Etch-free additive lithographic fabrication methods for reflective and transmissive micro-optics

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Abstract: With the widespread application of micro-optics in a large range of areas, versatile high quality fabrication methods for diffractive optical elements (DOEs) have always been desired by both the research community and by industry. Traditionally, multi-level DOEs are fabricated by a repetitive combination of photolithography and reactive-ion etching (RIE). The optical phase accuracy and micro-surface quality are severely affected by various etching artifacts, e.g., RIE lag, aspect ratio dependent etching rates, and etching artifacts in the RIE steps. Here we propose an alternative way to fabricate DOEs by additively growing multi-level microstructures onto the substrate. Depth accuracy, surface roughness, uniformity and smoothness are easily controlled to high accuracy by a combination of deposition and lift-off, rather than etching. Uniform depths can be realized for both micrometer and millimeter scale features that are simultaneously present in the designs. The grown media can either be used directly as a reflective DOE, or as a master stamp for nanoimprinting refractive designs. We demonstrate the effectiveness of the fabrication methods with representative reflective and transmissive DOEs for imaging and display applications.

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1. Introduction

Micro-optics have gained widespread interest in a large variety of areas due to their flexibility in light manipulation with a small form factor. As a generic platform, diffractive optical elements (DOEs) [1] are a class of thin and lightweight optical components with constituent feature sizes approaching the operating wavelengths. Particularly in imaging and display devices, DOEs are perfect candidates to perform multifunctional light modulation as well as to miniaturize optical systems. Thanks to modern photolithography techniques, the fabrication of DOEs is more easily accessible, allowing them to play more and more significant roles in the optical designers’ toolbox. More recently, the integration of diffractive optics and computational imaging has pushed forward more practical applications of DOEs in broadband imaging applications [2–5]. The demand of versatile high quality DOEs in such applications in turn necessitates effective yet easy-to-implement fabrication methods.

Traditionally a prevailing DOE fabrication routine is to combine photolithography and reactive-ion etching (RIE) techniques [6] because of the readily available equipment and standardized procedures borrowed from the semiconductor industry. The RIE step is key to defining the 3D microstructures on the substrate, and hence the final fabrication quality and optical performance. Although fine tuning of the parameters in RIE is possible to maintain high topological accuracy, RIE inherently suffers from several drawbacks in producing uniform and smooth microstructures, such as RIE lag, aspect ratio dependent etching (ARDE), and various etching artifacts [7, 8]. These limitations and artifacts cause serious optical artifacts when features of different scales are present on the same optical design. A simple example is the Fresnel lens, where the etched depths vary from center to edges due to RIE lag and ARDE. The uniformity of depths is challenging to maintain if there exist both micrometer and millimeter patterns. The surface quality is also
affected differently by these artifacts for varying feature sizes. In multi-level DOEs, errors accumulate throughout the process, leading to the drop in diffraction efficiency compared to theoretical calculations. In addition, care must be taken to maintain the stability of etching rates from time to time. In a word, RIE is the most error-prone step from both a fabrication quality and a yield perspective.

In recent years, emerging additive lithography techniques have been proposed as alternatives for DOE fabrication. A previous photoresist additive lithography method [9, 10] makes full use of the linear region between the bias and saturation regions of the resist response curve. With a series of different exposure doses and pattern masks, 3D multi-level microstructures can be formed on the photoresist on a stepper with automatic and accurate alignment. A single etching step is still needed to finally produce the required DOEs onto the substrate. The number of process steps are reduced in this method, but the linear range ultimately determines the maximum heights and number of levels, and careful characterization of the exposure time is important for successful implementation. Another additive fabrication method combines the area-selective deposition with patterned self-assembled monolayers (SAMs) [11] to grow patterned materials in a chemical way. A photoresponsive SAM assemble from the surface adsorption and form ordered crystalline monolayers. Upon exposure with a mask, selective areas are cross-linked, which are then deactivated from atomic layer deposition. Material growth is allowed in the unexposed regions, and inhibited in the exposed regions. Therefore, the induced chemical difference enables the required relief from deposition. Although it is feasible to fabricate semiconductor devices with these organic materials, optical component fabrication has not been reported yet. Another related fabrication method is soft lithography [12]. A master stamp is first fabricated (usually on photoresist) with conventional lithography techniques, and then transferred via a soft stamp to the final substrate.

