The influence of self-focusing and filamentation on refractive index modifications in fused silica using intense femtosecond pulses

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Abstract

The interaction of focused femtosecond infrared laser pulses at 1 kHz repetition rate with bulk fused silica is thoroughly investigated. The interplay between self-focusing and filamentation of the laser pulses is analyzed for a broad range of focusing conditions. It is shown that even in the case of very tight focusing, filamentation is observed as evidenced by the scanning electron microscope (SEM) pictures. Preliminary results show that using such a tight focusing geometry and at input powers above the critical power for self-focusing in silica, waveguide structures with elliptical cores are inscribed within the glass by moving the sample perpendicular to the laser beam propagation direction.

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1. Introduction

In recent years, the interaction of ultra-short powerful infrared laser pulses with transparent optical materials has attracted a lot of attention. Beyond the fundamental investigations on the nonlinear propagation of ultra-short pulses in different optical materials [1–5], numerous technological applications have emerged. Among them are the development of three-dimensional photonic structures in bulk optical materials, such as waveguides [6–9], gratings [10], couplers [11], and optical memories [12].

Optical breakdown (OB) [13–15] and laser pulse filamentation (FL) [16–18] have been identified as two principal nonlinear electronic excitation mechanisms leading to photo-structural modifications in glass. Such modifications appear in the form of refractive index changes, when a femtosecond infrared laser pulse is focused inside a transparent optical material. These processes can be initiated...
at laser intensities, typically of the order of $10^{13}$ W/cm$^2$ at the focal point, which are sufficiently high to induce nonlinear absorption in silica glass. Filamentation results from the dynamical balance between two effects: self-focusing due to the nonlinear Kerr effect and self-defocusing associated with the plasma formation in the self-focus region. The plasma is generated mainly through multiphoton excitation (MPE) of electrons from the valence band to the conduction band in the glass [16]. Filamentation is usually accompanied by a strong broadening of the laser spectrum extending from the near-infrared to the visible, which is called super-continuum generation or white light laser [3,16]. Optical breakdown, on the other hand, is characterized by the formation of localized dense plasma around the geometrical focus, caused by initial multi-photon excitation of electrons, followed by inverse Bremsstrahlung, impact ionization, and electron avalanche processes. It should be mentioned, however, that the relative contributions of OB and FL in plasma formation mostly depend on the pulse duration and external focusing condition [3,19]. The competition between OB and FL in silica glass was recently reported by our group [20]. The photo-generated plasma resulting from OB and/or FL processes transfer its energy to the glass network which leads to local heating and melting [21,22]. The molten glass is rapidly cooled; this leads to re-solidification of the silica glass and increase of density and refractive index in the irradiated region.

In this work, we show visual evidences of optical breakdown, self-focusing and filamentation which play an important role in the formation of geometrical structures. At sufficiently high pulse energies, the index modifications around the geometrical focus are accompanied by an irregular void-like structure. The spatial extent of such structure depends on the number of laser pulses per focal volume. Even under a very tight focusing geometry, we observed multiple refocusing of the laser pulse giving rise to repeated elongated zones, i.e., beyond the geometrical focus. By moving the sample perpendicular to the beam propagation axis in a tight focusing geometry, we produced tracks with an elliptical shape and asymmetric index profile which were able to guide laser light at 633 nm.

### 2. Experimental setup

A Spectra Physics chirped-pulse-amplification (CPA) Ti:sapphire laser system consisting of a mode-locked oscillator (Maitai: 300 mW, 80 MHz, FWHM ~40 nm) and a 2 W Spitfire regenerative amplifier, was used to generate femtosecond pulses at a central wavelength of 810 nm (line-width ~30 nm) with 1 kHz repetition rate. The sample was a polished bulk of pure fused silica. The laser beam diameter was ~4.6 mm (at 1/e$^2$ intensity, measured with a CCD camera). The beam, after passing through a vacuum spatial filter, was focused inside the glass using various standard achromatic microscope objectives (Melles-Griot). The schematic layout of the experimental setup is shown in Fig. 1. To visualise the evolution of self-focusing and filamentation versus focusing geometry, we chose microscope objectives with effective NAs of 0.03 ($1\times$, $f = 73.8$ mm) (long focal length), 0.075 (4$\times$, $f = 30.8$ mm) (intermediate case), 0.136 (10$\times$, $f = 16.9$ mm) (short focal length), and 0.85 (63$\times$, $f = 3.1$ mm) (very tight focusing). The beam spot size was estimated to be approximately 17.4, 7.2, 4.0, and 0.8 μm in silica, for $1\times$, 4$\times$, 10$\times$, and 63$\times$ objectives, respectively, assuming an undisturbed Gaussian beam profile. The temporal width of the transform-limited pulses was measured by an optical autocorrelator (Positive Light SSA) to be approximately 45 fs before the focusing lens. The input pulse energy was controlled by a half-wave plate placed before the compressor together with calibrated neutral density filters inserted before the sample, and was varied from 0.1 to 20.0 μJ. A shutter was used to
control the number of pulses incident onto the sample. Another objective lens (20×, NA: 0.4) was used to image the focal region onto a CCD camera (Hitachi KP-160) from the side. The video signal of the camera was recorded by a frame grabber card (Spiricon LBA-300) connected to a computer. After exposure, the irradiated spots were inspected under a phase contrast microscope (magnification: 100–400×). Each location in the glass was exposed only once by manually moving the sample in the vertical direction perpendicular to the laser beam axis (Fig. 1).

