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Stress distribution around femtosecond laser affected zones: effect of nanogratings orientation

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Abstract: Under certain exposure conditions, femtosecond lasers create nanogratings in the bulk of fused silica for which the orientation is governed by the laser polarization. Such nanostructure induces stress that affects optical and chemical properties of the material. Here, we present a method based on optical retardance measurement to quantify the stress around laser affected zones. Further, we demonstrate stress dependence on the nanogratings orientation and we show that the stress within single nanogratings lamellae can locally be as high as several gigapascals.

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References and Links

1. Introduction

Femtosecond laser exposure of fused silica below the ablation threshold leads to various structural modifications. Depending on laser exposure parameters, two types of structural modifications are of particular interest. In a first exposure regime, typically observed for pulse duration below 200 fs and pulses energy below 400 nJ [1,2], homogeneous structural changes (sometimes referred as type I) occur, leading to an increase of both refractive index [3] and etching rate [4] within the focal area. In a second exposure regime, corresponding to pulse duration above 200 fs and/or pulse energies above 400 nJ self-organized nanostructures - referred as ‘nanogratings’ or type II modifications - are found [5]. When the laser is linearly polarized, these structures orient themselves perpendicular to the electrical field vector [5,6].

Poumellec et al. [7] have shown that the induced stress in the laser affected zone depends on laser parameters such as polarization and energy per pulse. Furthermore, the polarization dependence on the optical retardance observed inside laser tracks depends also on stress and pulse front tilt [8]. On the other hand, we recently demonstrated that nanogratings induce a localized volume expansion and consequently, the build-up of stress around laser affected zones [9]. In particular, we observed the existence of a maximum stress level at a given energy deposition level, suggesting possible stress relaxation mechanism consecutive to crack formations above a given energy deposition level [9].

Here, we investigate a new method for measuring the stress surrounding laser affected zones for various exposure conditions and nanogratings orientations. Using a dedicated experimental method to expand the effect of stress-induced birefringence surrounding laser-written lines, we demonstrate that the stress distribution in the vicinity of the laser exposed area also depends on the orientation of the nanogratings. Furthermore, we use this observation to estimate the amount of stress surrounding individual lamellas that form the nanogratings.

The main application field of this study is metrology: more specifically, we provide a simple tool for rapidly quantifying laser-induced stress in glass. Stress not only influences optical properties but also etching rate [10] and the occurrence of cracks during laser exposure. In that context, the results presented here are of particular significance for assessing the stress distribution induced by femtosecond laser processing in the non ablative regime.

2. Hypothesis

Nanogratings consist of self-organized planar structures or lamellas. Based on recent observations from Canning et al. [11], we assume that nanograting lamellas are made of porous material (see Fig. 1 left). This porous material could be created by oxygen decomposition during the laser interaction in the nanogratings [11]. This hypothesis is consistent with the observed volume expansion reported in [9]. From these experiments, a pressure ($\sigma_0$) (Fig. 1 right) has been estimated as a function of the energy deposited in the material. A direct consequence of the lamellae-expansion hypothesis is that the stress surrounding laser-written lines should depend on the nanogratings orientation (i.e. the lamellas orientations with respect to the writing direction). In particular, this stress-orientation dependence observed at macroscale could be used to indirectly determine the stress surrounding individual lamellae at the nanoscale level.
Fig. 1. Left: Transverse and top view of a single nanograting made of porous material [11] Right: Top view of three lines with different nanograting orientation according to the writing direction and different $\sigma_0$ ($\alpha$ is the angle between the writing direction and nanogratings orientation).

3. Stress measurement in direct-write patterns

3.1 Methodology

Stress in transparent material induces birefringence that in turn introduces retardance for the light passing through the specimen. This retardance can be measured and is directly related to the principal stress components according to Eq. (1):

$$\sigma_1 - \sigma_2 = \frac{R}{T(C_1 - C_2)}$$

(1)

where $C_1-C_2 = C$ is the stress optic coefficient and is equal to $3.55 \times 10^{-12}$ Pa$^{-1}$ for fused silica at 546 nm. This coefficient is expressed by $C = (n^2/2) (\pi_{11} - \pi_{12})$ where $\pi_{11}$ and $\pi_{12}$ are the piezooptic constants for fused silica [12] and n is the refractive index. Since fused silica is isotropic, the tensor principal directions are considered collinear to the cylindrical coordinates. T is the thickness of the sample and R is the measured retardance.

