Shapeshifting Diffractive Optical Devices

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In optical devices like diffraction gratings and Fresnel lenses, light wavefront is engineered through the structuring of device surface morphology, within thicknesses comparable to the light wavelength. Fabrication of such diffractive optical elements involves highly accurate multistep lithographic processes that in fact set into stone both the surface morphology and optical functionality, resulting in intrinsically static devices. In this work, this fundamental limitation is overcome by introducing shapeshifting diffractive optical elements directly written on an erasable photoresponsive material, whose morphology can be changed in real time to provide different on-demand optical functionalities. First a lithographic configuration that allows writing/erasing cycles of aligned optical elements directly in the light path is developed. Then, the realization of complex diffractive gratings with arbitrary combinations of grating vectors is shown. Finally, a shapeshifting diffractive lens that is reconfigured in the light-path in order to change the imaging parameters of an optical system is demonstrated. The approach leapfrogs the state-of-the-art realization of optical Fourier surfaces by adding on-demand reconfiguration to the potential use in emerging areas in photonics, like transformation and planar optics.

1. Introduction

Diffraction gratings[1] are among the first optical devices ever realized. Light structuring through gratings results from the periodic phase modulation accumulated across the grating profile.[2,3] A diffraction grating made of periodically spaced grooves produces, far from the grating, a line of light dots (diffraction orders) along a direction perpendicular to the grating grooves (grating vector). Although simple, diffraction gratings represent the first example of structuring light by means of an engineered phase modulation.[4] This concept is at the core of holography and Fourier optics (when the field structuring can be considered as only due to the propagation of component plane waves).[3] Moreover, phase masks can be multiplexed, so gratings with different Fourier components can be combined together to achieve more complex bi-dimensional light structuring.[5,6]

Diffraction gratings represent also the first example of planar optics.[7,8] For a grating with sinusoidal profile made of a 1.7 refractive index transparent material, the maximum diffraction efficiency is achieved with a morphology modulation of 0.84 times the illuminating light wavelength (i.e., only 531 nm thickness for an illuminating light of 633 nm).[4]

Other diffractive optical elements (DOEs) of common use are Fresnel lenses, in practice, cylindrically symmetric gratings optimized to focus collimated light at a design distance.[9] Such lenses, used for instance in lighthouses, become handy to reduce space and weight constraints related to standard refractive lenses.[10–12]

Once the optical parameters of the material are fixed, the optical functionality of a DOE is controlled through the device morphology, in case of a grating: spacing, height and profile of the grooves. Such design principle has been continuously developed for DOE manufacturing, resulting in optimized...
elements for specific applications. An interesting discussion about advanced diffraction elements for spectroscopy is provided in a recent review. However, most of the standard DOE manufacturing techniques consist of at least two steps, typically a molding process, that involves the patterning of a lithographic material with the target surface geometry, followed by a development stage requiring a selective etching of a substrate, along with an eventual additional surface coating step in case of reflective DOEs. Direct machining technologies, instead, allow direct DOE fabrication by selectively grooving a metallic or dielectric plate through ruling engines or ion beam etching, but they provide coarser surface profile control. Only recently, a direct grayscale lithographic method based on thermal scanning probe lithography has been successfully employed to realize accurate complex diffractive surfaces on a polymer film. All the mentioned fabrication approaches, however, result in static DOEs, with surface geometry and optical functionality defined once for all during device manufacture. Here we report, instead, the direct realization of planar diffractive optical elements whose surface morphology can be changed in real time to provide different on-demand optical functionalities.

Phase and amplitude masks for diffractive optics can be also realized with different approaches. On one hand, there are fast but pixelated modulators, spanning from commercially available liquid-crystal-based spatial light modulators (SLMs) or digital mirror devices to recent 2D materials devices. On the other hand, in the last ten years, research on optical metasurfaces exploded, providing new devices with unprecedented modulation possibilities and resolution but, in fact, much limited in terms of optical tunability.

In this work, we develop a grayscale holographic photolithography scheme employing a reconfigurable polymer to realize diffractive optical elements that can be reshaped directly in the light-path to provide a specific optical functionality; in fact, shapeshifting diffractive optical elements that do not need any additional lithographic process (e.g., resist development, substrate etching, surface coating, etc.) for tunable and reprogrammable practical operation. Complex gratings are showed together with a reconfigurable monochromator. Finally, variable focal lengths diffractive lenses are demonstrated that allow magnification tuning of a complex optical system.