Here, we propose and demonstrate alternative etch-free additive DOE fabrication methods to mitigate the above challenges. Instead of removing materials from the substrate, we propose to form the microstructures by growing materials onto the substrate, i.e., additive fabrication. Our additive fabrication can be used to directly fabricate reflective designs by depositing Chromium (Cr) or Aluminum (Al) onto fused silica (FS) substrates by sputtering followed by lift-off. Refractive/transmissive designs can be achieved by using the same process to fabricate a master stamp of the design, and then transfer the final patterns to proper transparent substrates by nanoimprint. The final substrates and materials can be chosen depending on applications, either rigid (e.g., Poly(methylmethacrylate), or PMMA) or flexible (e.g., Polydimethylsiloxane, or PDMS). In principle, the proposed methods are different from existing additive lithography methods. We do not place stringent requirements on the choice of photoresists, as there is no need for demanding control of photoresist preparation. Microstructures are generated from the deposited materials, not from the photoresists. It’s therefore more flexible for both reflective and transmissive DOEs. Compared with soft lithography, our patterns are deposited on “hard” metals, and there are fewer topological artifacts being transferred from the photoresist to the stamps and final substrates by nanoimprint [12].

With the proposed additive fabrication methods, we are able to thoroughly eliminate the etching steps from the procedure. Compared with etching, the deposition thickness can be time-controlled very well with nanometer accuracy, and is more stable over time. The deposition is uniform across the whole wafer (typically 4-inch) for both micrometer and millimeter patterns. This results in high-quality surface roughness to suppress scattering and improve diffraction efficiency, which is important for many optical applications. By eliminating the etching steps, we significantly simplify the overall procedure compared with conventional etching based methods. The risk of breaking or wasting wafers can be reduced significantly, and hence the cost of fabrication is reduced. Last but not least, the process itself is suitable for mass production by the introduction of nanoimprint. In a research laboratory setting, the fabrication can be carried out
in a small scale with different designs from wafer to wafer. In mass production, a high-quality master stamp could be fabricated with metal structures, and replicated in a large amount in the nanoimprint step. We envision the proposed methods to be alternative options in the fabrication of micro-optics components, in both reflective and transmissive modes.

2. Etch-free additive lithographic fabrication

We first illustrate two successive fabrication cycles of the traditional RIE-based multilevel DOE fabrication in Fig. 1a. In each iteration, an FS wafer is first prepared with thin sacrificial layers. A photoresist (PR) layer is usually spin-coated for the pattern transfer, and a Cr layer is deposited as a hard mask because of its high selectivity in the RIE step. The photolithography step defines the spatial resolution via proper Ultraviolet (UV) light exposure followed by development. Subsequently, the open areas in the Cr layer are removed with a Cr etchant to form a hard mask. Substrate materials in the open areas are removed by plasma in the RIE step, and a certain height profile is created on the substrate after removing auxiliary layers. Each fabrication cycle doubles the number of microstructure levels on the previous profile. By repeating this cycle over \( N \) iterations, \( 2^N \) levels of microstructures can be achieved on the substrate.

Typical RIE artifacts [7,8] are shown in Fig. 1b. RIE lag refers to the difference in etched depths for different features. Normally smaller features gain shallower depths compared with larger features on the same wafer, given the etching conditions are the same. As the etching proceeds, the aspect ratio becomes larger and larger, therefore, the etching rate is slowed down, which results in shallower depths for the same etching period, or longer etching time to reach the same depth as in a previous RIE step, shown in Fig. 1c. In addition, trenching, faceting, spearheading, rounding, and notching (footing) could occur at the bottom of the etched regions, as shown in Fig. 1d. Achieving mirror-like surface finishes is even more challenging with dry etching methods.

