## 05

# Birefringent structures with high transmittance written in fused silica by ultrashort laser pulses

© N.I. Busleev<sup>1</sup>¶, A.E. Rupasov<sup>1,2</sup>, V.V. Kesaev<sup>1,2</sup>, N.A. Smirnov<sup>1,2</sup>, S.I. Kudryashov<sup>1</sup>, R.A. Zakoldaev<sup>2</sup>

 <sup>1</sup> Lebedev Physical Institute, Russian Academy of Sciences, 119991 Moscow, Russia
 <sup>2</sup> ITMO University, 197101 St. Petersburg, Russia
 <sup>¶</sup>e-mail: busleeni@lebedev.ru

Received November 08, 2022

Revised December 29, 2022 Accepted January 28, 2023

The writing process of birefringent structures in the volume of fused silica by focused ultrashort laser pulses with wavelength in visible range and various values of pulse energy, pulse duration, repetition rate, numerical aperture and moving substrate velocity has been studied. The retardance value of fabricated structures has been measured and influence of the subsequent annealing of these structures has been studied. It was shown, that combination of writing layered structures with subsequent annealing provides structures with high homogeneity, required retardance value and high transmittance.

Keywords: direct laser writing, femtosecond laser pulses, birefringence, annealing.

DOI: 10.61011/EOS.2023.02.55777.3-23

## Introduction

Currently, femtosecond lasers are a common tool for optical property modi?cation and transparent dielectric structuring, including glasses [1–3], sapphire [4–6], fluorite [7,8]. Depending on the parameters of the used laser emission, various types of modifications may occur in glasses: refraction index increase due to multiplexing [3], emergence of self-organizing nanoarrays and nanopores [1,2,9,10] and also void formation [3,11,12]. Nanoarrays and nanopores allow to form birefringence domains within fused silica. And the birefringence value may be controlled by recording created modifications, for example, by means of structure layer thickness variation. In fused silica, individual nanoarray elements constitute porous areas formed due to fast glass decomposition under the action of focused ultrashort laser pulses. Voids in these areas are caused by formation of oxygen molecules being the product of fused silica disintegration [13] and considerably reduce the refraction index of glass compared with the untreated volume.

Using orientation control of the recorded structures [14], whose orientation is perpendicular to polarization of the laser emission used for recording, various optical elements may be created [1,6,15,16]. Fused silica is a material which is interesting for various applications due to a wide transmittance range, high optical breakdown threshold and moderate refraction index dispersion. However, the created structures not always have high transmittance required for applications. This may be associated with periodicity fluctuation of the recorded structures and may be corrected by the increase in the number of pulses per point during structure recording at a lower scanning speed or specimen

movement that will result in formation of more uniform structures [16]. Also, transmittance loss may be reduced by reducing the size of recorded structure components.

The purpose of the study was to create uniform birefringent structures that would have a low retardance and high transmittance.

## **Experimental**

Satsuma (Amplitude Systémes) fiber laser with active Yb<sup>+3</sup> ion medium, 515 nm wavelength and linear polarization of exit emission. The pulse width controlled by the built-in compressor was equal to 300 fs, 1 and 2 ps, and the repetition rate is 250 kHz and 500 kHz. The fused silica specimen was attached to Prior H1P4A three-dimensional motor-operated platform designed to record structure layers at a velocity up to  $375 \,\mu$ m/s. microlens with numerical aperture NA = 0.1 and 0.25 were used for focusing.

To define dependence of structure characteristics on laser record parameters such as pulse energy and width, repetition rate and motor-operated platform velocity, singlelayer structure arrays were produced in a form of squares of side  $1000 \,\mu\text{m}$  and  $500 \,\mu\text{m}$ . Curves of retardance and transmittance of individual structures vs. record parameters are shown in Figures 1-3.

After analysis of the produced structures and selection of the best recording mode, multilayer structures were recorded. The first pane on the top of Figure 4 contains 12 layers of birefringent structures recorded with pulse energy  $0.5 \mu J$ , width 300 fs, repetition rate 500 kHz, NA = 0.25 and recording velocity  $375 \mu m/s$ . Distance between layers was equal to  $100 \mu m$ . The following square



**Figure 1.** Dependence of the retardance of individual squares on pulse energy for various recording parameters (pulse width, repetition rate, numerical aperture). Motor-operated platform velocity is  $375 \,\mu$ m/s. When retardance is lower than 100 nm, measurement error is  $\pm 1$  nm (not shown in the figure).