3. Experimental results and discussion

The photo-emission from the side, including plasma fluorescence and scattered laser light, was recorded on video for different incident pulse energies. The sample was exposed for period varying from 0.01 to 10 s corresponding to 10 and 10,000 laser pulses or shots, respectively. Typical images associated with 10,000 and 10 shot irradiations using 1× objective are shown in Figs. 2(a) and (b), where only the last video frame has been displayed. During laser irradiation we could observe, with the naked eyes from the side, a self-trapped reddish plasma column (namely a filament) with a typical length ranging from a few μm to approximately 1–2 mm. Note that due to a limited field of view the beginning portions of the filaments formed at higher pulse energies were not imaged. A spectrometer (Spectra Pro500i-Acton Research, grating: 300 grooves/mm) equipped with an ICCD (intensified charge-coupled device) camera was used to measure the spectrum of the plasma column from the side [20]. The extended white strip(s) associated with the filament shown in each CCD image corresponds to the dominant laser scattered signal at 810 nm, together with a plasma fluorescence near 650 nm (Fig. 3). The latter peak could be associated with some impurities or photo-generated defects (color centers) inside the glass. A similar femtosecond laser pulse-generated fluorescence band near 630 nm was recently reported in silica [23]. The observed peak at 450 nm corresponds to an isotropic plasma emission associated with optical breakdown around the geometrical focus [20].
We note that the filament length increases with pulse energy with its leading edge moving towards the lens (Fig. 2). This behavior can be well described qualitatively by self-focusing effect in light of the moving-focus model\[17,24\]. That is, a collimated Gaussian beam with a radius $a$ (at $1/e^2$ intensity), wave number $k$, and instantaneous power $P$ propagating through a Kerr medium with a linear and nonlinear index of refraction of $n_0$ and $n_2$, self-focuses at a position $z_f$ measured from the medium’s input surface [24]:

$$z_f = \frac{0.367ka^2}{\left\{\left[\left(\frac{P}{P_{\text{crit}}}\right)^{1/2} - 0.852\right]^2 - 0.0219\right\}^{1/2}}.$$  \hspace{1cm} (1)

Here $P_{\text{crit}} = 3.77\lambda^2/(8\pi n_0 n_2)$ represents the critical power for self-focusing in the medium ($\sim$4.4 MW for fused silica [16], $n_2 \sim 3 \times 10^{-16}$ cm$^2$/W) and $\lambda$ is the wavelength. In case of focusing with an external lens with focal length $f$, the position of first self-focus will shift to $z'_{f} = z_{f}f/(z_{f} + f)$ (where $z'_{f}$ specifies the beginning of the filament) as if there were two lenses one with focal length $f$ and the other, $z_f$.

At higher laser energies, multiple filaments occur (Fig. 2). They fuse towards the geometrical focus. Some inhomogeneities in the laser beam’s spatial profile could significantly trigger the formation of these structures, as already observed in water [25]. They arise from localized small scale self-focusing across the beam profile. These results suggest that controlled refractive index patterning of bulk optical materials by shaping the laser beam profile might provide promising applications for industrial laser micro-machining.

