MODIFICATIONS IN GLASS INDUCED BY FEMTOSECOND LASER RADIATION

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Abstract

The interaction of laser radiation with glass becomes nonlinear, when the electric field strength of a femtosecond laser exceeds the dielectric field strength of the glass. This effect can lead to a possible void formation along which the material can be precisely shaped. The parameter range of the laser-matter interaction was studied in order to find the optimal parameters for creating 3D void structures.

Key words: fs laser; glass; defect, void

Introduction

Locally confined permanent modifications inside a glass are induced via nonlinear absorption mechanisms using laser radiation with high intensity and ultra-short pulse duration [1]. If the interaction between the laser radiation and the material is nonlinear, it results in refractive index changes, stress formation, plasma, void (defect) formation etc. [1, 2, 3]. These modifications are spatially confined to the focal volume, which offers a possibility to create optical components like micro lenses, Fresnel lenses, gratings, and waveguides inside transparent bulk materials, as well as complex three-dimensional integrated optical and photonic devices [4, 5].

Inducing nanometer sized voids in the material is of great interest because of the possibility of 3D material processing. By systematic creating of micro-/nanovoids we try to produce a form or contour and cut the material along this contour. Arrangements of small voids would then lead to 3D structures with smooth surfaces. Fracture lines or cracks are not suitable for 3D material processing, they are much larger than voids, of irregular shape and would cause damaged surfaces of the 3D cut out object [6].

Theoretical

The interaction of the laser radiation with glass involves several processes that determine the energy dissipation in the matter: carrier excitation, thermalisation, thermal and structural processes (see Figure 1). These processes occur on time scales of femtosecond to nanoseconds after the excitation: first, the photons are absorbed in the matter (photon-electron interaction) in about ten femtoseconds, followed by the interaction of carriers. In a few picoseconds, the electrons start to transfer their energy to the lattice which can last up to nanoseconds depending on the material. Finally, the excited matter relaxes in some microseconds by phonon-phonon interaction [1, 6].
Most of the glass materials are transparent for infrared (IR) laser radiation as the energy of the single photons is smaller than the band gap\(^1\) of the glass. For high laser intensity, the probability of nonlinear absorption in the material increases rapidly and the electrons may be excited into the conduction band. There are two ionisation mechanisms: tunnel ionisation due to bending of the band gap, and multi-photon ionisation as a result of multi-photon absorption [7]. The probability of multi-photon absorption is dependent on the intensity of laser radiation - two-photon absorption becomes significant at a few MW/cm\(^2\), three- and more photon absorption demand GW/cm\(^2\) to TW/cm\(^2\) [8].

The interaction of thermalized electrons with the phonon system of the solid initiates melting and boiling of the material. If the energy input into the material is sufficient to exceed the critical electron density, plasma formation sets in, and material damage can occur. Several nonlinear effects have to be considered, the most important one being self focusing. It occurs above a critical laser pulse peak power which is given for a Gaussian pulse by

\[
P_{cr} = 1.8962 \frac{\lambda^2}{4\pi n_0 n_2},
\]

where \(\lambda\) is the laser wavelength and \(n_0\) and \(n_2\) are the linear and nonlinear refractive indices given by \(n(I) = n_0 + n_2 I\), where \(I\) is the intensity of the radiation [8]. The critical powers for the glasses used are \(3.08 \times 10^6\) W for borosilicate glass and \(3.68 \times 10^6\) W for fused silica glass.

To produce small defects without cracks, a strongly focused fs laser radiation has to be used to reach the threshold intensity for the optical breakdown; the short pulse length causes fast energy transfer to the lattice. Above a threshold intensity a plasma is formed, inducing material damage. Together with shock-wave propagation, a cavity surrounded by compacted material may be created.

Modification of the glass induced by multi-photon absorption is given by the input energy of the laser radiation, ns pulse irradiation may result in microcracks with dimensions in order of hundreds of micrometers, fs pulse irradiation can cause refractive index modifications in form of filaments or even voids. For high-resolution micro-structuring, achieving a very small focus is important. Therefore highly focused fs-laser radiation is used for the investigations.