In the proposed additive lithographic methods, we retain the conventional photolithography step for pattern transfer, but employ sputter deposition and bi-layer lift-off subsequently to replace the discarded etching step. Depending on the applications, an optional nanoimprint step could be included to obtain the reflective/transmissive mode design. The key steps to form the microstructures is via “additive” sputter deposition, instead of “subtractive” etching. The deposition rate is very linear with deposition time, and nearly independent of feature size, so the thickness can be time-controlled very well to achieve nanometer accuracy.

An overview of the proposed additive lithographic fabrication pipeline is shown in Fig. 2. Specifically, the workflow consists of several iterative patterning steps (a) and an optional nanoimprint step (b). In the patterning steps, the substrate (FS) is first coated with the target material (TM) as a base layer, where TM is typically a metal such as Cr or Al. We adopt the bi-layer lift-off method for easier operation, so a stack of Lift Off Resist (LOR) layer and PR layer are spin-coated successively. In the UV exposure, the dimensions of the design patterns are transferred to the top PR layer as usual, while the LOR exhibits slightly larger opening areas because the LOR has lower contrast and is more isotropically developed. Therefore, a re-entrant profile is created in the two stacked layers after development [13,14]. The difference in the critical dimensions in these layers is called undercut. To ensure sufficient space between the two layers and a discontinuity in the deposited materials between the substrate and PR, while the smallest features are still supported from the bottom in the LOR layer, the undercut should be characterized and optimized to the best range. On one hand, a large undercut may cause fine structures to disappear in LOR and small features in PR to collapse. On the other hand, a small undercut would not be able to create sufficient separation between the deposited TMs, so lift-off may fail and the structures in the corresponding areas would not be created in the end. The optimal undercut depends on the exposure dose and development time, given a fixed thicknesses of the two layers. Once the best re-entrant profile is obtained, a layer of TM is grown
onto the substrate in the opening areas through sputter deposition. The depth of the TM layer is controlled by the deposition time. Lastly, the auxiliary LOR and PR layers are removed by Dimethyl Sulfoxide (DMSO), leaving the required TM structures on the substrate. This process is iteratively repeated for multi-level structures. Figure 2a illustrates two cycles of the additive fabrication process for 4-level structures. With proper alignment, up to 16-level structures can be well fabricated.

In the case of reflective DOEs, the TM can be chosen as reflective materials, e.g., Al, and the fabrication workflow ends once the $N$-th iteration is finished. In the case of transmissive DOEs, one may deposit SiO$_2$ directly, but the deposition rate is very slow. Only thin structures can be fabricated this way. A more effective alternate is to first fabricate a hard stamp with Cr or Al, and replicate the stamp with transparent materials in a following nanoimprint step.

We use a variant of the UV nanoimprint technique, UV-NIL [15–17] for the pattern replication. The nanoimprint step begins with soft stamp preparation from the hard stamp, as shown in Fig. 2b. A glass substrate is cast with a drop of liquid UV curable flexible material (e.g., PDMS). It is then brought in direct contact with the hard stamp under pressure. The polymer chains are cross-linked when exposed under UV light, and the deformed material becomes solidified and glassy afterwards. The working soft stamp is then detached from the hard stamp, with an imprinted structured layer. To finally replicate the structures from the soft stamp to the product substrate, a fused silica substrate is prepared with liquid UV curable optical materials. Although various transparent resists (e.g., PMMA) are available for this purpose [18], the optical properties are of key concern in this regard. An excellent option for visible imaging applications is Norland Optical Adhesive (NOA). In the visible range, the refractive indices ranges from 1.36 to 1.64. Its excellent optical and mechanical properties have been demonstrated in widespread conventional optical applications. A few applications [19,20] indicate that, NOA is also suitable for patterning...
Fig. 2. Additive lithographic fabrication pipeline. (a) Iterative pattern transfer for additive multi-level DOE fabrication. The discarded RIE step in conventional lithography fabrication is replaced by additive sputter deposition and lift-off to form the desired 3D structures. (b) An optional nanoimprint step enables the replication of the multi-level structures from the hard stamp to the product wafer via a soft stamp, and imprinting onto UV curable optical adhesives on the substrate.