**Figure 2.** Dependence of the retardance of individual squares on motor-operated platform movement speed for various recording parameters (pulse energy, repetition rate). Pulse width is 300 fs, NA = 0.25. With retardance lower than 100 nm, measurement error is  $\pm 1$  nm (not shown in the figure).

also contains 12 layers, pulse energy is  $0.475 \,\mu$ J. Next squares were recorded with a lower number of layers, which did not allow to produce resulting uniform structures. Then the specimen was placed into a PID-control annealing furnace with the following conditions: initial temperature was equal to room temperature, heating was carried out at ~ 16.7°C/min up to 1000°C. After annealing at the

specified temperature during 2 h and 8 h, the specimen was gradually cooled down to room temperature during 12 h.

The laser emission energy was measured using Ophir 3A-P energy meter. retardance was measured by LCC7201 (Thorlabs) birefringence visualization system. transmittance of each individual structure was measured by MSFU-K (LOMO) microscope spectrophotometer.

## **Results and discussion**

Analysis of the resulting structures has shown that those structures that were recorded at high repetition rate would have been uniform. However, even in this mode, the degree of modification uniformity is reduced with decrease in pulse energy. The benefit of recording at low energy values is that the lowest retardance is obtained, which is required for production of optical elements containing multilayer structures with controlled resulting retardance. As a preferable recording mode, a mode with a pulse width of 300 fs, repetition rate of 500 kHz and NA = 0.25 (purple curve in Figure 1) was selected to record uniform structures with low retardance. A motor-operated platform velocity was set not higher than  $375 \,\mu$ m/s, since further velocity increase resulted in record degradation due to less accurate specimen positioning. To record structures at high velocities, a more accurate positioning system is required.

Curves of transmittance of individual recorded area vs. different pulse energies (Figure 3) demonstrate reduction of transmittance throughout the visible range when the pulse energy is increased, because this increases the imperfection of recorded structures and results in stronger light scattering on structures. When the platform velocity decreases below a certain threshold value, transmittance is also considerably reduced, which is associated with the achievement of too



**Figure 3.** Dependence of transmittance of individual squares on wavelength for various pulse energies and movable platform velocity. Pulse width is 300 fs, repetition rate is 500 kHz, NA = 0.25.



**Figure 4.** Multilayer structures in the form of squares of side  $1000 \,\mu\text{m}$  and  $500 \,\mu\text{m}$  (plan view) recorded within fused silica before (a) and after annealing in air at temperature  $1000^{\circ}\text{C}$  during 2 (b) h and 8 (c) h.



**Figure 5.** Dependence of transmittance of individual squares of multilayer structures on the wavelength for various pulse energies. Pulse width is 300 fs, repetition rate is 500 kHz, movable platform velocity  $375 \,\mu$ m/s, NA = 0.25.

high number of pulses per point during structure recording which results in the increase in modification density.

This study defined recording parameters for fused silica of structures having transmittance higher than 0.9 and retardance lower than 25 nm (purple curve in Figure 1, 3). Recording of several layers of such structures allowed to achieve higher uniformity of the resulting structure (Figure 4) and to retain transmittance higher than 0.8 arb.units (green curve in Figure 5) required for successful application of optical elements based on similar structures. Multilayer configuration of the resulting structure reduced transmittance only slightly. While the subsequent annealing of the specimen with recorded structures allowed to avoid undesired effects arising during recording due to material stresses around the recorded structures (Figure 4). The arising material stresses were caused by the fact that the layers are close to each other. Heat resistance of such nanostructures is explained by the fact that chemical recombination of oxygen whose molecules are involved in production of these structures is possible only at a temperature higher than  $1200^{\circ}$ C [13,17,18].

# Conclusion

Birefringent structure recording process within fused silica using focused ultrashort laser pulses was investigated herein. Dependence of the characteristics of recorded structures on laser recording parameters was analyzed. It was shown that combination of multilayer structure recording followed by annealing allowed to produce structures with high degree of uniformity, retardance lower than 25 nm and transmittance higher than 0.8.