High resolution retardance measurements in the immediate vicinity of a direct-written line are difficult to achieve due to the sharp intensity decay occurring over just a few microns [13]. To overcome this limitation, we enlarge the span of the stressed region using a dedicated laser-written pattern (shown in Fig. 2) suggested in [14] that we use here for a different purpose than it was initially designed for. In this pattern, several lines, forming an edge, are written one on top of another along the z-axis, and through the entire thickness T of the specimen. Lines are spaced with a given z increment (here, ten microns to avoid overlapping). We consider the stress homogeneous in the z direction. These edges are radially and regularly distributed around a circle (Fig. 2 left). Later, we refer to this particular shape as the‘sun pattern’.

Fig. 2. Left: top view of the laser pattern to investigate the dependence of the stress on nanogratings orientations. The laser beam is linearly polarized and the polarization is kept the same for all writing directions. Right: close-up three-dimensional view of two segments of the laser-written pattern shown in the left figure. (LAZ) stands for laser affected zones.
Nanogratings formed under linear polarization consist of a set of parallel planes with their normal vector collinear to the electrical field and perpendicular to the light propagation axis (k-vector). Consequently, in the case of the pattern above, if the polarization direction is set constant during the writing process (Fig. 2), each line of the radiating pattern will have a different nanogratings orientation with respect to the writing direction.

### 3.2 Stress profile between lines: analytical model

Using a method based on micro-cantilever deflection [9], we previously demonstrated that an expansion occurs in the laser exposed areas causing stress around laser affected zones. As a first order approximation in the case of the sun pattern, this expansion can be described by a homogeneous load distribution applied on both sides of a wedge (illustrated in Fig. 3). The stress distribution in such case was derived analytically by C. J. Tranter [15] who considered a plane-strain problem. Here, we just recall the main features of his model that we adapt to our particular case. In Tranter’s model, the shear stress is assumed to be equal to zero.

![Wedge model](image)

**Fig. 3.** Wedge model used for finding analytically the stress distribution in an angular section of the sun-pattern. To capture the stress resulting from the expansion of the laser-written lines, we consider homogeneous force distribution $(F_1, F_2)$ distributed along edges from a to b.

In Eqs. (2), the stresses, $\sigma_0$ and $\sigma_r$, are found using Mellin’s inversion formula [15]:

$$
\sigma_0 = \frac{1}{2\pi i} \mathcal{C} \left[ \int_{c-i\infty}^{c+i\infty} p(p+1)\phi r^{-p-2} dp \right],
$$

$$
\sigma_r = \frac{1}{2\pi i} \mathcal{C} \left[ \int_{c-i\infty}^{c+i\infty} \frac{d^2 \phi}{d\theta^2} - p\phi \right] r^{-p-2} dp,
$$

where $\phi$ is the stress function, $p$ a complex number, $r, \theta$ the cylindrical geometrical coordinates in Fig. 3. The edges are each subjected to an initial uniform pressure $(F)$ from the section a-b of the profile. The principal stress difference in the cylindrical coordinate system is given by Eq. (3):

$$
\sigma_\theta - \sigma_r = F(r) \left\{ \frac{2h}{r} \left[ \sin(\alpha) \cos(\theta) \cos(\alpha + \theta) \sin(\alpha - \theta) \xi^2 + \sin(\alpha + \theta) \cos(\alpha - \theta) \xi \right] \xi \log \left( \frac{b}{r} \right) \right\} d\xi
$$

$$
- \frac{2a}{r} \left[ \sin(\alpha) \cos(\theta) \cos(\alpha + \theta) \sin(\alpha - \theta) \xi^2 + \sin(\alpha + \theta) \cos(\alpha - \theta) \xi \right] \xi \log \left( \frac{a}{r} \right) \right\} d\xi,
$$

with:

$$
G(\xi) = \frac{\sin(\alpha - \theta) \cos h(\alpha + \theta) \xi^2 + \sin(\alpha + \theta) \cos h(\alpha - \theta) \xi}{\xi \sin(2\alpha) + \sin h(2\alpha \xi)}
$$