A very strong scattered laser signal appeared around the geometrical focus at an input energy per pulse of approximately 17 $\mu$J as shown in Fig. 2(a) and (b). In fact, by increasing the pulse energy, both intensity and free electron density increase around the geometrical focus due to the geometrical convergence of the whole beam. At such an input energy, the intensity in the focal volume exceeds the damage threshold so that the plasma density becomes sufficiently high to induce a permanent structural modification in the glass. The correlation between such observed signals and the real structural damages in glass was verified by the optical microscope pictures of the structures written without (Fig. 2(c), top) and with (Fig. 2(c), bottom) the appearance of the strong scattered signal. In Fig. 2(c) (bottom), a void-like morphology with a high index contrast is observed. This is similar to what was referred to in [12] as a micro-explosion around the geometrical focus. This should be contrasted to the smooth line created at lower pulse energy (Fig. 2(c), top). Such observation indicates that the plasma density along the filament and away from the geometrical focus is not sufficiently high to induce the structural damage in silica. Note that the appearance of this strong scattered signal around the geometrical focus at 17 $\mu$J is apparently irrelevant of the number of pulses (i.e. 10 versus 10,000), although some cumulative effects result in a higher intensity at 10,000 shots. Fig. 4 shows this variation of the intensity of the scattered laser light as a function of the number of shots for 17 $\mu$J-pulses. No significant change in the filament’s length is observed whereas the overall scattered laser intensity along the filament increases with the number of shots. Recent investigations on surface damage in silica glass induced by focused femtosecond laser pulses (800 nm, 150 fs, 1 kHz, 0.5 mJ) have revealed the formation of micro-holes at the irradiated region. This was accompanied by the ejected molten mate-

![Fig. 4. Variation of the accumulated intensity of scattered laser light from the damage zone in terms of laser shots. The input pulse energy was 17 $\mu$J and 1 x (f = 73.5 mm) objective was used to focus the laser beam inside the glass.](image-url)
rial containing some crystalline powder species [21,22].

In order to illustrate the influence of the focusing condition, we show the CCD images of the scattered laser light along the filaments after focusing with a 4× objective at different pulse energies (Fig. 5(a)). In parallel, the microscope photographs of the resulting refractive index modification regions are shown (Fig. 5(b)). A similar variation of the signal with input energy is noticed. That is, self-focusing of the pulse leads to the formation of filamentary structures similar to those observed for the case of 1× objective but with a relatively shorter filament length. At input energies higher than approximately 4 μJ a strong signal appeared near the geometrical focus which fully saturated the CCD camera. The image for 5 μJ was thus recorded with a lower gain of the camera. The damage zone with an irregular void-like morphology is shown in Fig. 5(b) at an input energy of 5 μJ. It can be distinguished from the region of uniform refractive index modifications (smooth track) when lower laser energy is used.

CCD images are shown in Fig. 6(a) corresponding to the structural damage resulting from focusing with a 10× objective. In this case, a considerably shorter filament with a typical length of ~20 μm is observed between the self and geometrical foci up to 1.0 μJ. For higher pulse energies a strong damage occurred right before the geometrical focus whose laser scattered signal extended over the region between the self and geometrical foci. Fig. 6(b) shows the evolution of the scattered laser light intensity from the damage zone when the number of laser shots at 1.3 μJ varies. The dependence of the spatial distribution of laser intensity and plasma density along the filament and around the geometrical focus on focusing condition and pulse energy was investigated in water using numerical simulations [26]. These results indicate that the on-axis plasma density would be higher near the geometrical focus as compared to that at the nonlinear self-focus. This is particularly valid for tight focusing conditions in which the avalanche ionization plays a major role in the formation of plasma around the geometrical focus. This could explain why in our experiments the structural damage often appears near the geometrical focus.

To investigate the influence of self-focusing and filamentation in the shape of modified refractive index regions in a very tight focusing condition, we used a 63× objective. We wrote various tracks at a depth of ~250 μm with input energies ranging from 50 nJ to 1 μJ, by translating the sample with a velocity of 100–1000 μm/s perpendicular to the beam propagation axis. The re-polished surfaces

Fig. 5. (a) Accumulated intensity distribution of the plasma photo-emission and scattered laser light at different pulse energies using 4× (f = 30.8 mm, NA: 0.075) objective and (b) microscope photographs of the corresponding regions of refractive index modification. At 5 μJ, the image was recorded with a reduced gain of the CCD to avoid further saturation of the camera. The number of laser shots was 10,000.

Fig. 6. (a) CCD images of the plasma photo-emission and scattered laser light using 10× (f = 16.9 mm, NA: 0.136) objective lens for 10,000 shots, (b) accumulated intensity variation of the scattered light from the damage zone versus laser shot at 1.3 μJ.
parallel to the laser beam axis were etched using a 50% hydro-fluoric (HF) acid solution for about 30 min. Afterwards, the cross sections of the tracks written under different pulse energies and for 500 μm/s scan speed, were inspected by a scanning electron microscope (SEM), as shown in Fig. 7(a). First note that for such scanning speed, each irradiated region in the glass received one or at most two laser pulses. For input energies lower than approximately 300 nJ, no visible structure was detected. This could be due to the very small size of the irradiated spots and/or very weak refractive index contrast (shallow etching) lower than the value detectable by the electron microscope. We were able to observe the irradiated spots for input energies approximately higher than 336 nJ. This energy is well above the pulse energy corresponding to the critical power for self-focusing in silica (~220 nJ). The laser beam propagates from the right to left side of the images. Transversely, the size of the zones where the refractive index appears to increase (corresponding to the dark area in the central part of each image) is approximately equal to the estimated beam spot size inside silica (~1 μm). However, along the beam propagation direction it appears that the size of the index modified region is longer than the estimated beam confocal parameter ~1.5–2 μm. Such

