Direct Observation of Femtosecond Laser Induced Modifications in the Bulk of Fused Silica by Phase Contrast Microscopy

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Employing optical phase-contrast microscopy we study the morphology of ultrashort pulsed laser-induced modifications in bulk fused silica and in other optical transparent materials for different conditions of irradiation. The influence of the input pulse energy, focusing depth, and number of pulses per site is investigated in order to establish optimal irradiation conditions for direct writing of waveguiding elements. The results obtained suggest that an increase of the refractive index is systematically accompanied by a region of lower index of refraction along the optical axis, presumably due to micro-explosion in the high energy-exposed regions, and the locations of this region is material dependent. Nevertheless, the creation of an hypothetical void in the interaction zone does not allow a complete explanation of the modifications observed and other mechanisms have to be invoked to explain the presence of regions with higher index of refraction. Different interaction regimes with respect to the input energy and to the number of shots per site can be established from these observations, emphasizing the role of nonlinear pulse propagation and plasma generation. To complement the experimental observations we simulate the propagation of an ultrashort laser pulse in bulk silica using the non-linear Schrödinger equation and determine the distribution of energy along the propagation axis. The results are in good agreement with the experimental observations and provides insights into the mechanisms controlling the interaction between ultrashort laser pulses and transparent materials.

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1. Introduction

Under intense femtosecond laser irradiation, optical properties of glassy materials can be permanently modified. This phenomenon with implications in emerging photonic applications is currently a hot area of research because the physics of the interaction still remains unclear. Fundamental aspects such as light propagation in transparent materials have to be further clarified despite of significant advances on this topic [1]. Besides the theoretical debate, very promising applications of technological interest have emerged. Several groups have reported photoinscription of optical components such as waveguides [2], diffractive elements [3], three-dimensional splitters [4] or data storage units [5] in bulk fused silica. Combined to etching techniques, the possibility to strongly focus ultrafast lasers beam into very small volumes makes them valuable tools for nanotechnologies, in particular for nanostructuring of transparent materials [6]. Focusing a short pulse of light tightly enables reaching intensities of the order of 10^{13} W/cm^2. Under those conditions non-linear mechanisms are triggered, notably multiphoton ionization and self-focusing, which is a natural response of the medium to light propagation. At high intensities, the index of refraction in fused silica follows the local intensity distribution. Hence, a Gaussian pulse induces a transient lens-like refractive index profile and the propagating beam tends to converge only due to its propagation. The laser beam can also ionize atoms on its way and create a free electron population. This electronic plasma brings a negative contribution to the index of refraction and therefore acts as a divergent lens. At a certain point focusing and defocusing effects may balance and give rise to a filamentary propagation. The step of ionization providing a defocusing electronic cloud is nonlinear too. The weak energy of the photons used in our experiments compared to the bandgap of the dielectric material implies that multiphotonic ionization processes are determinant. Depending on the focusing parameters and on the input energy, avalanche and tunneling can also play a significant role. Following this strong excitation, material-dependant relaxation processes take place and the optical properties of the material are locally modified. It appears that the

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induced permanent modification is an interplay between many mechanisms. Several groups [7,8] presented experimental evidences that femtosecond irradiation leads to defect creation on atomic scale. As a result of bond breaking, new stable atomic arrangements appear, such as E' centers or non-bridging-oxygen-holes centers (NBOHC) and one can establish a direct relation between the number of defects, their position into the bandgap, and the variation of the index of refraction [9]. Exposure to strong irradiation can also provoke a re-organization of the lattice with direct consequences on the optical properties. Raman spectroscopy measurements in irradiated fused silica [10] unambiguously show a local densification of the material caused by a rearrangement of the Si atoms. This local densification is due to the formation of 3- and 4-membered rings instead of the 5- and 6- membered rings initially present in the fused silica matrix, resulting in a decrease of the O-Si-O angle [11]. On a larger scale, several groups pointed out the presence of a permanent field of mechanical stress around the exposed region [12,13]. Nevertheless, for input energies of interest for writing optical components by local refractive index change, the material experiences significant modifications in its optical properties while keeping its transparency. In other words, under laser action, a phase object is created. Hence, traditional microscopy alone is of limited interest and only helps in determining cracked and strongly damaged zones that may appear when the mechanical damage threshold is reached. In the 30's, Zernike developed a microscopy technique allowing the observation of phase objects. This method, named phase-contrast microscopy, was successfully applied in the field of biology in different applications, e.g. cells observations or enzyme detection [14]. The purpose of this paper is to show that phase contrast microscopy can also be a natural tool for characterization of laser-induced phase objects in transparent media. In order to help understanding the underlying mechanisms responsible for material modification, a precise insight into the morphology of the laser modified areas is of first importance. Some techniques providing topological information about the effective refractive index map photoinduced under femtosecond irradiation such as cleaving [15], etching [6,16] or digital holography microscopy [17] have been demonstrated. Those techniques have the disadvantage to be destructive and require post processing of the sample. Contrary to those methods, in situ measuring is also possible [18]. Using for instance phase contrast microscopy, direct and instantaneous observation of refractive index profiles is easily achievable. Although quantitative phase contrast microscopy methods have been recently reported [19], phase contrast microscopy is here used as a contrast enhancement technique able to report modifications of the refractive index relative to the background value. No absolute values for the variation of the refractive index (Δn) are provided. The whole study is based on comparative observations, and only increases or decreases of the refractive index compared to the undisturbed bulk are taken as reliable information.