\(^1\)It is effective to use the band structure approach for glass even if it is non-crystalline.
Experimental setup

In our experiment, a high repetition rate femtosecond laser system Impulse Clark MXR Inc. was used with the parameters summarized in Table 1. The laser radiation was focused under the sample surface to generate defects in the glass volume. Three objectives with focal radii 3.5, 2.8, and 1.2 µm were used to induce the nonlinear absorption in the glass and locally heat the material leading to void generation.

<table>
<thead>
<tr>
<th>wavelength</th>
<th>pulse duration</th>
<th>max. frequency</th>
<th>max. pulse energy</th>
<th>pulse peak power</th>
<th>M^2 (beam quality factor)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1030 nm (≈1.2 eV)</td>
<td>180 fs</td>
<td>1024 kHz</td>
<td>7 µJ</td>
<td>34 MW</td>
<td>1.3</td>
</tr>
</tbody>
</table>

Results and discussion

First, the optics with 3.5 µm focal radius was employed. For a single pulse, the defects were hardly detectable by optical microscopy even for a high pulse intensity, but they were observable for 10 and more laser pulses shot at the same point with a frequency of 1024 kHz (Figure 2).

<table>
<thead>
<tr>
<th>band gap [eV]</th>
<th>borosilicate glass (BS)</th>
<th>fused silica glass (FS)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.7 eV (≈264 nm)</td>
<td>9 eV (≈138 nm)</td>
<td></td>
</tr>
<tr>
<td>refractive index n_0 (1060 nm)</td>
<td>1.51</td>
<td>1.45</td>
</tr>
<tr>
<td>nonlinear refractive index n_2 [10^{-16} cm^2 W^{-1}]</td>
<td>3.45</td>
<td>3.00</td>
</tr>
<tr>
<td>critical power P_{cr} [MW]</td>
<td>3.08</td>
<td>3.68</td>
</tr>
<tr>
<td>melting temperature [°C]</td>
<td>559</td>
<td>1900</td>
</tr>
</tbody>
</table>

| focal radius* [µm] | 3.5 | 2.8 | 1.2 |
| numerical aperture | 0.2 | 0.4 | 0.65 |
| focal length* [mm] | 12.5 | 10 | 4 |
| pulse intensity range* [TW/cm^2] | 33-76 | 12-16 | 35-86 |
| maximal pulse power P_{max} [MW] | 6.3 | 1.4 | 0.78 |
| smallest pulse energy to achieve defects [µJ] | 1.3 | 0.28 | 0.16 |
| pulse frequency [MHz] | 1.024 | 0.128 | 0.128 |
| average pulse power P_{AV} [mW] | 1331-3072 | 38.4-51.2 | 20.5-51.2 |
| smallest pulse number to achieve defects | 10 | 100 | 1 |
The smallest laser pulse energy to produce defects was 1.3 µJ corresponding to a pulse power of 6.3 MW, which exceeds the critical power for BS glass (3.08 MW) when self-focusing of the laser radiation occurs. This leads to filament generation. The elongation of the defect can be further assigned to the defocusing of the laser radiation before the focus due to high carrier concentration in the beam center. This is followed by a decrease of the intensity and impedes achieving voids. Influencing the irradiated region by inducing subsequent pulses gradually heats the irradiated volume resulting in a noticeably higher processing effect. The induced defects are clearly defined and evidence a length up to about 100 µm in the radiation direction and 6 µm in the cross section. The dark central area of the defects probably indicates a different density of the material.

![Figure 2: Defects induced by optics with 3.5 µm focal radius, 10 pulses, f=1024 kHz, on top plane view of the smallest defects, down side view of defects caused by an energy of 1 µJ (left) and 3.5 µJ (right). In the lower images, the laser radiation comes from the right side.](image)

Second, the laser radiation was focused into an area with 2.8 µm focal radius with pulse peak intensities between 12 and 16 TW/cm². Higher intensity were not used in order to prevent destruction of the lens. No defects were obtained by single-pulse irradiation. Therefore the pulse number was increased to 100 at a pulse repetition rate of 128 kHz. The smallest pulse peak power to produce the defect was 1.4 MW, which is about half the power needed for self-focusing, and the filaments are not generated any more. The diameter of defects was less than 3 µm and their length was up to 40 µm. Both dimensions are about twice smaller than the dimensions in the previous experiment with 3.5 µm focal radius (Figure 3). The effect of subsequent pulses is significantly reduced by the small pulse frequency (8 times smaller than in the former case). The distance between two following pulses is in order of microseconds, reaching the end of the resolidification phase.