3. Results

We present necessary characterization tests, and four example designs to demonstrate the fabrication quality of the additive methods. Before the workflow starts, we need to measure the deposition rates for different target materials.

3.1. Deposition rates

Metal deposition is a matured technique, and can be well implemented by sputtering [21]. Considering the cost and availability of different metals, we choose Al for direct additive fabrication of reflective DOEs, and Cr as hard stamp fabrication for transmissive DOEs. The measured sputter deposition rates for Cr and Al are shown in Fig. 3. Both metals exhibit linear deposition in the experimental time window, whereas Cr deposition is approximately 2x faster than Al. With the tested deposition rate, we can define a basic deposition step, and duplicate it a few times when the desired deposition depth is thicker than the tested values.

3.2. Fabrication workflow

We summarize the complete workflow with detailed recipes in Table 1. Step 1 and Step 2 are pre-fabrication steps for wafer decontamination. We use piranha solution (a mixture of sulfuric
Fig. 3. Sputter deposition rates for Cr and Al. Both deposition rates show very good linearity in the test time range.

Table 1. Additive lithographic workflow

| Step | Work                      | Tools/Chemicals       | Recipe                      |
|------|---------------------------|-----------------------|-----------------------------|
| 1    | wafer cleaning            | Piranha solution      | 10 min at 115°C             |
| 2    | wafer drying              | wafer drier           | 7 min                       |
| 3    | base layer deposition     | sputter               | 100 nm                      |
| 4    | wafer dehydration         | hotplate              | 5 min at 200°C              |
| 5    | adhesion promotion        | HMDS vapor prime      | 20 min at 115°C             |
| 6    | LOR5B spin coating        | spin coater           | 0.6 μm, 1500 rpm            |
| 7    | soft bake                 | hotplate              | 3 min at 180°C              |
| 8    | AZ1505 spin coating       | spin coater           | 0.5 μm, 3000 rpm            |
| 9    | soft bake                 | hotplate              | 1 min at 100°C              |
| 10   | UV exposure               | contact aligner (EVG6200) | 9 mJ/cm²                 |
| 11   | development               | AZ726MIF              | 18 sec                      |
| 12   | metal deposition          | sputter               | time depends on thickness   |
| 13   | lift-off                  | DMSO                  | soak at 80°C                |
| 14   | sonication                | ultra-sonicator       | 5 - 10 min                  |
| 15   | wafer cleaning and drying | acetone and nitrogen gun | manual cleaning         |
| 16   | repeat Steps #4 - #15 for multi-level structures |                        |                            |

acid and 30% hydrogen peroxide solution) at 115 °C for 10 min of cleaning, and a spin rinse drier for 7 min of drying. A base layer is sputter deposited on the cleaned substrate. A thickness of 100 nm is usually sufficient for this purpose. The sputter deposition is done in a vacuum chamber with 5 mTorr pressure. Argon flow is 25 sccm with a DC power of 400 W. Deposition time is
calculated according to the measured deposition rates shown above.

Before spin-coating the LOR and PR layers, the wafer is dehydrated on a hotplate at 200 °C for 5 min in Step 4, followed by adhesion promotion with Hexamethyldisilazane (HMDS) vapor prime at 115 °C for 20 min in Step 5. The HMDS step is optional, but we find it helpful to create fine features as small as 2 μm. The LOR family has a variety of resists. We choose LOR5B (Kayaku Advanced Materials, Inc.) as its undercut rate is moderate compared to others in the series [22], and since it can be properly tuned with the PR used in the following step. The spin speed is set as 1500 rpm in Step 6 to have a thickness of 0.6 μm. The LOR thickness should be controlled larger than the maximum TM thickness in subsequent steps. The LOR layer is soft baked on a hotplate at 180 °C for 3 min in Step 7. The soft bake temperature and time are critical for the proper undercut. These parameters are fine tuned such that 2 μm features can be well maintained. In Step 8, a photoresist AZ1505 is spin coated at 3000 rpm for 0.5 μm, followed by another soft bake at 100 °C for 1 min in Step 9.

