Article
Laser-inscribed stress-induced birefringence of sapphire

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Abstract: Birefringence of $3 \times 10^{-3}$ is demonstrated inside cross sectional regions of 100 µm, inscribed by axially stretched Bessel-beam-like fs-laser pulses along the c-axis inside sapphire. A high birefringence and retardance of $\lambda/4$ at mid-visible spectral range (green) can be achieved utilising stretched beams with axial extension of 30-40 µm. Conditions of laser writing chosen ensure that there are no formations of self-organised nano-gratings. This method can be adopted for creation of polarisation optical elements and fabrication of spatially varying birefringent patterns for optical vortex generation.

Keywords: femtosecond laser; birefringence; stress; sapphire

1. Introduction

Three dimensional (3D) structuring of materials with a high refractive index at sub-wavelength resolution has promise to advance the field of photonic crystals (PhC) and the integration of PhC’s into photonic chips [1–11]. Femtosecond laser micro/nano-fabrication as the no contact method can directly pattern subwavelength ripples[12,13] on the surface and 3D microstructures in the transparent materials(polymer[14,15] and glass[16,17]). a 3D nonlinear PhC has been successfully fabricated inside lithium niobate using femtosecond laser[18]. However, this is still a challenging task [19] to deliver a close to diffraction-limited focusing at arbitrary depths required for the 3D patterning when using Gaussian-like laser pulses [20]. Compensation of spherical aberrations can be successfully achieved for laser writing in high refractive index materials and large depths [21]. In this study we enhance (instead of compensating [21]) the spherical aberration by tailoring axial light intensity to be stretched along the propagation direction and to form a Bessel-beam-like axial intensity profile. Similar techniques are utilised for 3D patterning inside high refractive index materials [5,6,8,22]. We use stretched pulses to control 3D structuring and dielectric permittivity change over tens-of-micrometers along the entire typical length of fs-laser pulses of 100-300 fs.

Permittivity $\varepsilon = n^2$ changes between the laser inscribed region $\varepsilon_1$ (refractive index $n_1$) with width $t_1$ separated with a host material of permittivity $\varepsilon_2$ and width $t_2$, creates an artificial uniaxial form-birefringent structure with $\varepsilon_e - \varepsilon_o > 0$, where $\varepsilon_e$ denotes extraordinary and ordinary beams polarised $\parallel$ and $\perp$ to the optical axis OA, respectively. Controlling the width of $t_1$, its period $t_1 + t_2$ and depth $d$ (along the light propagation) are essential parameters to an engineers optical elements.
for polarisation control. Stress-induced birefringence is a well utilised phenomenon to create artificial birefringent materials and also used as a method for material inspection and characterisation [23, 24]. Generally, the inverse permittivity tensor is $\Delta(1/e_{ij}) = P_{ijkl}\partial_k u_l$, where $P_{ijkl}$ is the fourth-rank photoelasticity tensor, $\partial_k u_l$ is the gradient of the displacement from equilibrium with $u_l$ being the linear displacement from equilibrium and $\partial_k$ denotes differentiation with respect to the Cartesian coordinates. Birefringence induced by stress and expanding beyond laser structured regions is promising for polarisation micro-optics, especially when the laser modified regions are not used for optical functions due to considerable light scattering losses.

In any type of material, the modification of adjacent regions with resolution smaller than the wavelength can be used to change the effective refractive index which for extraordinary $n_e$ (along the optical axis) and ordinary $n_o$ indices depend on the volume fraction $f = t/\Lambda$ [25]:

$$n_e = \sqrt{f n_2^2 + (1-f)n_1^2}; \quad n_o = \sqrt{f n_2^2 + (1-f)n_1^2}$$

where $n_1$ and $n_2$ are the indices of the host and laser inscribed regions, respectively. If modification of a silica host $n_1 = 1.40$ becomes $n_1 = 1.45$, the form birefringence would only reach maximum of $\Delta n = n_e - n_o = -8.8 \times 10^{-4}$ when $f = 0.5$, i.e., $t = \Lambda/2$. Hence, for a considerable phase delay required for $\lambda/4$ or $\lambda/2$ polarisation optics, a tens-of-$\mu$m long axial modification $d$ would be required for engineering the retardance $\Delta n \times d$. Moreover, a well controlled laser inscription method developed here is required to reach the optimal conditions of $f = 0.5$ for the shortest modification $d$ at the chosen wavelength of operation (the strongest birefringence $\Delta n$). Such flexibility is currently not available for fabrication of micro-optical elements.

