Ultrafast Laser-Induced Metasurfaces for Geometric Phase Manipulation

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The recent advances in flat optics have challenged the limitations of conventional optics by implementing ultrathin planar elements that instead of relying on optical path differences manipulate light waves via optical resonators with spatially varying phase response.[1] In principle, the phase profiles of nearly any optical components including lenses, gratings, vortex phase plates, as well as elements capable of bending light in unusual ways could be designed on the basis of plasmonic metamaterials[2,3] or dielectric gradient metasurfaces,[4–7] referred to as geometric phase (Pancharatnam–Berry phase[8–10]) optical elements (GPOEs). The main advantage of such tailored surfaces is that the large phase shifts can be realized by nanostructured thin films within thicknesses smaller than the wavelength of light, and thus can be easily integrated into multifunctional optoelectronic systems.[11–13]

Designs of dielectric GPOEs have attracted great interest for being a promising alternative to control light waves. The most common approach for implementing such optical elements is to exploit nanostructured materials which exhibit form birefringence.[14] This means that the desired phase pattern of the wave interacting with an inhomogeneous birefringent element is directly encoded in the optical axis orientation and is equal to twice the rotation angle of the locally imprinted wave plate. Although numerous lithography techniques enabling the manufacturing of high efficiency and low scattering optical elements with complex phase gradients have been reported,[15–17] the technological flexibility remains a problem; it is still dictated by the material properties, and/or requires a long time to process. Here we propose a direct-write ultrafast laser nanostructuring as an alternative method capable of fabricating GPOEs in various media. One of the most fascinating aspects of this technique is the ability to induce tunable structures[16] with subwavelength periodicities that originate from birefringence.[18] The key advantage of using femtosecond pulses for direct laser writing, as opposed to longer pulses, is that they can rapidly deposit energy in solids with high precision. The light is absorbed and the optical excitation ends before the surrounding lattice is perturbed, which results in highly localized nanostructuring without collateral material damage.[19,20]

First observation of laser-induced periodic surface structures dates back to the 1960s, when Birnbaum reported ripple formation on the surface of semiconductors.[21] Since then, this phenomenon was observed on virtually any type of media including metal, semiconductor, dielectric solids, and thin films.[22–29] Processing conditions occurred to be broad with wavelengths ranging from the mid-infrared to visible spectrum and from continuous wave operation to femtosecond laser systems. A vast number of applications, including coloration,[30] control of surface chemical and mechanical properties,[31,32] and demonstration of various geometric phase designs including arrays of polarization microconverters and microlenses, polarization gratings (PGs) and computer-generated holograms with phase gradients reaching up to ≈1 rad μm⁻¹.

In order to identify the maximum birefringence of the laser-induced periodic thin-film structures, 80 μm long lines
Separation by an interline distance of 30 µm were imprinted under different writing conditions. First, the pulse density was varied from $1.25 \times 10^5$ to $5 \times 10^6$ pulses mm$^{-1}$ with various writing speeds to ensure the constant laser repetition rate of 100 kHz. Second, the writing speed was varied from 0.05 to 0.4 mm s$^{-1}$ with various laser repetition rates to ensure a constant pulse density of $5 \times 10^5$ pulses mm$^{-1}$. From here, the resulting retardance as a function of pulse energy varying from 0.005 to 0.2 µJ was obtained (Figure 1b).

At the pulse energies slightly above the ablation threshold (>0.05 µJ), the film is modified along its depth organizing into periodic structure that is qualitatively dependent on the pulse density (Figure 1b, top graph), while the significant dependence on the laser repetition rate or writing speed is not observed (Figure 1b, bottom graph). At pulse densities higher than $10^5$ pulses mm$^{-1}$, we detect the formation of birefringent subwavelength periodicity ($\approx \lambda_{\text{laser}}/3$) nanogratings oriented perpendicular to the laser beam polarization. The maximum retardation of $R_{546 \text{ nm}} \approx 0.55\pi$ is achieved at $5 \times 10^5$ pulses mm$^{-1}$ with the related writing speed of 0.2 mm s$^{-1}$ and repetition rate of 100 kHz. The retardance drops drastically with the further density.

