Fundamental processes of refractive index modifications during femtosecond laser waveguide writing

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ABSTRACT

By using focused ultrashort pulsed laser radiation refractive index modifications are induced in glass in order to generate optical components. The understanding of physically fundamental processes induced by laser radiation is the basis for the systematic control and maximization of the refractive index change for the realization of three-dimensional, optical components for integrated optics like in-volume waveguides. In this paper fundamental processes which are induced by focused laser radiation in the volume of borosilicate glass D263 and fused silica are investigated. The glass materials are structured by laser radiation in the infrared spectral range (λ = 1045nm). By using femtosecond laser pulses with high repetition rates (f = 500 kHz), thermal processes like heat accumulation effects are induced leading to heat affected zones and thus waveguide cross sections with dimensions larger than the focal spot. The absorptivity during modification in relation to the applied pulse energy is measured for different repetition rates in both glass materials. Furthermore, the laser induced structural change in the glass matrix by the increase of three- and four-membered ring structures is proved with Raman spectroscopy.

Keywords: in-volume waveguide, femtosecond laser radiation, refractive index change, fundamental processes, glass materials

1. INTRODUCTION

For several years in-volume waveguides are fabricated using tightly focused femtosecond laser pulses in glass and crystals. Non-linear absorption processes like multiphoton absorption and avalanche ionization in the focal volume result in the deposition of energy and lead to a strongly localized refractive index change of the irradiated material [1]. The control and maximization of the refractive index change is beneficial for the fabrication of photonic devices. Applying the technique of direct femtosecond laser writing integrated optical components such as waveguides, phase plates, beamsplitters and combiners can be fabricated [2,3,4]. Furthermore, active devices like amplifiers and waveguide lasers are realizable [5].

At high repetition rates (f > 200 kHz), waveguides with circular cross sections much larger than the focal volume are observed with transmission light microscopy. The compacted region surrounding the central modification is explained with heat accumulation effects [6]. On the other hand, the formation of a transient electronic excitation is observed which can lead to the formation of permanent color centers [7,8]. Nonelastic thermomechanical stress and/or the formation of color centers change the absorption properties and the refractive index of the irradiated material. For the fabrication of optical devices, a spatial control of the induced refractive index modification is necessary. Therefore, the fundamental processes regarding the absorbed pulse energy, the impact of heat accumulation effects and modified ring structures which are structural atomic changes in the material are investigated in this work.

1.1 Ring structures

The topology of glass is characterized by the amount and arrangement of different ring structures. A n-membered ring is defined by the shortest connection which is made up of 2n silicon-oxygen-bonds [9]. A four-membered ring consists of eight silicon-oxygen-bonds for example. In untreated glass the maximum of occurrence is at six-membered rings [9,10]. By the influence of laser radiation the maximum of occurrence can change. With molecular dynamical simulations the impact of laser-induced shock waves is calculated [9]. After relaxation of the material the maximum of occurrence is at...
three- and four-membered rings whereas the occurrence of six-membered rings decreases from 33 % to 25 % [9]. The bonding angles between silicon and oxygen atoms are smaller for three- and four-membered rings than for six-membered rings. Therefore, the atomic distances are decreased and the material has a higher density resulting in a positive refractive index change. Furthermore, the redistribution in the occurrence of ring structures means the formation of defects in the atomic structure. Thereby changes in the electronic characteristics of the material are induced [9]. With Raman spectroscopy the oscillation of oxygen atoms is detected in terms of a coherent movement which is excited in three- and four-membered rings [10,11]. By excitation with an argon-ion-laser two maxima of the Raman spectrum at $k = 605 \text{ cm}^{-1}$ and $k = 490 \text{ cm}^{-1}$ are measured which represent the excitation of three- and four-membered rings [12], respectively. The silicon atoms are not or only rarely excited [11].