![Fig. 7. (a) SEM images of the polished etched cross sections of the tracks written perpendicularly with different input pulse energies at 500 μm/s using 63x objective (NA: 0.85, f = 3.1 mm) and (b) far-field pattern of the transmitted laser beam at 633 nm for the waveguide written at 432 nJ.](image-url)
extension results from the self-focusing and filament formation between the self-focus and geometrical focus. As shown, the length of filament increases with pulse energy [16,26]. The right and left hand sides of the dark central region in each of the images in Fig. 7(a) specify nearly the beginning of the self-focus and geometrical focus, respectively. We have also investigated the influence of scan speed, or in other words the number of laser pulses received per given focal volume, on the shape of refractive index modifications. No significant increase in the size of the refractive index modified zone was observed by decreasing the speed from 500 to 50 μm/s. However, a higher etching contrast was observed for the spots irradiated at lower scan speeds. This suggests that the magnitude of the refractive index change increases with the number of laser pulses. A recent study on the laser-induced breakdown in bulk transparent materials also reported similar results [27].

A step-like ring structure is clearly observed around the irradiated regions (Fig. 7(a)) To our knowledge such structures have not been reported previously in bulk silica glass upon irradiation with intense femtosecond laser pulses. At this moment, we have no clear evidence to state the origin of their formation. However, it is suggested that thermal effects and shock-waves associated with the formation of hot and dense plasma in the focal volume might play an important role in these observations.

For sufficiently high input energies, the region of refractive index modification evolves into two or more separated zones with lower etching depths (namely lower refractive index changes) beyond the geometrical focus in a rather regular fashion (Fig. 7(a)). Such behavior is known to be associated with refocusing of the femtosecond pulse [3]. Note that a non-uniform plasma density is also observed along the filaments in Fig. 2, suggesting multiple refocusing of the laser pulse before the geometrical focus. Refocusing of the pulse during filamentary propagation in air [3,17,29], in fused silica [30] and in liquid [28] were experimentally observed and also theoretically demonstrated by numerical simulations [5,31,32]. The formation of multiple foci after the geometrical focus can be described as follows. When the peak power is higher than the critical power at the geometrical focus, there would be a chance for the diverging pulse right after the geometrical focus, to refocus again one or more times as long as the pulse peak power remains sufficiently above the critical power $P_{cr}$. Note that the pulse peak power at the geometrical focus should be lower than the input pulse peak power due to energy loss in generating the plasma, plasma absorption, and group velocity dispersion (GVD) in the silica glass.

The fine modified refractive index structures resulted from multiple refocusing of the pulse after the geometrical focus (Fig. 7(a)) were not visualised clearly in the case of focusing with 1×, 4×, and 10× objectives. In fact, the dependence of pulse refocusing effect on the beam size and external focusing condition has been investigated [28–30]. In particular, this dependence was discussed in silica [30] based on a model in which the pulse propagation is analogous to the movement of a classical particle in a potential well [33]. Such analysis demonstrates that pulse refocusing could occur as long as the numerical aperture of the lens is less than a critical value [30]. This therefore suggests that multiple refocusing of the pulse similar to that observed for the 63× objective case should exist and be observed clearly (i.e., using SEM images) under focusing with 1–10× objectives as well. In fact, we were able to detect such fine structures owing to sufficiently higher sensitivity of the SEM technique compared to imaging by the CCD and optical microscope. Our observations appear to contradict in part [30] where no refocusing was reported in silica upon focusing tightly using an objective with the NA of 0.85. We believe that is due to different experimental conditions namely with respect to the pulse duration which was 120 fs, instead of 45 fs used in our work. Another important difference is that they had used a less sensitive imaging technique using a CCD camera for the detection of plasma luminescence from the focal volume.

In order to verify the tracks written perpendicularly using 63× objective for the possibility of guiding light, we coupled a He–Ne laser beam at 633 nm into their polished input facets using a 20× objective. For example, the far-field pattern
of the transmitted light from the track written at 432 nJ and 500 μm/s is shown in Fig. 7(b). It consists of a central bright slightly elliptical disk surrounded by near concentric elliptical rings. This suggests the formation of a waveguide with elliptical core in silica [34] which confirms that the refractive index is higher in the central irradiated zone compared to that in the unexposed surrounding area, as illustrated through the SEM images.