Here, we show that form-birefringence patterns inscribed in crystalline sapphire (along c-axis) can reach retardance of $\lambda/4$ ($\pi/2$ in phase) for visible wavelengths using a simple approach to generate femtosecond pulses with an axially extended Bessel-like intensity distribution using a spatial light modulator (SLM).

2. Experimental

C-cut sapphire samples of 0.5 mm thickness were used for laser inscription. Sapphire has one of the highest Young modulus’ of $Y = 400$ GPa and can withstand high pressures strongly localised inside the crystal, as our earlier studies have shown [27]. The stress-induced birefringence inside the laser structured region patterned with single pulse irradiation reached $\Delta n \approx 1 \times 10^{-3}$ and the pressure was estimated to reach 1.3 GPa. At $\sim 2$ GPa, micro-cracks were developed when Gaussian fs-laser pulses were used in c-cut sapphire [27].

In this study, a fs-laser beam at second harmonic $\lambda = 515$ nm wavelength and pulse duration of $t_p = 280$ fs (Pharos, Light Conversion) was reflected from a phase mask designed on a spatial light modulator (SLM) and directed onto a tight focusing objective lens for laser structuring. Typical pulse energy used for stress-induced birefringent gratings was $E_p = 574$ nJ (at focus) to inscribe single modification lines inside sapphire at laser repetition rate of 10 kHz at a beam scanning speed of 0.5 mm/s (if not specified otherwise). The phase mask pattern on the SLM was selected to create close-to-linear intensity distribution along the propagation direction and was characterised by a stretch factor $f_{st}$. For the $f_{st} = 0$ the spherical aberration was compensated at the position of the Gaussian beam, while the largest value of $f_{st} = 12$ was at the maximum stretch to obtain a linear intensity distribution over the entire pulse length $ct_p/n$, here $t_p$ is the pulse duration. The inscribed structure was approximately 10-15 $\mu$m below the sample surface. A laser structured sapphire sample was cleaved to expose the structured regions and was etched in 20% HF for 60 min at 130°C degrees before SEM observation.
3. Results and discussion

By choice of repetition rate and scanning speed, it was possible to create a single line without movement of the sample (or beam). By scanning the beams linearly modified regions, lengths of tens-of-micrometers were recorded. For the most axially stretched intensity distribution, single lines were recorded without usual formation of nanogratings \[4,28\]. First, we present structural characterisation of inscribed modified lines at a wider range of parameters and subsequently present results of optical characterisation of patterns which can deliver a $\lambda/4$ waveplate performance.

3.1. Direct write of nanoplanes

Inscription of long axial modifications in a crystalline sapphire were made by focusing onto a c-plane sample. Typical results of laser inscription are summarised in Fig. 1. For smaller stretch factors $f_{st}$, formations of nanogratings with period $\Lambda = 0.3 \mu m$ are observed along the optical axis. The expected period is $\Lambda = 514 \, [\text{nm}]/(2 \times 1.7) = 151 \, \text{nm}$ for the normal incidence. At larger angle of incidence $\theta$, $\Lambda/(1 - \sin \theta)$ and for $\theta = 30^\circ$ a period twice as large is expected as $\sin(30^\circ) = 1/2$. In the Bessel-like beam, light was propagating onto optical axis at a given angle and the observation was consistent with expectations. For smaller pulse energies and/or larger $f_{st}$, the individual laser damaged nano-regions, which were not initially interconnected into a single line, were observed connected after wet etching. With an increasing number of pulses (larger repetition rate or slow scanning), those single damage regions formed a line, which was further revealed after etching. Wet
etching in a more concentrated 20%vol. HF solution and at higher 130°C temperature was employed due to the very high contrast of etching between crystalline sapphire and laser amorphised regions such as those seen previously when utilising silica and boro-silicate glasses [16,29–33]. Etching up to 0.8 mm into the depth of sapphire sample (along z-axis; inset in Fig. 1(a)) was observed when continuous inscriptions of strongly modified regions were formed at smaller stretch factors.