2. EXPERIMENTAL SETUP

2.1 Waveguide fabrication

The glass materials borosilicate glass D263 and fused silica are irradiated with femtosecond laser radiation from the fiber laser Satsuma from Amplitude Systèmes with the central wavelength $\lambda = 1030 \text{ nm}$, the pulse duration $t = 400…500 \text{ fs}$ and the maximum average output power $P_{av} = 5 \text{ W}$. The repetition rate is variable in the range $f = 0.1…27 \text{ MHz}$. The laser radiation is focused with a high numerical aperture microscope objective ($\text{NA} \geq 0.4$) into the volume of the material. The surfaces of the glass samples have to be optically polished. The focal position is placed about 200 $\mu\text{m}$ underneath the surface. By moving the glass sample transversally with an air-bearing translation stage a local modification is generated in the material. In the experiment the laser process parameters pulse energy, repetition rate and writing velocity as well as the numerical aperture of the microscope objective are changed systematically.

3. CHARACTERIZATION

3.1 Transmission light microscopy

All written waveguides are investigated using transmission light microscopy. Thus, the dimensions like width and height of the waveguide cross section as well as the width of the waveguide observed from the top can be determined. For cross sections the end facets of the samples which lie perpendicularly to the written waveguides have to be polished. Especially, the size of the heat affected zone surrounding the central modification of the waveguide is investigated by transmission light microscopy. Furthermore, induced micro cracks can be observed in dependency on the applied laser parameters.

3.2 Intensity distribution in the near field

The intensity distributions in the near field of the waveguides are detected using helium-neon-laser radiation which is focused onto the facet of the polished waveguide cross sections with a microscope objective. At the other end of the waveguide the guided radiation is collected using another microscope objective. The intensity distribution is collected by a CCD-camera. The black-and-white pictures of the camera are normalized and converted into false color images for comparison.

3.3 Absorption measurement

Femtosecond laser pulses are absorbed by nonlinear absorption processes. The value of the nonlinear absorptivity in glass materials depends on the applied pulse energy and the repetition rate of the laser pulses [13]. The experimental setup used in this work consists of a power detector which is placed underneath the glass sample (Figure 1).

Measurements of the transmitted pulse energy are taken during modification when the focal volume is scanned through the sample (Figure 1a) and when the focal volume is slightly above the sample for reference (Figure 1b). The applied power is measured without any sample in the light path (Figure 1c). The scattered radiation is neglected.

The laser radiation is focused perpendicularly to the surface into the volume of the glass sample. When high numerical aperture objectives are used for waveguide writing an additional optic has to be added underneath the sample to collect all of the transmitted radiation and guide it completely to the power detector for measurement. The measured signal
collected by the power detector is logged with an oscilloscope. There is a linear correlation between the measured voltage of the oscilloscope and the applied pulse energy.

With the measurement of the transmitted, the reference and the applied power (Figure 1) the absorbed pulse energy in the material can be calculated with consideration of reflection at the sample surfaces.

3.4 Raman spectroscopy

With Raman spectroscopy the laser modified waveguide cross sections are investigated in terms of atomic oscillation modes of oxygen atoms in glass. Laser radiation with the wavelength $\lambda = 532$ nm is focused onto the polished surface of waveguide cross sections by a microscope objective with the numerical aperture $N_{\text{AO}} = 0.85$. A frequency doubled Nd:YAG solid state laser is operated at continuous wave mode. The power of the laser radiation is reduced to a few milliwatts behind the microscope objective in order to not change the initial laser modification during measurement. The spectroscopic lattice constant $g = 556$ nm and the measurement range $k = 1 – 1790$ cm$^{-1}$ are selected. The exposure time is $t = 30$ s for each measurement whereof 60 measurements are accumulated. For each measurement the measurement period is $t = 30$ min.