Figure 1. Femtosecond laser-induced periodic a-Si:H film structures. a) Retardation of nanogratings induced in the bulk of silica, on the surface of silica, and in the thin-film silicon materials ($n = 3.5–4.5$), based on the experimental reports$^{[33,34]}$ and effective medium theory.$^{[18]}$ Both the thickness and the periodicity of structures were set to 300 nm with the duty cycle of 1/6. b) Separated lines with its retardance dependence on pulse density (top graphs) and writing speed (bottom graphs) as a function of pulse energy. On the right, optical and electron microscopy-generated images of periodic structures induced under (left) $5 \times 10^5$ pulses mm$^{-1}$ and (right) $5 \times 10^6$ pulses mm$^{-1}$ at pulse energy of 0.125 µJ: identification of subwavelength birefringent structure and wavelength-size nonbirefringent ripples formation. Scale bars are 4 and 1 µm, respectively. c) Retardance and transmission of imprinted wave plates as a function of pulse energy. The measurement system was operating at 546 nm wavelength. The dependence of retardance on the interline distance is shown in the inset. d) Spectra of phase retardation and transmittance for wave plate imprinted at 0.125 µJ. In (c) and (d) the interline distance (d) was kept at 3 µm. The system was set to the pulse density of $5 \times 10^5$ pulses mm$^{-1}$ with corresponding 0.2 mm s$^{-1}$ writing speed and 100 kHz repetition rate. Black arrows indicate the laser writing direction and blue arrows indicate the polarization state.
increase (>10^6 pulses mm^{-1}), as the nanogratings are replaced by nonbirefringent wavelength-size (=\lambda_{min}) ripples oriented parallel to the laser beam polarization.

Contrary to the separated lines, the planar wave plate elements were fabricated by partially overlapping the written lines. The calibration of imprinted elements was performed under maximum retardation conditions by changing the pulse energy from 0.005 to 0.2 \mu J at a fixed interline distance of 3 \mu m (Figure 1c), and varying the interline distance from 1 to 7 \mu m at a fixed pulse energy of 0.125 \mu J (Figure 1c, inset). The observed retardance linearly grows with the increase of the interline distance reaching the peak value of \approx 0.5\pi at 4 \mu m, which correlates well with the spot diameter of the focused beam. However, despite the lower retardance value, the most uniform wave plates are fabricated using the interline distance of 3 \mu m. When the retardance is defined as

\[ \varphi = h(n_e - n_o)2\pi/\lambda \]  

where \( h \) is the thickness of the film, and \( n_e, n_o \) are extraordinary and ordinary effective refractive indices, the birefringence of the imprinted optical elements with \( h = 300 \) nm and 0.5\pi retardation at 546 nm wavelength corresponds to the form birefringence of \( \Delta n = n_e - n_o \approx 0.5 \). This is two orders of magnitude higher than commonly observed in uniaxial crystals such as quartz, ruby, sapphire, or femtosecond laser nanostructured silica glass and is comparable to the birefringence achieved in amorphous silicon by electron-beam nanolithography.[36]

Optical performance of the fabricated wave plates is strongly dependent on the linear absorption and scattering of the nanostructured material, which can be significantly improved by reducing the intrinsic losses. This can be achieved by increasing the laser pulse energy as it triggers the oxidation processes and ultimately enhances the transmission reaching a threefold value at the pulse energy of 0.2 \mu J, while the birefringence remains of the same order (Figure 1c). Alternatively, this problem could be partially overcome by achieving highly regular nanostructures with a reduced scattering, and/or by implementing other high-index materials with a transparency window in the visible spectrum. Thus, the realization of nanostructuring in lossless materials could challenge not only the nanolithography techniques that are strictly limited to the fabrication of silicon-based or plasmonic metasurfaces but also the conventional optics.

The dispersion analysis performed in the spectral region from 450 to 680 nm revealed a chromatic behavior of the wave plates. In the case of pulse energy of 0.125 \mu J and interline distance of 3 \mu m the retardation value varies from 0.29\pi to 0.47\pi (Figure 1d). Less than 5% variation in retardance is observed in the spectral region from 532 to 546 nm wavelength. The transmission coefficient at 532 nm, where the phase retardation is about 0.45\pi, is roughly of \approx 0.1. The transmission as high as 50% can be achieved at longer wavelengths (>700 nm) where the lower scattering is ensured, as well as the amorphous silicon band gap is approached (=1.7 eV[35]). Although the losses are high compared to other nanolithography techniques,[15,16] the performance is sufficient for the majority of applications such as polarization sensitive beam shaping, information and imaging technologies, quantum optics.