To further characterize these structures, the refractive index profiles of the cross sections of two of the waveguides were measured along two perpendicular axes of $x$ and $y$ (as defined in Fig. 7) using an optical waveguide analyzer (OWA-9500, EXFO Inc) (Fig. 8). Before installing the sample, the glass surface near the waveguides was polished down to 170 μm. This was necessary because only the waveguides within a depth of less than 100 μm could be tested by the instrument. The observed strong asymmetry between the index profiles confirms the influence of self-focusing and filamentation during pulse propagation. It appears that the index profile along the laser propagation direction ($y$ axis) is non-uniform and modulations with a period of $\sim 2–3$ μm are pronounced. We checked this point many times and observed similar index modulations measured for the other waveguides as well. We do not know yet the origin of their formation while further investigations are in progress. A second smaller wide peak located 12–13 μm away from the main modulated peak is also observed along the propagation direction.

![Graph](image_url)

Fig. 8. Refractive index profiles of the waveguide structures written perpendicularly using 63× objective lens along ($y$) and perpendicular ($x$) the laser propagation direction at (a) higher input energy and (b) lower input energy.
for the waveguide written at higher input energy (Fig. 8(a)). This is attributed to refocusing of the pulse beyond the geometrical focus which is in good agreement with the SEM images. We have also measured the index profiles of the waveguides written at lower input pulse energies. In agreement with the SEM pictures, they have revealed no secondary refractive index peak as shown in Fig. 8(b).

Waveguide losses depend on the writing parameters such as scan speed and pulse energy. For the waveguides written in the parallel configuration [8], a typical propagation loss of 2–3 dB/cm at 633 nm was measured in a 25 mm long sample, however it is believed that less than or on the order of 1 dB/cm is achievable. The loss measurement was performed by recording the intensity of the diffused light at 633 nm during propagation along the waveguide using the CCD camera. We have not yet measured losses for the elliptical waveguides written in the perpendicular geometry, but we expect higher values so that improvements in the optimization of the writing conditions are necessary in order to make these waveguide structures of practical use.

It should be mentioned that optical waveguides with a few micrometer core size were demonstrated in borosilicate glasses under a very tight focusing (NA = 1.4) in perpendicular writing geometry using a femtosecond laser oscillator at 25 MHz repetition rate [9]. The symmetric shape of the cross sections of the waveguides is attributed to the fact that the pulse energy used (several nJ) was much lower than the energy corresponding to the critical power for self-focusing in the glass. Note that for a sufficiently tight focusing condition one can reach the threshold intensity for optical breakdown at input powers below the critical power for self-focusing. Accordingly, we considered the possibility of writing such waveguides in silica at input powers less than the critical power but at scan speeds much slower than that used in [9]. This was to provide the same fluence at each exposed point as used in that work. Nevertheless, we could not realize this experiment because the estimated scan speed (~0.8 μm/s) required for satisfying the multi-shot irradiation condition given in [9] was many times smaller than the minimum translation speed of the motorized stage used in our work. However, it is believed that even after being able to write under such slow translation speeds, the created waveguide with a sub-micrometer core would not be practically useful to guide efficiently the visible and in particular telecommunication wavelength at 1.5 μm but could be useful for guiding near UV radiation. To further explain this point, note that the extent of thermal effects and their role in controlling (increasing) the size of the irradiated molten region in glass could be significantly different for the pulse repetition rates used. That is, unlike the case of irradiation with 1 kHz beam, the size of the refractive index modification zone in glass upon irradiation with 25 MHz laser beam strongly depends on the number of pulses received by the focal volume [9].

4. Conclusion

In conclusion, we studied the dependence of the femtosecond laser pulse-induced filamentation and structural damage in fused silica on pulse energy, focusing geometry, and the number of laser shots. The plasma photo-emission and laser scattering signals together with the modified refractive index structures in the glass were visualized using CCD, SEM and optical microscope images. For the pulse energies at which the photo-induced modifications in silica are observable, self-focusing and filamentation are inevitable and can significantly extend the size of the refractive index modification zone. In the case of a very tight focusing geometry using 63× objective lens and at input powers above the critical power for self-focusing in silica, elliptical structures with a non-uniform index profile were created by moving the sample perpendicular to the beam propagation axis, which could guide the laser light at 633 nm. We believe that the optimization of the writing conditions can significantly reduce the expected high propagation losses for such structures and make them useful practical devices.

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