2. Results

The central equation to design within the realm of the scalar Fourier optics a phase mask \( \varphi(x, y) \) for the operation of a transmissive DOE is\(^{1,4} \)

\[
\varphi(x, y) = k_0 (n - n_0) h(x, y)
\] (1)

Once the refractive indices of the grating material (\( n \)) and the surrounding material (\( n_0 \)) are fixed, together with the illuminating light wavelength \( \lambda_0 \) (wavevector \( k_0 = 2 \pi / \lambda_0 \)), the phase mask, and hence the output diffracted field from the DOE, is entirely defined by the surface morphology \( h(x, y) \). For an operating DOE, the surface profile needs to be faithfully transferred on a dielectric substrate using accurate lithographic processes. To this aim, we use here a polymer containing azobenzene molecules (or simply azopolymer) that requires only a single photostucturation step to directly realize the working phase mask (details about the polymer can be found in the Experimental Section). When illuminated by a specific light pattern, the surface of an azopolymer film is directly deformed by a reversible light-induced material transport, resulting in a morphology that follows the illuminating light distribution. For example, illuminating such polymer with a 2D light pattern with sinusoidal profile, results in a surface relief with similar profile, i.e., a diffraction grating (see also Figure S4, Supporting Information). In this case, the periodic light distribution can be as simple as that produced by two interfering light beams. The same configuration has been demonstrated to be suitable also for the direct realization of more complex multiplexed gratings. In our experiment we used, instead, a holographic setup that projects arbitrary grayscale light patterns on the polymer surface to realize the profile of complex DOEs. With our system, the surface morphology is analytically calculated and converted in a grayscale bitmap image that defines the holographic light pattern irradiated on the polymer surface for any designed DOE. Such light patterns can also be changed in time by simply updating the bitmap image with a new and completely independent one, making the system ready for dynamical surface structuration of a photo-convertible material as the azopolymer we use. The details of our experimental setup can be found in the Experimental Section.

Figure 1 shows the principle of dynamical holographic surface structuration process by realizing, as a first simple DOE, a transformable (by light) sinusoidal diffraction grating (see Figure S2, Supporting Information). The writing beam is a holographically patterned laser beam with a wavelength of 491 nm (circularly polarized). The generated light pattern is projected onto the polymer film surface by means of a 50X long working distance objective (Figure 1A). The formation of the diffraction grating on the surface can be monitored in real time by detecting the light power in the first diffraction order of a probe laser beam at wavelength of 633 nm, collimated through the same objective. Figure 1E shows the rising diffraction efficiency during the writing process (see also Movie S1, Supporting Information). In our configuration, another circularly polarized laser beam with wavelength of 405 nm also illuminates the grading area, from the substrate side. The function of the 405 nm beam is twofold (see also the Experimental Section): at low power density (\( \approx 0.5 \text{ W cm}^{-2} \)) it keeps active the photosensitive molecules in the polymer, avoiding partial saturation, assisting and speeding-up the writing process;\(^{45-47} \) at high power density (\( \approx 1.0 \text{ W cm}^{-2} \)), the violet beam erases the polymer surface\(^{48-51} \) (see also Figure S5, Supporting Information), that is then ready to write the next DOE. It is important to note that, once the writing process is concluded, the realized DOEs are still stable for years at room temperature and ambient illumination conditions, due to structural stability of the patterned surfaces and the absence of preferential orientation of the photosensitive molecules during the writing (see also Figure S6, Supporting Information). Figure 1F,H shows the atomic force microscope (AFM) micrograph of the polymer film surface after writing and after erasing, respectively. Figure 1E also shows the decreasing of the diffraction efficiency over the erasing process: both the morphology (see also Figure S7, Supporting Information) and diffraction residuals are negligible, demonstrating fast (\( \approx 10 \) s) and complete surface erasure. At this point, a new DOE can be realized on the film surface; for instance, a new grat-
Figure 1. Direct all-optical realization of reconfigurable diffraction gratings. A–D) Schematic illustrations of optical configurations used for the writing, erasing, and the real-time monitoring of dynamically reconfigurable diffraction gratings. In the writing steps A,C), the writing beam (light blue color) illuminates a photoresist area (≈200 μm diameter) with holographically-controlled sinusoidal intensity patterns through a 50X (NA = 0.55) microscope objective. A collimated beam (λ = 405 nm) is used at low intensity to enhance the surface structuration dynamics (light violet beam) and at higher intensity (dark violet beam) in the erasing steps B,D) when the writing beam is switched off. E) Time-evolving first-order diffraction efficiency curves recorded by two photodiodes (PD1 and PD2 shown in panels A and C) for a He–Ne probe beam (red beam) during the reconfiguration of two gratings of different periodicity (green curve for G₁ and orange curve for G₂). F–M) AFM micrographs and relative horizontal topographic profiles of the surface at the instants (tᵢ) of the time sequence in E). Scale bars in AFM images are 10 μm. N–R) Dynamical tuning of diffraction dispersion obtained through multiple grating periodicity reconfigurations. Top panels show optical micrographs of the surface (scale bar 10 μm). The colored diffraction patterns produced by the surface from a white LED source are imaged by a CCD camera in the surface Fourier conjugate plane. A fixed aperture on the CCD camera intercepts color bands of shifting central wavelength.