4. RESULTS AND DISCUSSION

4.1 Structural and optical characteristics

The picture of a waveguide cross section in D263 taken with transmission light microscopy shows an elongated shape in the beam propagation direction for the repetition rate $f = 100$ kHz (Figure 2, top left). The intensity distribution in the near field shows two light guiding regions on the left and the right side of the induced structure (Figure 2, top middle) which are located by comparison with the micrograph. The false color image further illustrates the intensity distribution in the near field of the waveguide showing both intensity maxima lying close to the modification symmetrically (Figure 2, top right). It is supposed that a stress induced refractive index change leads to such light guiding areas.
For \( f = 500 \text{ kHz} \) the intensity distribution in the near field shows a circular shape with radial periodical light guiding structures with several intensity maxima (Figure 2, bottom middle and right). By comparison of the near field picture with the micrograph (Figure 2, bottom left) the light is determined to be guided in the modified outer region of the structure cross section. The circular region has a higher density than the unmodified material and exhibits a higher refractive index change making light guiding possible. In the dark region in the center of the modification which can be seen in the micrograph no light is guided. Therefore, a negative refractive index change is supposed here. For other waveguides structured with repetition rates \( f > 500 \text{ kHz} \) there is a similar circular shape detected in the micrographs as well as in the intensity distribution in the near field showing the light guiding areas. The circular structure in the waveguide cross sections is generated by heat accumulation effects. Due to the high repetition rate the temperature of the materials is increasing pulse by pulse until saturation. In contrast to single pulses the temperature does not return to the initial material temperature but increases during several laser pulses [6]. By using high repetition rates thermal induced refractive index changes are generated due to cumulative heating of the material. A radial structure with compacted material is formed in which light is guided.

In summary there is a large difference in the intensity distribution in the near field of propagating radiation in waveguides fabricated with \( f = 100 \text{ kHz} \) and \( f \geq 500 \text{ kHz} \) due to the described heating effects.

### 4.2 Absorptivity

In general the absorptivity \( A \) is increasing with increasing applied pulse energy and repetition rate for both investigated glass materials D263 (Figure 3, left) and fused silica (Figure 3, right).

In D263 the absorptivity increases for \( f = 500 \text{ kHz} \) from \( A = 10 \% \) of the applied energy at \( E_p = 50 \text{ nJ} \) to \( A = 88 \% \) at \( E_p = 600 \text{ nJ} \) (Figure 3, left). For larger pulse energies a saturation is observed with an absorptivity of about \( A = 90 \% \).
The increase of the absorptivity is similar for \( f = 100 \) kHz compared to \( f = 500 \) kHz but the absolute values are smaller. For \( f = 100 \) kHz the absorptivity is increasing from \( A = 33 \% \) at \( E_p = 100 \) nJ to \( A = 78 \% \) at \( E_p = 1 \) \( \mu \)J. The saturation is shifted to larger pulse energies \( E_p > 1 \) \( \mu \)J.

For larger repetition rates the temperature in the focal volume and the surrounding material is larger due to heat accumulation effects and a larger number of electrons is thermally excited into the conduction band. By increasing number of laser pulses absorbed in one spot the absorption volume increases leading to a higher value of absorptivity. Therefore, a larger repetition rate leads to a larger absorptivity in the glass material.

In fused silica the absorptivity increases for \( f = 100 \) kHz from \( A = 14 \% \) at \( E_p = 100 \) nJ to \( A = 66 \% \) at \( E_p = 1.1 \) \( \mu \)J (Figure 3, right). The pulse energy for saturation is \( E_p > 1.1 \) \( \mu \)J. However, a characteristic effect is observed for \( f = 500 \) kHz in fused silica. Until \( E_p = 450 \) nJ with \( A = 54 \% \) the absorptivity is increasing. For \( 500 \) nJ \(<\ E_p \ <\ 800 \) nJ the absorptivity remains nearly constant at \( A \approx 52 \% \). For larger pulse energies the absorptivity increases again up to \( A = 77 \% \) at \( E_p = 1 \) \( \mu \)J.