In the purpose of understanding the way the optical properties of the irradiated target are modified, optical observations are correlated with numerical simulation results. The nonlinear Schrödinger equation that describes pulse propagation in a nonlinear medium is solved in order to determine the energy deposition pattern after focusing and propagation in the bulk. In the numerical simulations, the concurrent phenomena of plasma defocusing and self-focusing are taken into account. The solutions obtained reveal the fluence distribution around the axis of propagation.

In this paper, phase contrast microscopy images of femtosecond laser induced bulk modifications in fused silica (a-SiO_2) and BK7 optical borosilicate crown glasses are reported. Several sets of pictures corresponding to different conditions of irradiation in a tight focusing regime are shown. First, the influence of the input energy, the importance of the incubation effects, and of spherical aberrations for single pulses are studied. Then, experimental observations about waveguiding structures and discussion about the optimal writing conditions are also presented. In parallel with the presentation of the experimental results, a synopsis for the global interaction is proposed, based on both phase contrast pictures and results of numerical simulation.

2. Experimental setup

We use an ultrafast laser system (Quantronix) delivering 170 fs pulses at a wavelength of 800 nm. Structures requiring a high number of shots per focal volume (e.g. waveguides) are written at a repetition rate of 1 kHz. Structures requiring a low number of shots are realized by scaling down the repetition rate of 100 Hz in combination with an electromechanical shutter (Uniblitz) which delivers a final repetition rate of 1 Hz. A set of optical density filters is used to select the input energy. Pulses are focused in the target samples using a 50× long working distance microscope objective with a numerical aperture (NA) of 0.45. SiO_2 and Bk7 parallelepipeds (3×20×10 mm) are investigated. Those samples are 4 sides polished, allowing writing and performing side microscopic observation simultaneously. A 3 axis positioning system including 2 motorized stages (Physik Instrumente) and one manual translation stage is used in order to drive the samples with a submicrometric precision. The phase contrast apparatus consists of an Olympus BX-41 microscope combined with a Jai CV-A1 camera providing images with a resolution up to one megapixel.

3. Results and discussion

When a femtosecond laser pulse is tightly focused in the bulk of a transparent material, structural modifications are photoinduced as a result of a strong electronic excitation. In a general case, the transmittance of the focal region after irradiation can be written as

$$T(x,y,z) = A(x,y,z)e^{-j\varphi(x,y,z)}, \quad (1)$$

where $A(x,y,z)$ represents the attenuation part and $e^{-j\varphi(x,y,z)}$ is the dephasing term. If the density of energy is low enough, the material keeps its transparency and the attenuation part can be neglected. In higher ranges of intensities, significant shockwaves are generated and the material network is not able to dissipate this mechanical energy without experiencing a permanent damage. Microcracks may occur and, as a consequence, the transmission of the irradiated region is affected. The modulus of the transmittance can be studied with a conventional optical microscope. The purpose here is to concentrate on the phase (φ) function, corresponding to a variation of the real part of the dielectric function. Phase contrast microscopy is well adapted to such a study. This special technique is able to provide directly a topological map of the different refractive index regions. As observable on Fig. 1, the surrounding medium, i.e. unaffected bulk of fused silica, appears mid gray. Any darker pixel indicates an increase of the index of refraction while a decrease is revealed by a lighter region.

We draw attention on a phase contrast picture showing

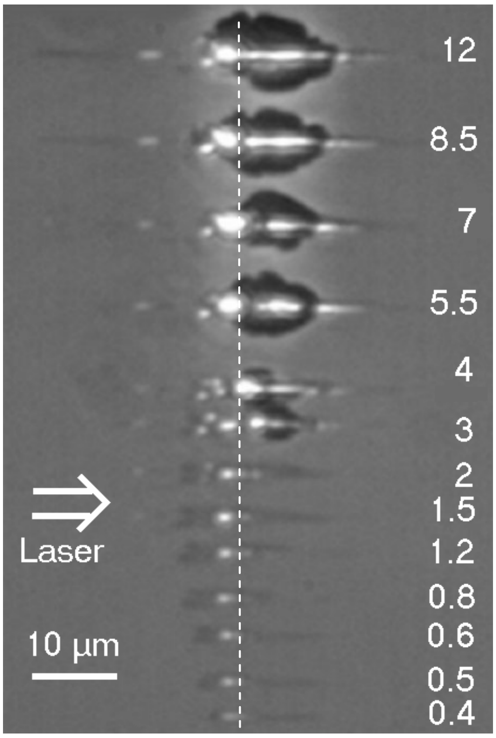

Fig. 1 Energy dependence of laser-induced traces for a single pulse interaction in bulk fused silica. Aberrations are minimized by focusing 200 µm into the bulk. On the right column, input energies in µJ are specified. The dotted line represents the estimated focal plane.