As next step, we tested the possibility of having reconfigurable DOEs in the light-path of an operating optical device. To this aim, we realized the proof-of-concept of a reconfigurable monochromator (see also Movie S2, Supporting Information). In this case, the light diffracted by the grating is detected by means of a CCD camera. The probe beam is a white light beam provided by a LED. Figure 1N–R shows the effect of reshaping the grating periodicity. As expected, the smaller the grating pitch, the higher the diffractive power and the separation between the light spectral components. In this case, changing the DOE directly in the light-path allows real-time tuning of the spectrometer resolution through the grating dispersion. In the same configuration, fixing an aperture on the camera (same as using a physical aperture and a point detector) allows to observe how the wavelength diffracted...
in the detection area changes as a function of the realized grating. This last configuration works in fact as a tunable monochromator, without any moving part, that can be used together with a broadband source to select an illuminating wavelength band while preserving the original alignment of the entire system.

Another unique characteristic of our lithographic approach stands in the noise reduction of the holographic light pattern and the resulting enhanced quality of the morphology produced on the azopolymer surface. Usually, light structuring through digital holography suffers of a random speckles distribution that degrades the contrast and limits the achievable gray levels in the projected light pattern, also compromising the definition of small features. To minimize such effects, we continuously refresh the light pattern at a refresh rate of 20 Hz. Each new light pattern has a random speckle distribution that averages down during the writing time, improving in fact the quality of the structured polymer morphology.[44] For instance, in order to realize a grating with a single vector (like that of Figure 1F–J), we expose the
Diffractive lens. A) Target surface (normalized) of a Gabor phase zone plate, designed to focus light of wavelength $\lambda = 633\,\text{nm}$ at distance $f_0 = 500\,\mu\text{m}$ from the surface. B) Scanning electron micrographs (SEM) of the holographically structured photoresist surface. C,D) 3D views for a section of the target C) and AFM measured D) surface. E) Target and experimental profiles traced along the radial directions shown in C) and D), respectively. F–I) Comparison of the normalized axial F) and transverse G) Point-Spread Functions (PSFs), simulated for the target lens profile in A) through Fresnel diffraction integral, and experimental axial H) and lateral I) PSFs produced by the experimental surface in B) under illumination with a collimated beam at design wavelength. J) Comparison of simulated and experimental focusing diffraction efficiency as function of average surface modulation amplitude $h$ (defined in E)).

This approach can be used for example to realize, with an identical experimental configuration as the previous one, DOE with any distribution of grating vectors, directly encoded in the analytical design of grayscale digital holograms (see also Figure S9, Supporting Information). Figure 2 shows some examples. First a RGB grating, designed as even superposition of three grating vectors (Figure 2A), is realized (Figure 2B) to diffract three different wavelengths ($\lambda_1 = 633\,\text{nm}$, $\lambda_2 = 532\,\text{nm}$, and $\lambda_3 = 488\,\text{nm}$) into the same diffraction order. The surface profile (Figure 2C), realized on the polymer film in the single grayscale holographic exposure step, faithfully match the targeted DOE profile (see also Figure S9, Supporting Information), that correctly produce the designed white diffraction spot (Figure 2D).

Then, the same design can be extended even to more complex Optical Fourier Surfaces. Figure 2F shows a 2D quasicrystal structure characterized by 6 grating vectors $g_i$, differently oriented in the transverse plane (Figure 2E). Figure 2H shows the effect of having same vectors orientation as Figure 2F but different vectors lengths (different periodicity) in the designed superposition of sinusoidal functions. This results into a spiral quasicrystal DOE, whose fabrication would be very demanding in standard sequential interference lithography.

Additionally, Figure 2I-L shows the realization of a blazed grating that can diffract a probe beam into a preferential diffraction order. In this case, the blazed structure results from combining 6 gratings vectors with same direction but different lengths and weighted amplitudes. Besides being reconfigurable, DOE structures like those showed in Figure 2 are comparable with the state of the art of the most recent multiplexed structures obtained by static and serial lithography techniques.