The Pancharatnam–Berry phase is a geometric phase achieved by space-variant polarization manipulations.[14] In the next step, we designed GPOEs by controlling the azimuth of the slow axis, \( A(x, y) \), of locally imprinted nanogratings. As a result, the circularly polarized light transmitted through the GPOEs experiences the relative phase change equal to

\[ \phi(x, y) = \pm 2A(x, y) \]  

where the sign is defined by the handedness of the input polarization.

All GPOEs demonstrated in this work were imprinted using the pulse energy of 0.125 \mu J and pulse density of 5 \times 10^5 pulses mm^{-1} with corresponding 0.2 mm s^{-1} writing speed and 100 kHz repetition rate. The interline distance was ranged from 3 to 4 \mu m to balance the homogeneity and magnitude of the laser-induced retardation.

Local orientation of the laser-induced nanogratings was continuously controlled by rotating the half-wave plate mounted before the objective lens. The accuracy of \approx 0.002 rad is ensured. The resulting geometric phase as a function of rotation of the half-wave plate, \( \theta(x, y) \), is expressed as

\[ \phi(x, y) = \pm (4\theta(x, y) - \pi) \]  

where the orientation of the half-wave plate corresponds to the orientation of the polarization of the linearly polarized laser beam, and the phase offset of \( \pi \) occurs due to the nanogratings orientation, which is always perpendicular to the incident polarization.

In particular, when designing GPOEs by laser direct writing, it is important to ensure the continuity of the generated nanogratings. If the phase gradient is introduced, the local field as well as the induced structures is perturbed by the previously imprinted structure. Thus, the induced retardance value drops with the decrease of the grating period (a) (Figure 2a).

The efficiency of the GPOEs imprinted in a-Si:H thin films was specified by fabricating the PG with the geometric phase varying in the x-direction as \( \phi(x) = (2\pi a)x \mod 2\pi \) (Figure 2b). The periodicity of the PG was set to 30 \mu m as the achieved phase retardation shows the value of \approx 90% of the possible maximum value. For an incident plane wave with the polarization state \( |E_x\rangle \) the resulting field generated by the PG is\[38]

\[ |E_{xx} = \eta_L|E_x\rangle + \eta_R e^{2iA(x)}|R\rangle + \eta_A e^{-2iA(x)}|L\rangle \]  

where the \( \eta_L = 1/2(t_{ex} + t_{ex} e^{i\phi}) \), \( \eta_R = 1/2(t_{ex} - t_{ex} e^{i\phi})E_{xx}|L\rangle \) and \( \eta_A = 1/2(t_{ex} - t_{ex} e^{i\phi}) E_{xx}|R\rangle \) are the complex field efficiencies with \( E_{xx}|R\rangle \) as an inner product of the left-handed \( |L\rangle \) and right-handed \( |R\rangle \) circular polarizations. The retardation of the imprinted element, and \( t_{ex, y} \) is the real-amplitude transmission coefficients for light polarized perpendicular and parallel to the optical axis. From Equation (3), the imprinted PG with \( t_{ex, y} = 0.14 \) (Figure 1d) and \( \phi = 0.39\pi \) (Figure 2a) at a wavelength of 532 nm is expected to diffract around 5% of the light intensity, while 9% would propagate as a nondiffracting beam. Figure 2c shows good agreement as roughly 64% of the transmitted light is projected to the 0th order and 36% to the \pm 1st order. Also, the handedness of the diffracted circularly polarized beam is flipped. As the polarization of the generated beams does not vary spatially, polarization filtering could be applied in order to completely eliminate the nondiffracted light. In addition, if the
half-wave retarder is fabricated, the ≈100% efficiency could be achieved. Potentially, the retardation value could be enhanced by introducing a thicker layer of a-Si:H films.

The flexibility of the direct-write femtosecond laser nanostructuring technique allows recording of nearly any wavefront as a GPOE. The phase profile of an incident plane wave can be manipulated radially, azimuthally or both simultaneously resulting in complex designs.