In our case, the angle between lines is set to $\theta = 2\alpha = 15^\circ$. From [15] and integrating $F$ over $r \in [a, b]$, the stress function $\phi$ is found. The integrals are solved between $\theta \in [0, \pi/2]$ with the residue method for the real part of $p$, which is equal to $-1$. 
The integral in Eq. (3) cannot be calculated analytically for $\theta \in [0,\pi/2]$. These two integrals, given below, are solved numerically for a particular angle:

$$f_a(r,\theta,\alpha, a) = \frac{1}{\pi} \int_0^\infty G(\xi) \sin \left(\frac{a \log\left(\frac{a}{r}\right)}{\xi}\right) d\xi$$

$$f_b(r,\theta,\alpha, b) = \frac{1}{\pi} \int_0^\infty G(\xi) \sin \left(\frac{b \log\left(\frac{b}{r}\right)}{\xi}\right) d\xi$$

Finally, from Eqs. (3)-(5) the difference of principal stress is expressed by Eq. (6):

$$F(r,\alpha) = \kappa \left[ \frac{2b}{r} \sin(\alpha) \cos(\theta) - f_a - \frac{2a}{r} \left[ \frac{\sin(\alpha) \cos(\theta)}{2a + \sin(2\alpha)} - f_b \right] \right]$$

Where, $f_a = f_b$ are numerical estimation of Eqs. (5) and are both equal to $0.78$ for $\alpha = 15^\circ$, $a = 100$ microns and $b = 600$ microns.

To take into account both the effect of the laser-written line width (which can be up to 2 microns depending on exposure condition and which is therefore not negligible) and the ‘bulk’ nature of our pattern as opposed to a slice as in Tranter’s model, we introduce a fitting factor, $\kappa$ (here 14), that we estimated using finite element simulations.

### 3.3 Stress profile between lines: finite element model (FEM)

To test and refine the analytical method, we use a finite element model of the stress induced within the sun pattern. Here, we consider a plane-strain problem (i.e. the sun pattern is considered infinitely thick). Since laser affected zones expand [9], we apply a uniform pressure $F$ (blue arrows in Fig. 4 left) around rectangles that are 2 µm-wide and 500 µm-long (i.e. the size of one laser-written line as measured with an optical microscope equipped with a 40x objective).

Fig. 4. Finite element simulation for the homogeneous case. (i.e without taking into account the nanogratings) Left: plane-strain problem definition and boundary conditions (forces are indicated with blue arrows) and right, simulated stress for identical forces applied on each line. (Here, for the sake of clarity, the polarization state is assumed to be the same for each line.)

The initial pressure ($F$) is found by calibrating the finite element simulation so that the maximum stress measured between lines (and therefore the retardance, in Eq. (1) matches the experimentally measured ones (in paragraph 4.1). The relation between the maximum stress in between the lines and the initial pressure applied on the edges is found to be approximately linear.

In the following section, we will use both models for finding the stress resulting from the expansion of the laser-written lines.
4. Experimental results

To write sun patterns, fused silica substrates (Corning 7980 of UV grade, 2 x 2 cm square and 250 +/- 25 µm thick) are exposed to a femtosecond laser (fiber-amplifier from Amplitude Systèmes, Femtoprint model) emitting 275 nJ pulses at 1030 nm and at a repetition rate of 800 kHz. The pulse duration is 270 fs. The numerical aperture of the objective (OFR/Thorlabs 20X-1064 nm) used for focusing the laser beam is 0.4. The branches of the sun-pattern are single lines stacked over the entire sample thickness (T = 250 µm). The energy deposition [16] is set by tuning the writing speed and the repetition rate. The retardance is measured with a commercial instrument (CRI-Polscope using a 10x objective).