Related to the quality of the obtained structures, it is to be considered that the polymer that we used is fully compatible with a further lithographical step, where the obtained morphology...
can be transferred to a PDMS stamp for high quality replicas (see also Figure S9, Supporting Information) that provides our process with technological significance as new photolithography technique to be applied to optical systems as well as functionalized surfaces and microfluidics.

To further highlight the potential of our approach, in Figures 3 and 4 we show the realization of reconfigurable diffraction lenses working in real imaging systems. DOEs with such profiles are known as Gabor phase zone plates \(^{53}\) (details about such design can be found in the Experimental Section) and have been out of reach of previous experiments on azopolymers.\(^{54,55}\) Figure 3A shows the computed profile for a lens with 500 μm focal length and numerical aperture NA = 0.19. The light distribution in the yz plane around the focal position can be calculated through the Fresnel diffraction integral \(^{3}\) (see also the Experimental Section) The calculated diffraction limited spot of this lens has FWHM of \(\approx 1.7 \mu m\) when illuminated by a beam with 633 nm wavelength (Figure 3F). The simulated features are well reproduced in the measured intensity distribution (Figure 3H,I), obtained from the experimental diffractive lens shown in Figure 3B. Figure 3J also compares the predicted and realized efficiency for this DOE as a function of average grooves height, achieved in different ex-
posure times. For this planar lens, the predicted maximum efficiency of ≈34% is achieved in 50s exposure (see also Figures S10 and S11, Supporting Information).

Figure 4 demonstrates the possibility of re-shaping our diffractive lens in real-time in the aligned optical system. Figure 4A–C shows three lenses of different focal length realized one after the other in the same polymer area. The respective experimental focal field distributions are reported in Figure 4D–F as the focal length increases according to the design, the numerical aperture decreases and the focused light spot increases according to the fixed diameter of our lenses (see also the Experimental Section). Notably, the focal length reconfiguration resulting from the surface transformation demonstrated in Figure 4D–F, achieves about 70% shift of the focal position, without involving any mechanical movement in the system. This large deformation range is comparable to electrically tunable LC-based DOE lenses, characterized by a faster switching speed, which comes at the expenses of complex electrode configurations, reduced spatial resolution and significantly larger thicknesses with respect to our flat diffractive devices.

The tunable shapeshifting lenses demonstrated by means of our approach can be used to realize an imaging system able to dynamically provide different magnifications of extended scenes. Standard zooming systems require the axial movement of at least two lenses to produce a magnified image in the camera plane. In our system instead one of the mechanical motions can be replaced by the lens reconfiguration into the needed new optical element (see also Figure S12, Supporting Information). This is evident in the magnified images of our Institutions logos presented in Figure 4G–N. In this case, one lens of the optical system is physically shifted to regain the focal position while changing on-demand the focal length of the reconfigurable diffractive lens. The observed magnification factors reproduce the expected values from focal length ratios: \( f_1/f_\text{L} = 1.2 \) in Figure 4I,J; \( f_1/f_\text{L} = 1.6 \) for Figure 4K,L; \( f_2/f_\text{L} = 2 \) for Figure 4M,N.

An important aspect to highlight is that our devices are just made of a structured surface, thinner than the illuminating light wavelength, on a polymer film spin coated on a glass coverslip. The photolithography process to obtain such lightweight and planar devices is fully scalable and compatible with raster scanning method for increased throughput and with curved substrates, opening new possibilities in functionalizing surfaces of objects as diverse as wearable items and vehicle parts.

3. Conclusions

In conclusion, we have proved that diffraction optical elements with efficiency equal to the theoretical efficiency can be fabricated by direct structuring of the surface of a photosensitive polymer, avoiding any further lithographic step. The realized gratings and lenses can be reshaped completely while aligned in the optical setup. Grating periodicity can be changed; lenses focal length can be tuned; one optical element can be morphed into another optical element with completely different optical functionality, without affecting the alignment of the specific optical setup. More than 100 years after Michelson was reporting about optical effects from imperfect gratings, we show that it is possible to realize optical elements with theory-matching efficiency and practical use, reconfigurable on demand right where and when needed.

4. Experimental Section

Azopolymer Synthesis and Characterization: The photoresponsive material used in this work is an azobenzene-containing polymer (azopolymer) in amorphous state. All reagents were purchased from Merck and used without further purification. The azopolymer was synthesized, purified, and characterized as previously reported (\( M_\text{n} = 70000 \); phase sequence: Glass 67 °C Nematic 113 °C (isotropic; \( \lambda_{\text{iso}} = 350 \text{ nm} \)) [26–28]. The solution for film deposition was prepared by dissolving (70 mg) the polymer in (0.50 mL) of 1,1,2,2-tetrachloroethane and filtered on 0.2 μm PTFE membrane filters. The desired film thickness (typically 1.0 ± 0.1 μm) was obtained by spin coating the solution on 24 x 60 mm cover slides at 300 rpm for 4 min. In the final stage, the samples were kept under vacuum at room temperature for 24 h to remove solvent traces.