Third, the optics with 1.2 µm focal radius was used for both BS and FS glasses. The pulse number varied from 1 to 5 pulses with repetition rate 128 kHz and pulse peak intensity ranging from 35 to 86 TW/cm². The intensity was sufficiently high to produce defects even after irradiation with a single pulse. The smallest energy needed for obtaining defects was 200 nJ (corresponding pulse peak power 0.97 MW) for BS glass and 160 nJ (0.78 MW) for FS glass, respectively. If this pulse intensity could be applied for the optics with 2.8 µm focal radius, the corresponding maximal pulse power would be 4.3 MW, which exceeds the critical power for both glasses. Therefore using this optics is not advantageous for creating voids.
The defects in FS are much sharper and better defined. Surprisingly, the energy needed to achieve defects in FS glass is smaller than in BS (see Figures 4 and 5).

Figure 3: Optical image of the smallest defects induced by optics with 2.8 μm focal radius, 100 pulses, f=128 kHz. Left: plane view, right: side view. Defects were observed for pulse energy / peak power intensity 280 nJ /11 TW/cm² and higher (right).

This can be ascribed to the thermophysical parameters of the material, e.g. larger band gap which causes a bigger process selectivity, higher melting temperature (see Table 2), and larger hardness of FS.

For higher energy and higher number of pulses, the defects are in the plane view surrounded by dark areas, which might be an evidence of strain (refractive index modification). The dark areas are not likely to be cracks because no damage of the material was observed in the side view. In BS glass, strain can be observed for ≥5 pulses with pulse energies 280 nJ and higher, in the FS, strain appears for 2 pulses and 210 nJ.

Figure 4: Optical image of defects induced in BS glass by optics with 1.2 μm focal radius, 1-5 pulses, f=128 kHz. On the left: plane view of the defects, on the right: side view (laser radiation coming from the bottom).
Consequently, the absorption of the laser radiation is limited to a very small volume. For BS, the intensity to initiate the multi-photon absorption is sufficient in the focal volume already, but is not high enough to induce voids or cracks.

To verify the presence of voids, a field of defects caused by single laser pulses with 350 nJ pulse energy (intensity 76 TW/cm$^2$) and by optics with 4 mm focal length was induced in FS and subsequently the substrate was broken along these defects. Some of the defects were visible by confocal microscopy indicating the presence of voids. The cut edge was then covered by a metal layer and studied by scanning electron microscope (SEM, see Figure 6). Because of the large roughness of the cut plane, only two defects were detected. One of them is a crack and another one is a void. Both defects evince the presence of densified material. The void is about 2.5 $\mu$m long and 0.3 $\mu$m wide, the size of the densified material surrounding the void is about 4.5 x 1.5 $\mu$m.

In the experiments with the optics with larger focal radius, the generation of voids was not achieved even after multi pulse irradiation. It means that the effect of subsequent pulses for pulse frequencies 1024 and 128 kHz contributes to the defect size only.
Conclusion

The influence of several laser parameters on the defect creation in BS and FS glasses was studied. When the laser radiation was focused into a region with radius 3.5 µm, the defects could clearly be observed after irradiation of 10 pulses with peak intensity 53 TW/cm². The defects are very probably refractive index modifications. For the irradiation with focal radius 2.8 µm, the average laser power had to be reduced down to 20 mW and therefore smaller defects were observed first after 100 pulses. The best results were obtained for 4 mm focal length (focal radius 1.2 µm), where the material modification occurs even after a single pulse irradiation. For this focal length, the presence of voids in FS glass was assumed and proved by a SEM measurement.

References


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