The longest inscription of modifications of up to 60 µm along the propagation direction (on y-axis) were inscribed with ~ 850 nJ/pulse energy at \( f_s = 12, f = 10 \) kHz repetition rate and scanning speed of \( v_s = 0.1 \) mm/s along z-axis (Fig. 1(a)). At these conditions approximately \( n = d/v_s/f \approx 46 \) pulses were accumulated over the diameter of the focal spot \( d = 1.22\lambda/\text{NA} \approx 465 \) nm.

The use of an even distribution of pulse energy along the propagation axis can be applied in generation of high pressure and temperature phases of materials due to better energy delivery via resonant absorption [34]. The proposed phase control using SLM can, in principle, be adopted for experiments exploring a temporal evolution of fs-laser pulse induced micro-explosions using femtosecond X-ray pulses of a free electron laser (FEL) for probing, whilst coaxially propagating Bessel-like beams [35] can be utilised for optically triggered micro-explosions. The current study confirms formation of an amorphous phase of sapphire which is typical in conditions of high pressure [36].

3.2. Engineering of birefringence

Optical characterisation of laser inscribed gratings are shown in Fig. 1. Gratings with \( \Lambda = 10 \) µm period were inscribed with duty cycle of 0.5, i.e., 10 µm were inscribed with a separation of \( \Delta x \) ranging from 200 nm to 500 nm between axially extended linear modifications. Between the laser inscribed regions, 10 µm separations remained. The footprint of the gratings were 100 × 100 µm², an acceptable size for many applications in the field of micro-optical elements.
Figure 3. Retardance $|\Delta n|d$ measured at several wavelengths from 475 nm to 650 nm with 10-nm-bandpass filters. Sample was fs-laser inscribed at pulse energy $E_p = 574$ nJ; sample is shown in Fig. 1. Rectangular regions of interest (ROIs) show locations from where an average retardance was measured. Two lines of gratings with different stretch factors of 11 (the length of inscribed line $d = 40$ µm) and 10 ($d = 30$ µm) were analysed using liquid crystal compensator [26]. Note logarithmic ordinate was used to reveal single exponential decay of retardance with $\Delta x$.

Single lines without the formation of self-organised nanogratings were inscribed by stretching the incoming fs-laser pulses. The stretch factors of 10 and 11 corresponded to a single line (a plane under scanning) inscription for 30 and 40 µm, respectively. Inspection of the laser inscribed regions with scanning electron microscopy (SEM) revealed the width of the structurally modified lines corresponding to ~100 nm. When those modifications were written with $\Delta x = 200$ nm separation, cracks formed during the laser writing (Fig. 1), however, for larger separations the gratings were stable. The writing depth was approximately 10 µm below the surface and also extending into the sample. Strong stress-induced birefringence was observed inside the gratings in the regions without laser damage as well as between the gratings as revealed by cross-polarised imaging.

To determine the sign of refractive index change $\Delta n = n_e - n_o$, a $\lambda$-waveplate at 530 nm was inserted at 45° in respect to the orientation of polariser and analyser. In this setting, Michael-Levy color charts can be used to determine color changes corresponding to $+|\Delta n|$ and $-|\Delta n|$ (Fig. 1(c)). For the $\lambda$-plate of 530 nm wavelength oriented vertically, the blue color indicates the stress-induced regions. Since the blue color on the Michael-Levy chart corresponds to the higher absolute birefringence and the orange to the lower, the change $\Delta n$ has to be negative $n_e > n_o$, where $n_o$ is refractive index of the ordinary beam (perpendicular to the optical axis OA). The slow axis of the form-birefringent pattern (grating) is along the vertical direction (Fig. 1(c)) and the refractive index is $\Delta n = n_{||} - n_{\perp} = n_e - n_o > 0$. Hence, the form birefringent structure acts as a negative uniaxial crystal.