Refractive index of the fabricated film was measured via ellipsometry. Measured values at some relevant wavelengths (633, 532, 488 nm) are: \( n_{633} = 1.70 \); \( n_{532} = 1.74 \); \( n_{488} = 1.78 \).

Holographic Illumination Setup: The experimental configuration for the azopolymer surface photopatterning is based on a phase-only Computer-Generated Holograms (CGHs) system. Its schematic representation is shown in Figure S1 (Supporting Information). A laser diode source (Cobolt Calypso) emits a TEM00 beam at wavelength \( \lambda = 491 \text{ nm} \) and, after a beam expander, is phase-modulated by a computer-controlled reflective phase-only Spatial Light Modulator (SLM, Holoeye Pluto). The modulated beam is propagated through a 4f lenses system with the input plane located in the SLM plane. The output plane coincides with the back focal plane of an infinity-corrected long-working distance 50X objective (Mitutoyo), with numerical aperture NA = 0.55. This configuration allows the reconstruction of a structured intensity pattern in the focal plane of the objective (where the azopolymer is placed).

Arbitrary intensity patterns can be generated imposing the proper phase profile (kinoform) for the beam in the SLM plane. The phase hologram is calculated according to the Fourier transform relations [14] existing between 4f system conjugate planes. The focal lengths of the lenses \( L_3 \) and \( L_4 \) (Figure S1, Supporting Information) are chosen in order to maximize the spatial resolution in the hologram reconstruction planes. [24,25] This choice also defines the diameter (≈200 μm) of the accessible circular area in the objective front focal plane, which can be used to structure the photosensitive surface in a single illumination step. The position of the sample near the objective focal region is accurately controlled by means of a x-y-z translation stage. Average intensity in the range 12.7–14.0 W cm\(^{-2}\) and circular polarization are used for the structuration of the azopolymer surface. For visual inspection, and proper focusing of the holographic pattern on the photosensitive surface, a beam splitter placed in the light path redirects the light retroreflected by the surface and re-collimated through the objective toward a tube lens. This lens forms an image of the holographic pattern in the second focal plane, where a CCD camera is positioned to observe the surface plane in real-time (Figure S1, Supporting Information).

When needed, an additional diode laser beam at 405 nm illuminates the photosensitive film from the substrate (glass microscope coverslip) side. The beam has circular polarization and is set on different intensity levels depending on its intended functionality. When the intensity is in the range 0.4–0.8 W cm\(^{-2}\), the beam favors the surface structuring process, acting as a writing assisting beam. At intensity higher than 0.9 W cm\(^{-2}\), its absorption causes the erasure of previously inscribed surface structures, acting as an erasing beam (see also Figure S5, Supporting Information).

For the DOEs with optimized diffraction efficiency demonstrated in Figure 3 (and Figure S4, Supporting Information) a total exposure time of 50 s is used over an area of 3.14 × 10\(^3\) μm\(^2\). This provides an estimation of processing throughput of our structuration method of ≈630 μm\(^2\) s\(^{-1}\).

Algorithm for Calculation of Phase-Only Holograms: The Iterative Fourier transform algorithm (IFTA) used for the calculation of the phase profile of the writing beam in the SLM plane is the mixed region amplitude freedom (MRAF) algorithm [17] based on an extension of the standard Gerchberg–Saxton (GS) algorithm [18]. The IFTA has been implemented in MATLAB, using Fast Fourier Transform (FFT). In the first step of the calculation, a target 8-bit grayscale image (see also Figure S2, Supporting Information), representing the desired intensity pattern (e.g., sinusoidal)
to be reconstructed in the polymer plane, is analytically defined. Next, with an available calculation loop, based on direct and inverse FFTs, the algorithm returns a grayscale (8-bit, 256 phase levels) image (1080 x 1920 px) for the phase profile (the koinform) to be imposed on the writing beam in the SLM plane. In the MRAF algorithm, the reconstruction plane is divided in two regions: one that is considered important, called signal region, that corresponds to the area of sample effectively illuminated; the region outside that domain (the noise region) is used to spatially redirect some of the calculation noise intrinsically related to IFTA methods (speckle, ghost hologram, etc.) in the periphery of the system field of view. A single parameter (the mixing parameter) in MRAF[57] controls the relative fraction of light power directed in each of the two domains of the reconstruction plane. An optimum value for the mixing parameter was selected to maximize intensity contrast in the target patterns. An iris placed in the intermediated hologram reconstruction plane (Figure S1, Supporting Information) is used to filter out the noise regions.