In Step 10, the UV exposure is performed on a EVG6200 contact aligner with a dose of 9 mJ/cm² in hard+vacuum mode. The LOR and PR bi-layer is developed for 18 sec in AZ726MIF developer (2.38% TMAH in H₂O) before cleaning in Step 11. Some TMAH based developers may attack Al, so AZ developer is preferred for minimal Al etching [14]. With the LOR5B we use, however, we find AZ726MIF performs best in dissolving LOR5B in a short period (< 18 sec) along with AZ1505 for high resolution patterning. Negligible Al attack is observed with this structure. The re-entrant structure is now ready for sputter deposition in Step 12, which has the same parameters as in Step 3. The deposition time is tailored for specific thickness according to the deposition rates.

The residual metals and auxiliary resists are removed in Dimethyl Sulfoxide (DMSO) soak at 80 °C in Step 13. The lift-off period depends on the metal type. Experimentally we find that, Cr can be lifted off easily within 10 min, while Al needs longer time to complete lift-off, typically a few hours or even overnight. Step 14 is an optional sonication step to remove residual metals in the lift-off step, and should be done gently for 5-10 min to prevent metal re-deposition onto the sample surface. Finally the wafer is cleaned and dried with acetone and N₂ manually.

The procedure from Step 4 to Step 15 is a basic additive fabrication cycle. They can be repeated N times to achieve 2ᴺ-level structures. Practically, the maximal number of N is determined by the pattern visibility under the microscopy, and the requirement for alignment tolerance. We find that the alignment from layer to layer becomes more difficult when N > 4, as the alignment error would not be well controlled to be within 1 μm. In our current workflow, the limiting number of iterations is N_max = 4.

3.3. Fabrication examples

We present two groups of typical reflective and transmissive DOEs to demonstrate the fabrication quality. In each group, we show two popular examples for widespread optical applications. The reflective DOEs are fabricated on Al using only the additive workflow in Table 1. The transmissive DOEs are fabricated first on Cr as the hard stamp, and then are replicated by nanoimprint on NOA63 (Norland Products, Inc.).

**Reflective Tilt-Gaussian-Vortex phase.** The first example is a reflective DOE for an astronomy application. In the upgrading Gemini Planet Imager calibration unit (CAL2.0), a scientific goal is to validate the Fast Atmospheric Self coherent camera Technique (FAST) [23]. A reflective Tilt-Gaussian-Vortex (TVG) focal plane mask [24,25] is required on the focal plane. The mission of the TVG is to send most of the rejected starlight into the off-axis reference beam pinhole, so the TVG phase consists of a central tilted Gaussian phase, and a background Vortex, as shown in Fig. 4a. The phase function [23] is

\[ \phi_{\text{TVG}}(x, y) = T + G + V, \]  (1)
where
\[
\begin{align*}
T &= 3.16\xi_0 (x \cos \theta_0 + y \sin \theta_0), \quad r < e, \\
G &= g \exp \left(-\frac{1}{2} \left(\frac{r}{\sigma}\right)^2\right), \quad r < e, \\
V &= l_p \theta, \quad r > e,
\end{align*}
\]

where \(r = \sqrt{x^2 + y^2}\), and \(\theta = \tan^{-1}(y/x)\). \(\xi_0\) and \(\theta_0\) are the tip/tilt parameters. \(g\) and \(\sigma\) are the Gaussian amplitude and width parameters. \(l_p\) is the topological charge of the Vortex. \(e\) is the radius of the Tilt-Gaussian region.