Here, we demonstrate GPOEs with azimuthal phase variation serving as polarization and phase converters that generate optical vortices with radial and azimuthal polarizations. Initial experiments were carried out with a radially symmetric 1 mm diameter GPOE (Figure 3a–d). The topological charge of the element was set to \( c = -1 \), indicating that the resulting orbital angular momentum has opposite sign compared to the input spin momentum.\(^{[39]}\) The average value of the induced retardance was ≈0.5\( \pi \) at 532 nm wavelength, which corresponds to the quarter-wave plate value.

The generated radially (azimuthally) polarized vortex with the orbital angular momentum \( l = -1 \) (\( l = 1 \)) is considered as a superposition of two circularly polarized beams, one possessing the orbital angular momentum \( l = -2 \) (\( l = 2 \)) and the other having a plane front.\(^{[39]}\) The circularly polarized vortex with topological charge 2 can be separated by filtering with a quarter-wave plate and linear polarizer.

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**Figure 2.** Femtosecond laser direct writing of GPOE designed as a polarization grating (PG). a) Retardance dependence on the PG periodicity; b) Imprinted PG with period of 30 \( \mu \)m: top image—the azimuth of the slow axis of the imprinted nanogratings; bottom graphs—the profiles (white dashed line in top image) of retardance and slow axis extracted from the birefringence image. Pseudo colors (inset) indicate the local orientation of slow axis. c) Frequency-doubled Nd:YAG cw laser beam profiles of the transmitted right-handed circular (top) and left-handed circular (bottom) polarizations, and its corresponding diffraction efficiencies and polarizations states (white and red circular arrows) produced by PG.

**Figure 3.** Radial and azimuthal polarization optical vortex converter for circular incident polarization. Imaged a) azimuth of slow axis orientation and b) averaged retardance value of roughly 0.5\( \pi \) specifies GPOE as a quarter-wave plate working at wavelength of 532 nm. Pseudo colors indicate the direction of slow axis. Color bar: 0–0.8\( \pi \) rad. c,d) Linearly polarized optical transmission images of polarization sensitive element illuminated by circularly polarized light. Scale bar is 300 \( \mu \)m. e–g) Simulated and h–j) measured far-field intensity profiles of frequency-doubled Nd:YAG cw laser beam generated by the radial (azimuthal) polarization vortex converter. Blue arrows indicate the polarization state of the incident beam and red arrows indicate the polarization state of the light imaged on the camera detector.
To verify the radial/azimuthal polarization optical vortex, the converter was inserted into the path of a left/right-handed circularly polarized frequency-doubled neodymium-doped yttrium aluminium garnet (Nd:YAG) cw laser beam. For comparison the optical test system was modeled using the algorithm based on Jones matrix formalism and Fourier propagation.[40] The propeller-shaped interference pattern, which is typical for double charged vortex beams, obtained after filtering with a linear polarizer confirms successful realization of the imprinted radially symmetric GPOE (Figure 3e–j).

In the next step, we imprinted a 2 mm × 2 mm array of 100 micrometer diameter converters (Figure 4). Same as in Figure 3, an intricate swirling interference pattern was observed in the far-field indicating the presence of the orbital angular momentum with \( l = \pm 2 \), where the handedness of the input circular polarization defines both the orientation of the generated whirlpool beams and the resulting polarization state (Figure 4b,c). This shows that the technique enables imprinting of elements with high resolution and can be exploited to develop miniaturized optics for optical manipulation of micro-objects on lab-on-a-chip platform,[41] or directly imprint waveplate arrays for polarization sensitive detectors.[42]

To realize the most basic GPOEs with radially varying phase profiles, we fabricated a 9 × 9 array of 100 μm diameter geometric phase lenses with focal lengths of \( f_1 = \pm 0.4 \text{ to } 1.2 \text{ mm} \) (Figure 5). Each lens was repeated nine times to form a 9 × 9 subarray (Figure 5a,b). The orientation of the half-wave plate...
used to control the resulting geometric phase is expressed through the phase function of a conventional aspheric lens

\[ \theta(r) = \pm \frac{1}{4} \left( \pi + \frac{2\pi}{\lambda} R \left( 1 - \sqrt{1 - \frac{r^2}{2R^2}} \right) \right) \]  