Optical Real-Time Characterization of Structured Surfaces: For real-time observation of the polymer surfaces during the structuration process, the holographic setup was integrated with a collimated white LED source for a bright-field transmission microscopy system (see also Figure S3A (Supporting Information)). Scattered light from the sample surface is collected by the objective. Real time image of the sample surface is obtained with the same configuration for holographic pattern acquisition. The retro refected holographic pattern, propagating along the same light path, can be eventually discarded in the imaging using a low pass filter before the camera.

For the collection of real-time diffraction patterns shown in Figure 1 and in Movie S2 (Supporting Information), a second beam splitter redirects part of the light emerging from the tube lens through a positive lens realizing a 2f configuration with the tube lens. A second CCD camera, positioned in the second focal plane of the lens, provides a Fourier transform image of the structured surface, corresponding to the far-field diffraction pattern.

A second configuration, shown in Figure S3B (Supporting Information), allows to produce real-time diffraction patterns from photoresist structured surface using a He–Ne as probe beam, whose wavelength (\(\lambda = 633 \text{ nm}\)) lies outside the absorption band of the azopolymer[25,23] not interfering with the writing process. The probe beam is focused in the back focal plane of the objective using the beam splitter and the tube lens. The collimated beam, emerging from the objective, illuminates the same photoresist area simultaneously structured by the holographic writing beam. For the time-dependent analysis of diffraction efficiencies produced by 1D diffraction gratings (Figure 1E), photodiodes are used to measure the light power of the probe beam diffracted in far-field in the first diffraction order.

Morphological Characterization of Structured Surfaces: Topographic characterization of polymer surfaces is performed using AFM and SEM.

For AFM measurements, a WITec AlphaRS300 microscope is used. The AFM is operated in tapping mode using a cantilever with 75 kHz resonance frequency and nominal force constant of 2.8 N m\(^{-1}\). AFM tips (Arrow FM type from Nano World), with nominal radius of curvature of \(\leq 10 \text{ nm}\), are used in all the experiments. The maximum scanned area has a size of 100 x 100 \(\mu\text{m}\)^2.

Scanning electron microscope images are acquired with a field-emission gun (FEG-SEM) FEI/Thermo Fisher Nano NanoSEM 450 microscope. Samples are sputtered with a layer of Au/Pd using a Denton Vacuum Desk V TSC coating system prior to observation.

Design and Diffraction Properties of Complex Diffraction Gratings: The scalar wavefront modulation provided by thin transmissive DOEs can be described by a phase-only space-dependent complex transmission function \(t(x,y)\), able to modulate an incident light field. For an incident monochromatic plane wave (at wavelength \(\lambda\), propagating through the structured DOE layer, the modulation function is

\[
t(x,y) = e^{i\phi(x,y)} = e^{i\phi(x-n\lambda)}(x, y)
\]  

(2)

The phase modulation \(\phi(x,y)\) is the result of the differences in the optical path that the incident monochromatic plane wave accumulates with respect to the surrounding medium (air in the case \(n_i = n_0 = 1\)) as the wave propagates in DOEs, medium of refractive index \(n\). In Equation (1), \(k = 2\pi/\lambda\) is the wavevector of the incident plane and the surface relief geometry is described using a normalized structural function \(s(x, y)\) (with \(s(x, y) \in [0, 1]\)), defining the DOE pattern, and the total relief amplitude \(h\), which defines the maximum phase modulation depth provided by the DOE.

For the complex diffraction gratings reported in Figure 2, the structural function \(s(x, y)\) is designed as sum of \(N\) sinusoidal patterns

\[
s(x, y) = \frac{1}{2\pi} \sum_{m=0}^{N-1} A_m \left( 1 + \sin \left( \frac{2\pi x}{\Lambda_m} + \frac{2\pi y}{\Lambda_m} + \varphi_m \right) \right)
\]  

(3)

