in calculations, and could also not fully explain the observed trends in Fig. 3(b). For example, in the extremely unlikely case of 100-nm misalignment of adjacent grating layers, FDTD simulation shows the diffraction efficiency to only fall from 95% to 77.2% over 8 layers of volume gratings. The presence of both positive and negative refractive index changes (Fig. 4(a)) together with a complex combination of the above variances possibly underpins this discrepancy in diffraction efficiency. An accurate profiling of the refractive index contrast on a high resolution scale of \(< 200 \text{ nm} \) is required to better assess and fully harness laser direct writing for generating highly efficient multi-layered phase gratings.

6. Discussion

The insight into the Talbot self-imaging oscillation first presented in Fig. 1 revealed a fundamental restriction on the available diffraction efficiency from high resolution, low refractive index contrast phase gratings. This sparked the concept of stacking grating layers that was theoretically found to enhance or diminish the 1st order diffraction efficiency with the separation between gratings aligned or misaligned according to the periodic Talbot planes. Moreover, this enhancement could be optimized with selection of the appropriate number of grating layers. Femtosecond laser fabrication of multi-level phase gratings was applied inside bulk glass and found to verify the importance of layering on Talbot planes as well as demonstrate an 8-fold enhancement of the diffraction efficiency with the appropriate assembly of 8 grating layers. Further improvements in the laser fabrication technique will therefore permit multi-layered phase gratings to generate a potentially high diffraction efficiency of up to 95% as compared with only 4.4% for thick gratings.

In this paper, we introduced a new approach to understanding the limitations of weak diffractive elements in bulk glasses that was followed with a method to dramatically improve the diffraction efficiency through coherent layering of these phase elements. In the case of simple periodic grating structures, the placement of the layers at the periodic Talbot planes presented an arriving electric field pattern with correct phase coherence to build up strong interference contrast and generate strongly diffracting orders. These principles were confirmed by FDTD modeling and verified experimentally through laser direct writing of multi-layered phase gratings in bulk glass at high resolution. This concept of phase control for multilayered structures can now be combined with the manipulation of the light amplitude
towards new unexplored directions in the design and fabrication of DOEs that now go beyond those first proposed by Hargrove et al. in 1962.

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