with \( R \) as the radius of curvature (\( f = 2R \)), and \( r \) as the radial position, which corresponds to \( \sqrt{x^2 + y^2} \). Sign of the phase determines the geometry of the imprinted element, which must be chosen according to the handedness of the input circular polarization.\(^{[43]}\) Therefore the same geometry works as a converging or diverging lens for left-handed and right-handed circular polarizations, respectively (Figure 5c). These GPOEs were incorporated into the polarization sensitive optical imaging system operating at 546 nm wavelength. By changing the position \( z_i \) of the microscope objective, the imaged object was projected at different locations on the detector with the corresponding relative magnification of \( M_z = \frac{f_z}{f_z - d_0} \), where \( f_z = 10 \text{ cm} \) (Figure 5a). It should be noted that the absolute value of position \( z_i \) is defined by the overall measurement system, and its relative value is proportional to the focal length of the imaging microlens.

The final example of GPOE showing the simultaneous radial and azimuthal phase manipulation is a computer-generated geometric phase Fourier hologram (CGH) that converts the initial Gaussian beam into the target intensity distribution. Using the adapted weighted Gerchberg–Saxton algorithm,\(^{[44]}\) the eight-bit grayscale CGH element with 0.1 megapixel and pixel spacing of 3 \( \mu \text{m} \) was designed to encode the “Light” logo (Figure 6). During the continuous writing process, the maximum relative continuous phase change of \( \pi \) between the two adjacent pixels was achieved.

By using the Fourier transforming properties of a positive lens, the target image was reconstructed within the spectral range of 450–950 nm (Figure 6c–h). As it was mentioned before (Equation (3)), to attain the high efficiency of an imprinted GPOE, the half-wave retardation must be ensured. However, even if the retardance is below this value, the nondiffracted beam can be completely removed by the means of polarization filtering (Figure 6a). In addition, the geometric phase is independent of wavelength. Therefore, the phase profile for different wavelengths transmitted through the same GPOE will be the same. In this case, the broadband sources can be implemented with a filtering efficiency, \( \eta = \frac{I_{1st}}{I_{1st} + I_{0th}} \), as high as \( \approx 100\% \) (Figure 6d–h). It also should be mentioned that the same accumulated phase due to the different wave vectors causes each wavelength to diffract at different angles.

In summary, we have demonstrated potential implementations of laser-induced periodic thin-film structures as the geometric phase manipulating elements including polarization gratings, Fourier holograms, microlenses, and optical vortex microconverters. The direct-write ultrafast laser nanostructuring is a high precision, flexible, and time efficient technique. Through nonlinear light–matter interaction, the subwavelength resolution is ensured providing the possibility of reaching phase gradients higher than 1 rad \( \mu \text{m}^{-1} \). As a result, the applicability to any material that supports laser-induced periodic structures including high-index dielectrics and lossless dielectrics deposited on any substrate with different textures could revolutionize the fields of integrated flat optics and provide new methods of manufacturing. The overall fabrication process of the millimeter-sized elements being on the time scale of minutes could facilitate innovative solutions in fields such as security marking, data storage, solar cells, sensors, and detectors.

**Experimental Section**

The experiments were carried out with the 300 nm thick a-Si:H films deposited on a silica glass substrate by the plasma-enhanced chemical vapor deposition upon the decomposition of the mixture of silane (SiH\(_4\)) and argon (Ar) at substrate temperature of 250 °C. The ratio between the gases in the reaction chamber was 25% SiH\(_4\) and 75% Ar.

The film was processed with the mode-locked ytterbium-doped potassium gadolinium tungstate (Yb:KGW) ultrafast laser system (Pharos, Light Conversion Ltd.) operating at a wavelength of 1030 nm with the pulse duration fixed at 360 fs. The laser beam was focused to a 4 \( \mu \text{m} \) spot on the film via a 0.13 numerical aperture (NA) objective lens.

Surface imaging of laser-induced periodic thin-film structures was performed with a scanning electron microscope Zeiss Evo50 and optical transmission microscope Olympus BX51. The imprinted elements were optically characterized and/or visualized with the VIS/NIR microspectrometer (CRAIC Technologies, Olympus BX51) and the quantitative birefringence measurement system (CRI Abrio, Olympus BX51) operating at 546 nm wavelength. Nd:YAG cw laser (Spectra-Physics) frequency-doubled to 532 nm and supercontinuum fiber laser
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(Fiamium) emitting a broad optical spectrum in the range of 450–950 nm were used to characterize the Gaussian beam propagation through the imprinted CPOEs.

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