To determine birefringence $\Delta n(\lambda)$ at several wavelengths, a recently developed method was utilised for birefringence imaging [26]. A single wavelength measurement of birefringence (as for example in the popular Abrio tool) leaves an ambiguity of the true $\Delta n$ value due to a possible $2\pi$ folding of the phase and is avoided by carrying out measurements at several wavelengths in our method [26]. The measurement of $\Delta n$ was made at five different wavelengths whilst using narrow 10-nm-bandpass filters spanning the visible spectral range of 475 - 650 nm. A good linear fit through the origin point in the retardance ($\Delta n \times d$ [nm]) vs. wavenumber ($1/\lambda$) plot was obtained with a single slope which defines the $\Delta n$ averaged over the tested spectral range [26]. Figure 1 shows the experimentally determined retardance for the grating patterns. Regions of interest (ROIs) were set to average retardance on the grating and in its vicinity. The highest $\Delta n \times d$ was observed in between gratings and was scaling with separation $\Delta x$ between nano-planes. For the largest retardance of 112 nm...
Figure 4. Retardance $|\Delta n d/\lambda|$ [waves] measured at 650 nm; sample are shown in Fig. 1. (a) Retardance map calculated at a single pixel level for VGA 640×480 pixel area. The $\delta n = 0$ contour lines are shown to distinguish regions affected by stress-induced birefringence; the maximum was 0.22. Horizontal single pixel cross sections are plotted in (b). The slope of retardance $\gamma = 4.8 \times 10^{-4}$/pixel or $(3.27 \times 10^{-4})/\mu m$ at the used magnification was achieved. One pixel corresponds to $1.4 \mu m$ in the image while the optical resolution for the $NA = 0.2$ lens was $0.61 \lambda/NA = 2 \mu m$.

Figure 5. Retardance $|\Delta n d/\lambda|$ [waves] measured at 625 nm with higher resolution $NA = 0.4$; sample are shown in Figs. 1 and 1. Retardance map calculated at a single pixel level. Cross sections for two regions inscribed with stretch factors $f_{st} = 11$ and 10 are shown as one-pixel line for $\Delta x = 200 \mu m$; for $f_{st} = 10$ five separate lines and their average (+-marker) are plotted. The optical resolution for the $NA = 0.4$ lens was $0.61 \lambda/NA = 0.95 \mu m$. Rectangular markers show positions of the inscribed regions for the $f_{st} = 11$ grating.

and stretch parameter of 11 ($d = 40 \mu m), \Delta n = 2.8 \times 10^{-3}$. Only slightly smaller birefringence was determined for the stretch factor of 10 and 30-µm-long inscribed gratings (Fig. 1).

Even the larger $\Delta n$ values were observed inside gratings in the 10-µm-wide openings reaching 22% retardance at the longest wavelength of 650 nm, selected for the measurement of birefringence (Fig. 1(a)). For shorter wavelengths in visible range, a $\lambda/4$ waveplate condition was achieved by direct write of nano-inscribed modifications without changing the axial position of the modified region during inscription. Single-pixel cross sections (Fig. 1(b)) of the retardance maps show difference in the gradient of $\Delta n$ between laser inscribed regions. By placing few regions of nano-planes at different depths from the surface, it should be possible to fabricate $\lambda/2$ waveplate retarders. Stress-induced
regions between larger extended laser-structured patterns should allow to reduce light scattering observed from laser inscribed areas which are utilised for fabrication of optical elements [4,37].

Optical resolution of large area birefringence mapping (Fig. 1) have provided only a couple of points measured per 10 µm regions inside the grating. It illustrates a 0.1λ modulation of retardance. Measurements at approximately twice a higher resolution (Fig. 1) confirmed the modulation amplitude of retardance shown in Fig. 1. Retardance at 625 nm wavelength (Fig. 1) is shown in relative units of waves since the reference retardance required to make calibration was not possible to measure from the stress-free region on the same image (as area “Ref.” in Fig. 1). The cross sections of the measured retardance map clearly shows that stress-induced phase delay between inscribed 20-µm period and 0.5 duty cycle grating were clearly resolved between the laser inscribed regions with axial extension tens-of-µm.

4. Conclusion and outlook

We demonstrate λ/4 phase retardance at visible wavelengths in sapphire recorded by direct write of nano-inscribed modifications tens-of-micrometers long. This modality of laser structuring opens a flexibility in stress-induced optical element fabrication and eliminates light scattering since the regions of tailored birefringence are outside of laser structured regions. Patterns of tailored birefringence can be produced at different depths along the light propagation direction or even in different micro-plates for the final optical element. One particular field of application can be spin-orbital couplers where spatially variant birefringence can be inscribed with complex 3D topology similar to the polymerised 3D couplers [38].

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