Here, \(\Lambda_m = (\Lambda_{m1}^2 + \Lambda_{m2}^2)^{1/2}\) is the spatial periodicity of the \(m\)th sinusoidal pattern, characterized by a grating vector oriented at the angle \(\gamma_m = \arctan(\Lambda_{m1}/\Lambda_{m2})\) with respect to the x direction, \(A_m\) is the amplitude weight for each sinusoid in the superposition, \(\Lambda_m\) and \(\varphi_m\) are relative phases between sinusoids. The field diffracted by a phase mask described by Equations (1) and (2) can be obtained by expanding the transmission function \(t(x,y)\) in Fourier series

\[
t(x,y) = e^{i\phi(x, y)} = \sum_{m=0}^{\infty} \sum_{q=0}^{\infty} f_{mq} e^{i(k_{mq} x + q_{mq} y)}
\]  

(4)

where \(f_{mq}\) are the complex Fourier coefficients and \(g_{mq} = 1/\Lambda_m\) and \(g_{mq} = 1/\Lambda_{mq}\) are the coordinates in the Fourier space. The solution of the scalar Helmholtz equation for a plane wave of amplitude \(U_0\), incident on DOE surface with and angle \(\theta_i\), is[31]

\[
U(x, y, z) = U_0 \sum_{m=0}^{\infty} \sum_{q=0}^{\infty} f_{mq} e^{i(k_{mq} x + q_{mq} y)}
\]

\[
\times e^{-ik_0 z} \left( k_{mq} \sin \vartheta_i \frac{\Lambda_{mq}}{\Lambda_m} + q_{mq} \right)
\]

(5)

Only a finite number of diffraction orders, (defined by the indices \((m, q))\) which make the quantity \(k_0^2 - (k_{mq} \sin \vartheta_i) + n \frac{2\pi}{\Lambda_m} + n \frac{2\pi}{\Lambda_{mq}} > 0\) (depending also on the periodicities of the superimposed sinusoids in \(s(x, y)\)), propagates in far-field from the surfaces as plane waves. All the other possible (infinite) orders give rise to evanescent waves localized at the DOE surface. Each term of the expansion (4) carries a power \(P_m = U_0^2 |f_{mq}|^2\), which also defines the diffraction efficiency \(\eta_m = |f_{mq}|^2\) for propagating orders.

For the realization of the complex diffraction gratings reported in Figure 2, relation (2) was used to analytically design the surface pattern \(s(x, y)\). This pattern is encoded in a grayscale 8-bit digital image (1080 x 1920 px and 256 discrete intensity levels in the interval [0 255]), used as target image in the MRAF algorithm for hologram calculation. The parameters \(\Lambda_m\), \(\varphi_m\) and \(\gamma_m\), used for the design of the surfaces reported in Figure 2 are summarized in Table S1 (Supporting Information).

Design of Gabor Phase Zone Plates: A Gabor phase zone plate[31] is a diffractive phase mask, designed to focus light at a specific distance. The radial profile \(h(r)\) of a surface relief acting as a Gabor phase lens is

\[
h(r) = \left( 1 + \cos \frac{\arctan^2 r}{2} \right) h
\]  

(6)

where \(h\) is the relief amplitude and \(r\) is a structural parameter that sets the lateral size of the relief, defining the focal distance \(f\) on the optical axis. The relation between the size parameter \(r\) and the focal length \(f\) of the lens can be found considering the total phase delay accumulated by a plane wave of wavelength \(\lambda\) in the propagation through the surface relief. Each point of the surface introduces a space-dependent phase delay on
a portion of the incident plane wavefront determined by the propagation distance $2\pi f/(\text{field})$ (according to Equation (1))) in the material and the optical path accumulated due to propagation in air from the plane $z = h$ to the target axial focal point at $z = h + f$. Imposing that the diffracted light from each of the topographic maxima (located at $r = \sqrt{(2\pi M)/\alpha}$, M integer number) constructively interferes at the focal plane (having a phase delay of $\pm 2\pi M$), the following relation is obtained

$$\alpha = \frac{2\pi}{\lambda^2 - 2zf}$$ (7)

In addition to the focus at the distance $f$, the phase mask (5) produces also secondary foci of decreasing intensity at the distances $f_m = f/m$ (with $m = \pm 1, \pm 2,...$). (53)

The smallest focal distance $f$ realizable with the system is determined by the discrete nature of the SLM pixels, which limits the spatial resolution in the holographic light distribution and induces large speckle noise affecting the contrast in the holographic intensity patterns. A lower limit of $f = 300 \mu m$ for lenses maintaining good structural quality was found. This parameter, together to the lens diameter of $\approx 2\mu m$ defined by the field of view of the holographic system, set also the maximum numerical aperture NA $\approx 0.32$ achievable with the specific experimental configuration. Using an improved holographic system, involving a higher resolution SLM and a larger NA microscope objective, the maximum achievable NA for the diffractive lenses can be enhanced even further. The lower NA limit, for the same lens diameter, is instead mainly determined by the photostructuring properties of the azopolymer, which limit the focal length at approximately the value $f = 1200 \mu m$ and minimum NA $\approx 0.08$.