#### Funding

The authors are grateful to the Russian Science Foundation for the financial support of these research within project 20-71-10103.

#### **Conflict of interest**

The authors declare that they have no conflict of interest.

#### References

- M. Beresna, M. Gecevičius, P.G. Kazansky. Opt. Mater. Express, 1, 783–795, (2011). DOI: 10.1364/OME.1.000783
- [2] A.E. Rupasov, P.A. Danilov, M.P. Smaev, M.S. Kovalev, A.S. Zolot?ko, A.A. Ionin, S.I. Kudryashov. Opt. i spectr., 128 (7), 918 (2020). DOI: 10.21883/OS.2020.07.49564.48-20
- [3] R. Stoian. Appl. Phys. A, **126**, 438 (2020).
  DOI: 10.1007/s00339-020-03516-3
- [4] L. Rapp, R. Meyer, R. Giust, L. Furfaro, M. Jacquot, P.A. Lacourt, J.M. Dudley, F. Courvoisier. Sci. Rep., 6, 34286 (2016). DOI: 10.1038/srep34286
- [5] S. Lavin-Varela, S. Madden, K. Yan, M. Ploschner, A.V. Rode, L. Rapp. Opt. Express, **30**, 6016–6036 (2022). DOI: 10.1364/OE.449230
- [6] S. Xu, H. Fan, Z.-Z. Li, J.-G. Hua, Y.-H. Yu, L. Wang, Q.-D. Chen, H.-B. Sun. Opt. Lett., 46, 536–539 (2021). DOI: 10.1364/OL.413177
- S.I. Kudryashov, P.A. Danilov, M.P. Smaev, A.E. Rupasov,
  A.S. Zolot'ko, A.A. Ionin, R.A. Zakoldaev. JETP Lett., 113,
  493–497 (2021). DOI: 10.1134/S0021364021080075
- [8] S.I. Kudryashov, P.A. Danilov, A.E. Rupasov, M.P. Smayev, A.N. Kirichenko, N.A. Smirnov, A.A. Ionin, A.S. Zolot'ko, R.A. Zakoldaev. Appl. Surf. Sci., 568, 150877 (2021). DOI: 10.1016/j.apsusc.2021.150877
- [9] M. Sakakura, Y. Lei, L. Wang, Y.-H. Yu, P.G. Kazansky. Light Sci. Appl., 9, 15 (2020). DOI: 10.1038/s41377-020-0250-y
- [10] G. Shayeganrad, X. Chang, H. Wang, C. Deng, Y. Lei, P.G. Kazansky. Opt. Express, **30**, 41002–41011 (2022). DOI: 10.1364/OE.473469

- [11] J. del Hoyo, R. Meyer, L. Furfaro, F. Courvoisier. Nanophotonics, 10, 1089–1097 (2021).
  DOI: 10.1515/nanoph-2020-0457
- [12] C. Vetter, R. Giust, L. Furfaro, C. Billet, L. Froehly, F. Courvoisier. Materials, 14, 6749 (2021).
   DOI: 10.3390/ma14226749
- [13] M. Lancry, B. Poumellec, J. Canning, K. Cook, J.-C. Poulin,
  F. Brisset. Laser Photonics Rev., 7, 953–962 (2013).
  DOI: 10.1002/lpor.201300043
- [14] I.V. Gritsenko, M.S. Kovalev, N.G. Stsepuro, Y.S. Gulina, G.K. Krasin, S.A. Gonchukov, S.I. Kudryashov. Laser Phys. Lett., 19, 076201 (2022). DOI: 10.1088/1612-202X/ac7136
- [15] M. Beresna, M. Gecevičius, P.G. Kazansky, T. Gertus. Appl. Phys. Lett., 98, 201101 (2011). DOI: 10.1063/1.3590716
- [16] R. Drevinskas, P.G. Kazansky. APL Photonics, 2, 066104 (2017). DOI: 10.1063/1.4984066
- [17] E. Bricchi, P.G. Kazansky. Appl. Phys. Lett., 88, 111119 (2006). DOI: 10.1063/1.2185587
- [18] Y. Wang, M. Cavillon, N. Ollier, B. Poumellec, M. Lancry. Phys. Status Solidi A, **218**, 2100023 (2021). DOI: 10.1002/pssa.202100023

Translated by E.Ilyinskaya