The unique challenge in fabricating this reflective phase is the simultaneous presence of micrometer (Tilt-Gaussian) and millimeter (Vortex) features. In the 16-level discretization, the 4 fabrication master masks, from coarse (L1) to fine (L4), are shown in Fig. 4b. With traditional RIE fabrication, it is challenging to etch the Vortex part uniformly in high micro-surface quality, due to the drastic radially expanding structures from the center to the edges. On the contrary, the proposed additive fabrication method is capable of maintaining all the structures for both the Tilt-Gaussian and the Vortex phases. The TGV phase is designed at the wavelength of \(\lambda = 1.3 \mu m\), so the nominal maximum height is \(h_{\text{max}} = \lambda/2 = 650\) nm for the continuous profile, and \(\tilde{h}_{\text{max}} = 15h_{\text{max}}/16 = 609\) nm for the 16-level discretization. The radius of the central Tilt-Gaussian region is 65 \(\mu m\). To achieve as high reflectivity as possible, we choose Al as the TM. The 3D measurements on a Zygo profilometer (NewView 7300) are shown in Fig. 4c (20x objective) and 4d (5x objective). It is clear that the high resolution Tilt-Gaussian region retains its fine structures, and the Vortex region is reproduced smoothly in the flat areas across the fabrication area of 3 mm \(\times\) 3 mm. The deposited Al heights from L1 to L4 are 41 nm, 81 nm, 163 nm, and 325 nm respectively. The crosslines in the central Tilt-Gaussian region and the Vortex region are shown in Fig. 4e and 4f to illustrate the actual total heights after fabrication. The fabrication error is controlled within 1% of the nominal height.

**Reflective Computer Generated Holography.** Holograms can be generated with DOE in many imaging and display applications. This kind of DOEs are often designed with the Gerchberg-Saxton (GS) algorithm [26, 27] in an iterative scheme. Given a displayed intensity pattern, the GS algorithm repeatedly transforms the amplitude and phase from the spatial domain to the Fourier domain to search for a possible DOE phase. Owing to the unconstrained optimization, such DOEs usually contain structures with high spatial frequencies. Here we show an exemplary reflective DOE for CGH fabricated on Al. The design wavelength is at \(\lambda = 1.3 \mu m\) (as above). Each pixel on the DOE is 5 \(\mu m\), and the dimension is 8 mm \(\times\) 8 mm. The optimized pattern is shown in Fig. 5a, with the fine features shown in the zoom-in region. The fabricated samples are measured on the Zygo profilometer with 20x objective (Fig. 5b) and 50x objective (Fig. 5c) to illustrate the “random” structures in different scales. A 3D height map of the local features is shown in Fig. 5d. The additively fabricated 16-level DOE can reproduce the designed CGH structures reliably.

**Transmissive Fresnel lens.** The third example is the transmissive diffractive Fresnel lens, which is a popular component in many diffractive optics applications. Variants of the Fresnel lens have been designed to work together with computational algorithms for novel imaging and display applications in recent years [3, 4, 28]. Here we show the fabrication results for both positive and negative Fresnel lenses to demonstrate the performance of fabricating such rotational symmetric structures.
The phase profile of a Fresnel lens is

$$\phi_F(x, y) = \text{mod} \left( \frac{2\pi}{\lambda} \cdot \frac{(x^2 + y^2)}{2f} \cdot 2\pi \right), \quad (3)$$
where $f$ is the focal length, and $\lambda$ is the design wavelength. We first fabricate a hard stamp on Cr, and then transfer the structures onto NOA63. Therefore, the stamp heights are determined by the refractive index of the final material NOA63 ($n = 1.56$) and the operating wavelength. Here we design a Fresnel lens at $\lambda = 610$ nm for a focal length of $f = 90$ mm. The maximum height on NOA63 is $h_{\text{max}} = \lambda/(n - 1) = 1089$ nm for the continuous profile. In the fabricated 16-level form, the deposited Cr heights on the hard stamp are 68 nm, 136 nm, 272 nm, and 545 nm respectively. In Fig. 6a and 6b, the fabricated 16-level height profiles on the Cr hard stamps are shown for two Fresnel lenses with focal lengths of 90 mm and -90 mm. The diameters of the apertures are 4 mm in both cases. After nanoimprint, the patterns are successfully transferred to NOA63, as shown in Fig. 6c and 6d respectively.

**Transmissive cubic phase.** Cubic phase is one of the first optical elements used for extended depth-of-field imaging applications [29]. Its characteristic is the combination of low frequency and smooth yet curved high frequency patterns from the center to the edge.