Up to about \( E_p = 500 \) nJ the waveguides observed by transmission light microscopy from the top exhibit a homogenous central modification (not shown here). For \( 500 \) nJ \(<\ E_p \ <\ 800 \) nJ the structure becomes inhomogeneous with irregular modifications in the direction of the waveguides. These irregularities are in the pulse energy range where the absorptivity is decreasing. For \( E_p > 800 \) nJ an additional outer region is formed surrounding the central modification of the waveguides (not shown here). The modification is mainly driven by heat accumulation effects. Although the central modification still exhibits an inhomogeneous structure the absorption volume is larger and leads to compacted material in the surrounding area (compare Figure 2, bottom left).

With quite high values of absorptivity of more than \( A = 90 \% \) of the applied pulse energy in D263 and around \( A \approx 80 \% \) in fused silica energy efficient modification and structuring of glass materials is realizable using femtosecond laser radiation.

4.3 Raman spectroscopy

Raman spectroscopy is performed for two positions on a waveguide cross section which is structured with the repetition rate \( f = 100 \) kHz, the pulse energy \( E_p = 1 \) \( \mu \)J, the scan velocity \( v = 1 \) \( \text{mm/s} \) and the numerical aperture \( NA_{OB} = 0.65 \).
The intensity distribution shows relatively wide maxima (Figure 4, right), as the spectra of amorphous materials are inhomogeneously broadened in contrast to spectra of crystalline materials [14]. As the wave numbers $k = 605 \text{ cm}^{-1}$ and $k = 490 \text{ cm}^{-1}$ represent the excitation of oscillations of three- and four-membered rings, the intensity values at these wave numbers are observed in particular. In the measured spectrum the intensity maxima are obtained at $k = 603 \text{ cm}^{-1}$ and $k = 490 \text{ cm}^{-1}$ (Figure 4, right) and thus are in accordance with literature values. The absolute values of the intensity depend on the measurement position of the waveguide cross section. The spectrum of the dark region in the waveguide (position 1) with a negative refractive index change is similar to the reference spectrum of the unmodified material. There is only a slight increase of the intensity values at $k = 603 \text{ cm}^{-1}$ but the relative height of the maximum is unchanged. Therefore, there is no evidence for a structural change related to oxygen atoms in this region of the waveguide.

A clear change in the Raman spectrum is observed for position 2 with a positive refractive index change compared to the unmodified material (Figure 4, right). The increase of the intensity values for $k = 603 \text{ cm}^{-1}$ represents an increase in the amount of three-membered rings of silicon and oxygen atoms. Moreover, there is an increase of the intensity value at $k = 490 \text{ cm}^{-1}$ whereby the increase of four-membered rings in the region of positive refractive index change is established. Additionally to other effects like thermal or other structural effects which are responsible for refractive index changes, the increase of three- and four-membered rings leads to a compaction of the material. The positive refractive index change is explained with a structural change and a rearrangement of silicon and oxygen atoms in fused silica.

5. SUMMARY

In-volume waveguides are fabricated in borosilicate glass D263 and fused silica in order to investigate fundamental processes induced by laser radiation. The structural and optical changes in the material are characterized using optical light microscopy and near field measurements. For $f = 100 \text{ kHz}$ two light guiding regions are observed in D263 whereas for $f \geq 500 \text{ kHz}$ a circular shape of intensity maxima surrounding the central modification is determined. Furthermore, the absorbed laser energy in the materials is measured and related to the applied pulse energy. In general, the absorptivity is increasing with increasing pulse energy. For fused silica there is a characteristic and sudden decrease of the absorptivity for $E_p = 800 \text{ nJ}$ which is explained with a more inhomogeneous structure in the central modification before at $E_p > 800 \text{ nJ}$ the modification is mainly driven by heat accumulation effects leading to higher absorptivity again. With Raman spectroscopy the excitation of ring structures consisting of silicon and oxygen atoms is investigated. After modification there is an increase in two special intensity maxima in the Raman spectra demonstrating an increase in three- and four-membered ring structures. Ring structures with a decreased bonding angle lead to compacted material and a positive refractive index change.
REFERENCES


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