It should also be noted that, from Equation (6), the focal distance of a Gabor phase zone plate does not depend on the total relief amplitude $h$. This degree of freedom can be used in the design to impose constructive interference for other points of the topography, allowing the improvement of the total diffraction efficiency of the device. The optimum value for $h$ can be obtained by simulating the diffracted field in the lens focal region.

Fourier Integral for Focusing Efficiency Simulations: For the simulations at $\lambda = 633$ a refractive index $n = 1.70$ for the azopolymer as provided by ellipsometry measurements was used.

A unitary amplitude for the incident plane wave in the structured surface relief area was assumed, so the input field can be written as $U_{in}(x,y) = A_o(x,y)e^{ikz}$. Here, $A_o(x,y) = \text{circ}(R)$, is a mask circular function, whose value is 1 within the circle of radius $R$, where the DOE is present, and 0 outside. The minima of the surface are located at $z = 0$ plane and the phase modulation provided by the relief gives the output modulated field in the plane $z = h$ as: $U_{out}(x,y) = e^{i\psi(x,y)}$. The field $U(x,y,z)$, propagated in any plane at the axial position $z$ is obtained solving Helmholtz equation, considering the Rayleigh–Sommerfeld diffraction integral in Fresnel approximation (61,11)

$$U(x,y,z) = \frac{e^{i\psi(x,y)}}{4\pi} \int \int U_{out}(x',y') \exp \left\{ \frac{j}{\lambda z} \left( (x-x')^2 + (y-y')^2 \right) \right\} dx'dy'$$ (8)

For the simulation, Equation (2) is numerically implemented through a MATLAB script using the convolution theorem with the transfer function of the Fresnel propagator

$$U(x,y,z) = FT^{-1} (FT (U_{out}(x',y')) \times H(f_x,f_y))$$ (9)

Equation (3) is also used to simulate the focusing diffraction efficiency for the diffractive lenses as function of relief amplitude $h$, reported in Figure 3J. Efficiency in the simulation is defined as the ratio of the light power integrated over an area of size $(3 \times \text{FWHM})^2$ in the focal plane (where $\text{FWHM}$ is the Full Width at Half Maximum of the simulated focus) and the power of the unitary incident plane wave in the circular area of the structured surface (of radius $R = 100 \mu m$). Maximum efficiency ($\approx 34\%$) in the focal plane ($af = 500 \mu m$) is obtained for a value $h \approx 330 \mu m$. For this relief amplitude, the lenses produces secondary foci at $f/2$ with efficiency $\approx 10.6\%$) and at $f/3 (\approx 1.8\%)$ and virtual foci at distances $z = \pm f/m$ (with $m = 1,2,3...$). Experimentally, the maximum diffraction efficiency from the lens can be achieved by tuning the exposure time of the polymer film surface to the relative holographic writing pattern, as demonstrated in Figure 3J; and Figure S10 (Supporting Information).

Focusing Efficiency and Axial Point Spread Function Measurements: The measurement of focusing efficiency reported in Figure 3J of the main text was performed using a He–Ne laser beam, incident on the diffractive lens from the substrate side. Light focused by the lens is collected through the same microscope imaging system used for brightfield surface observation. An iris and a power meter are used to intercept and measure only the light power transmitted through an aperture of the same diameter as the image of the structured area. Additional details about the experimental configuration can be found in Figure S11 (Supporting Information). Focusing efficiency is calculated as the ratio of the light power measured when the lens is written on the surface with respect to the same measurement with no structures in the photoresist area.

For the measurement of the axial point spread function of the reconfigured diffractive lenses, the iris is removed and the images of the transmitted He–Ne laser beam obtained axially translating the diffractive lens with 1 $\mu m$ (Figure 3J) or 3 $\mu m$ (Figure 4) steps are collected with the CCD in the focal plane of the tube lens (Figure S11, Supporting Information).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

This work has been financially supported by the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme “METAmorphoses”, Grant Agreement No. 817794. This work has been supported by Fondazione Cariplo, Grant No. 2019-3923, and by the project ACTRIS-Aerosol, Clouds and Trace Gases Research Infrastructure funded by Italian MIUR Programma Nazionale Infrastrutture di Ricerca (PNIR), Grant No. CIR01_00015.

Open Access Funding provided by Istituto Italiano di Tecnologia within the CRUI-CARE Agreement.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Received: September 10, 2021
Revised: December 6, 2021
Published online: January 24, 2022

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