4.1 Stress measured around laser affected zones

We consider sun patterns written with energy deposition level of 186 J/mm² (writing speed 1 mm/s). The measured stress (extracted from the retardance data) is fitted with the FEM simulations by simply changing one parameter, the initial pressure F (in this case the pressure is the same around every line forming the sun), which defines the boundary condition. As seen in Fig. 5, the best fit is found for F = 65 +/- 5 MPa which gives a corresponding stress measured in the middle of two consecutive radiating lines with an accuracy of +/- 1 MPa. This accuracy matches the stress measurement error. The error in measuring the position of the written pattern (located at r = 110 µm +/- 1 µm), gives a maximum error of 0.8 MPa for the initial pressure. Figure 5 right shows the measurement versus the analytical solution and the finite element simulation results. A reasonable match around the maximum is found for both the analytical and finite element models. While both models capture the stress decay with increasing r, the analytical solution is less accurate and overestimates the stress as r increases. The difference could be attributed to the geometry of the patterns. In the wedge case, the lines intersect, whereas for the measurement and simulation, the lines are distributed around a circle of 200 µm diameter. It nevertheless demonstrates that the profile birefringence effectively originates from the volume expansion of the laser-written lines and confirms our previous observations [9].

![Fig. 5. Left: Measured profile between two laser written lines versus simulations to define a range of pressure, in which the maximum principal stress fits best. The measurement error bars are not shown on this graph for clarity but are shown on the right graph. Right: Comparison of analytical model, finite element model and measurement.](image)

4.2 Polarization effects on accumulated stress

The sun patterns give an opportunity to test the influence of the nanogratings orientation on the stress surrounding the laser affected zones (Fig. 6). Nanogratings are oriented perpendicular to the electric field of the light [5,6]. Therefore, if the laser polarization is kept constant while writing the sun-pattern, the nanogratings will gradually turn with respect to the writing direction for consecutive lines.
Figure 6 clearly shows that the stress distribution amplitude depends on the nanograting orientation.

To model this orientation dependence, we use a multiscale approach (Fig. 7):

- In a first step, called ‘nanoscale model’, we simulate the influence of nanograting orientation along a single line, assuming that only the laser affected part of the nanogratings expands. The size and space are defined from SEM observation (from Fourier-transform of SEM images such as the one shown in Fig. 9, the nanogratings spatial period is estimated to be 275 nm \(\pm\) 5 nm, and the width is 100 nm \(\pm\) 5 nm). Following our initial hypothesis, we assume that each lamella (red segment in Fig. 7) of nanogratings expands. As boundary conditions, a homogeneous pressure, \(\sigma_{NG}\) is applied all over the lamellae (black arrows in Fig. 7 right). The shear, normal and transverse components of the stress tensor are simulated for every lamellae orientation. The resulting stress vector \(\sigma_R\) is used as input for the full-pattern simulation (step 2).

- In a second step, called ‘microscale model’, we simulate the stress distribution between lines, for the full-pattern. In this description, lines are represented by an homogeneous rectangle of finite thickness. By calibrating the finite element model with the experimental observations reported in paragraph 3.1 (for nanogratings oriented perpendicular to the writing direction and speed at 1 mm/s), we found that the stress around a single lamella is about 2 \(\pm\) 0.2 GPa. The complete simulation is performed between 0 deg and 90 deg with the angle increasing its increments of 15 deg. Note that according to our study on the effect of deposited energy [9, 16], the energy deposition considered here does not induce the maximum etching rate. It is therefore foreseeable that the etching rate could be even higher in the optimal cases of energy deposition. The dependence of stress-induced birefringence on energy deposited will be investigated in another study. Here, we focus on the role of nanogratings orientation on the stress distribution.

In Fig. 7, left, this methodology is described for three different line angles. The blue arrows represent \(\sigma_R\). Applying this method for the full patterns gives the results shown in Fig. 7 right.
Fig. 7. Multi-scale approach for investigating the effect of nanogratings orientation on stress distribution. Step 1 – ‘nanoscale description’, a uniform pressure $\sigma_{NG}$ (black arrows) is applied around a single lamellae (red segment) that constitutes the nanogratings for different lamellae orientation. Step 2 – ‘microscale description’, the resulting principal stress $\sigma_R$ (blue arrows) is applied on the equivalent rectangles, representing the nanogratings. This technique is then applied to the full patterns as can be seen on Fig. 7 right.