The cubic phase is expressed as

$$
\phi_C(x, y) = \text{mod} \left[ \alpha \left( x^3 + y^3 \right), 2\pi \right],
$$

where $\alpha$ is a control parameter for phase deviation. In this example, we use $\alpha = 10^{-9}$, at the design wavelength of 610 nm. The discretized 16-level height map is shown in Fig. 7a.
3D structure in the curved area on the Cr hard stamp is shown in Fig. 7b under 5x objective in the Zygo profilometer. The staircase fine features are clearly visible. In the nanoimprint step, the patterns are then replicated on NOA63, as shown in Fig. 7c. Note that the difference in the presented structures in Fig. 7b and Fig. 7c is owing to the fact that they are taken from similar regions on the Cr and NOA samples, but not the same.

3.4. Discussion

The advantages of the proposed additive fabrication methods lie on three aspects. First and foremost, the additive process offers a unique uniform patterning capability for feature sizes ranging from micrometers to millimeters at the same time. It makes it possible to fabricate optical devices that are too challenging for RIE based fabrication. Second, the microsurface roughness can be controlled very well due to the high uniformity in the deposition step. Therefore, the smoothness in the flat regions is better than in the etching methods. Improved roughness helps reducing the scattering effects in optical applications, thus higher diffraction efficiency can be maintained. Third, the additive hard stamp is a good resource for the nanoimprint process, so mass production is easily achievable with the proposed methods. The proposed methods can also be generalized with other deposition and nanoimprint methods to substitute those in the proposed workflow. For example, sputtering deposition can be used in combination with e-beam evaporation, and thermal embossing can be used as an alternative for UV nanoimprint.

A few drawbacks still exist in the current DOEs fabricated with our methods. Some nanometer thin spikes are present around the edges, especially in the third and fourth layers when the target
height goes larger and larger. This could be magnified by the nanoimprint step and becomes visible, as shown in Fig. 6c and 6d. This may arise in the lift-off step from some residual thin metal film remaining on the PR side walls, and the thickness difference between LOR and TM is narrow. A possible mitigation would be to use thicker negative PR instead of the bi-layer structure. Second, both the lateral and vertical edges in the deposited metal layer may suffer from roundness in the corners. In the 50x measurements of the CGH example, the round corners are visible in Fig. 5b-d. The lateral corners may arise from the photolithography step. Sharp edges are rounded due to diffraction during the projection from the master mask to the PR. The vertical corners are from the bi-layer deposition step, in which some TMs stay on the side walls on the PR. This effect is not necessarily a flaw for DOEs. Since the 16-level discretization is an approximation of continuous profiles itself, a sharp corner is not the intended way from the first place. The rounding effects, however, may serve as a smooth transition from level to level. Last, experimentally we find that the final NOA sample may sometimes be peeled off along with the intermediate stamp from the substrate locally, or completely, if the NOA does not stick to it firmly. This would require adhesion promotion resist before the NOA is coated onto the wafer. Other candidates of the final substrate material with better mechanical properties could be explored depending on the applications.

4. Conclusion

We have experimentally demonstrated a class of additive lithographic fabrication methods by replacing the conventional etching steps with sputter deposition and lift-off. Instead of removing
substrate materials, the proposed methods selectively grow target materials from bottom up onto the substrate. The fruitful benefits of additive fabrication overcome the inherent drawbacks and artifacts that frequently occur in conventional RIE. The proposed methods are suitable for both reflective and transmissive DOE fabrication. An optional nanoimprint step is compatible with the additive fabrication routine, making it possible for reflective/transmissive mode transform, as well as mass production. We present the fabrication workflow with detailed recipes, and showcase four fabrication examples in which spatially varying features co-exist in the design. The results demonstrate the ability of the proposed additive fabrication methods to produce uniform structure depths, and low surface roughness. The current parameters are fine tuned to achieve spatial resolution of 2 μm for up to 16-level DOEs. We envision the additive fabrication methods to be alternative processes to complement the etching based methods for DOE fabrication in particular cases where spatially varying features are of great concerns.

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**Data availability.** Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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