Figure 8 shows the measured maximum difference of principal stress ($\sigma_m$) depending on the line angles, for a particular set of exposure conditions (writing speed of 1 mm/s and energy deposited of 186 ± 2 J/mm²). A periodic distribution of stress is observed for both measurement and simulations. We simulated the stress for different spaces between lamellas (250, 300, 350 nm) because the width of a single lamella, in reality, is not constant in z direction (since it is elliptical). In this case the maximum width variation is ± 50 nm (One lamella is about 100 nm in width). The graph shows that the width of the lamellas does not dramatically change the trend and amplitude of the stress in between lines. This result confirms the validity of our model and that the nanogratings orientation influences the stress distribution. Since all other parameters were fixed, this variation is attributed to the nanograting orientation.

This result shows that the nanogratings orientation and therefore the laser polarization has a non-negligible influence in the stress induced in the material. The maximum simulated stress ($\sigma_m$), on the profile between lines can vary as much as 30 to 50%. The corresponding
refractive index change \((n_\perp - n_\parallel)\) resulting from stress induced birefringence around laser affected zones varies within the range of \(2.1 \times 10^{-5}\) to \(3.5 \times 10^{-5}\) for the experimental conditions of Fig. 7. This model further estimates that the stress around individual lamellae can be in the order of several GPa. Such high stress may locally create the conditions for crack nucleation and the collapse of the nanogratings structure resulting in stress relaxation phenomena as suggested in [14, 16]. Note that the occurrence of cracks inside nanogratings lamellae, were observed experimentally in [16]. It may also induce localized densification as well as the formation of high pressure phases, possibly surrounding the nanobubbles forming inside lamellas and causing the localized volume expansion.

4.3 Energy deposition effects on nanogratings

To further test this hypothesis of stress relaxation, we investigate the morphology of nanogratings for energy deposition levels ranging from 16 J/mm\(^2\) to 240 J/mm\(^2\) and for both polarizations (nanograting perpendicular to the trench and then parallel to the trench). The cantilever method described in [9], is used to identify the levels of stress and energy deposition at which relaxation occurs for both polarizations. Here, we examine the morphology of the lines for the chosen laser parameters (Fig. 9). In practice, several trenches at different energy depositions are written in a fused silica substrate. A solution of heated potassium hydroxide (KOH) (1M concentration at 80°C, 3h) is used to reveal the nanogratings [17].

![Fig. 9. Scanning Electron Microscope images of the nanogratings according to the energy deposition. The periodicity is about 275 ±5 nm and the size of the features is 100 ±5 nm.](image)

The nanograting periodicity is about 275 nm (± 5 nm). Their size and period do not seem to change according to the energy deposition level, although their morphology does. The nanograting regularity starts to gradually degrade (between 22 J/mm\(^2\) and 28 J/mm\(^2\) for transverse to the writing direction polarization, and between 28 J/mm\(^2\) and 34 J/mm\(^2\) for the longitudinal to the writing direction polarization) and finally becomes disorganized as the deposited energy increases further.

This result supports a model where the stress gradually increases inside nanograting lamellas, eventually causing their collapse and the degradation of the nanograting homogeneity.

5. Conclusion

Using a method based on retardance measurement, we have demonstrated that nanograting orientation influences the stress distribution induced in the material. To explain this phenomenon, we introduce a multiscale model that considers the local expansion of individual lamellas that constitutes the nanogratings. As the deposited energy increases, nano-cavities form (possibly as a consequence of the decomposition of SiO\(_2\) [11]) and cause the inflation of the lamellae volume which further results in localized stress generation and eventually the collapsing of the nanogratings structure when the stress becomes too high. In a separate experiment, we have shown that indeed, such a collapsing of the nanogratings is observed as the energy deposited is increased.
These results are particularly important for femtosecond laser processing in the non-ablative regime (the so-called ‘direct-write’ processes). Indeed, the stress not only influences optical properties (by introducing undesired birefringence effects) but also etching rates [10] as well as the formation of undesired cracks during the writing of arbitrary patterns. Therefore, an accurate knowledge of the stress-state induced in the material after laser exposure is essential for the reliability and repeatability of the direct-write process.

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