the morphology of the interaction results for different input energies in bulk fused silica (Fig. 1). In order to ensure that the depth of writing is the same for each trace, the perpendicularity between the sample and the writing beam has been carefully checked. All the single pulse traces have been written in a sequence, translating the sample perpendicular to the laser beam direction. Important observations arise from this picture. For the whole range of energy scanned, it comes out that for a NA=0.45 an increase of the refractive index (i.e. a dark region) is systematically accompanied by a region of lower refractive index. The higher refractive index zone corresponds roughly to the position of the geometrical focus. Two types of interaction clearly emerge: a soft mode, up to input energies of 2 µJ and a strong mode, taking place for input energies from 5.5 µJ. This latter mode is characterized by a filament followed by a black stain region. A transition regime is visible for input energies of 3 and 4 microjoules. Those tracks still exhibit a conical shape very similar to the low energy traces but one can distinguish as well the onset of the black stain typical of the strong interaction mode.

In the soft interaction regime (i.e. between 0.4 and 2 µJ), one can see a white volume surrounded and followed by a region of higher refractive index. In order to understand the interaction mechanisms, we developed a simulation code for ultrafast pulse propagation similar to the one employed by A. Couairon et al. [20], taking into account both self-focusing and plasma defocusing on the generated electron population. Since the formalism was presented in great detail [20], we will not insist here on the calculation steps but concentrate on the numerical results. The numerical simulations provide the temporal envelope of the pulse. This gives access to the fluence distribution deposited in and around the focal region along the propagation axis. Results are plotted on Fig. 2. The figure indicates that most of the energy is deposited before the geometrical focus, where the area of low index of refraction appears. This region may presumably be a microcavity, showing a smaller density than the surrounding material. The concept of a microexplosion has been suggested by Glezer et al [21] from atomic force microscope measurements. The presence of a void visible in scanning electron microscopy has been reported for higher numerical apertures by Schaffer et al. [22]. Nevertheless, the term of microexplosion may sound a bit excessive as explosive forces still have to be clearly identified. Alternatively, we believe that, as this white region receives the highest fluence, a transition towards a lower density state occurs and the microcylinder of lower refractive index may only be the result of thermal expansion in the heated material. As only a fraction of the energy experiences this effect, the deposited energy remains limited, the corresponding temperature increases

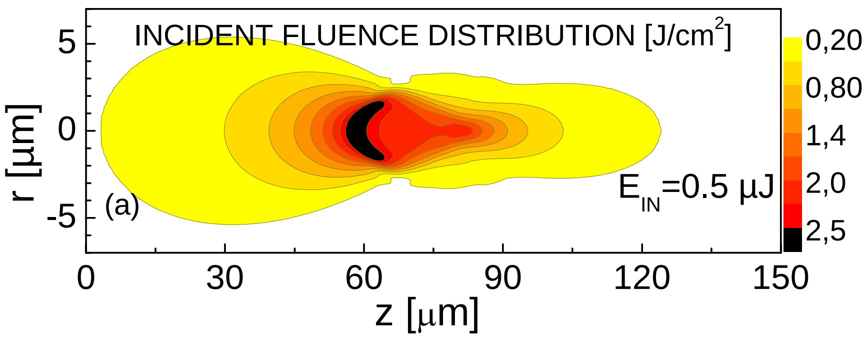

Fig. 2 Numerical simulation of the fluence distribution around the propagation axis z for a 500 nJ single pulse. The geometrical focus is located in the center of the window at z=75 µm.

moderately within the affected region, and the resulting interaction happens in a soft mode. Tracks corresponding to the strong regime of interaction (between 5.5 and 12 µJ) have a different morphology. The threshold for a transition to low density phase, corresponding to white areas, is reached on a large volume, over a length of 10 µm. Presumably, much more energy is being deposited and liquid phase is reached in the regions of maximum exposure. For the same reasons as before, a soft black zone extends far away from the initial focal point. A different black region comes out in this regime, appearing like a deep black stain around the white region and corresponding to a zone of high index of refraction. We believe that this feature is not simply due to electronic excitation, but also to a significant field of compression because of an important thermal expansion in the white region generating shock waves into the material. Interestingly, on the left side of those high energy tracks a filament followed by a white dot appears. This filament exhibits a surprising similitude with the lowest energy track (0.4 µJ) at the notable difference that it has been written from the right to the left. This suggests that a sufficient amount of energy is reflected from the tail of the pulse on a electron hole plasma mirror and exceeds the material threshold during its back propagation. It is worth noticing that the modification threshold may be substantially lower, due to the pre irradiation of this region by the transmitted part of the incoming pulse.

Figure 3 illustrates the effect of spherical aberrations. This picture corresponds to a non compensated propagation of roughly 500 µm in fused silica. As paraxial and non

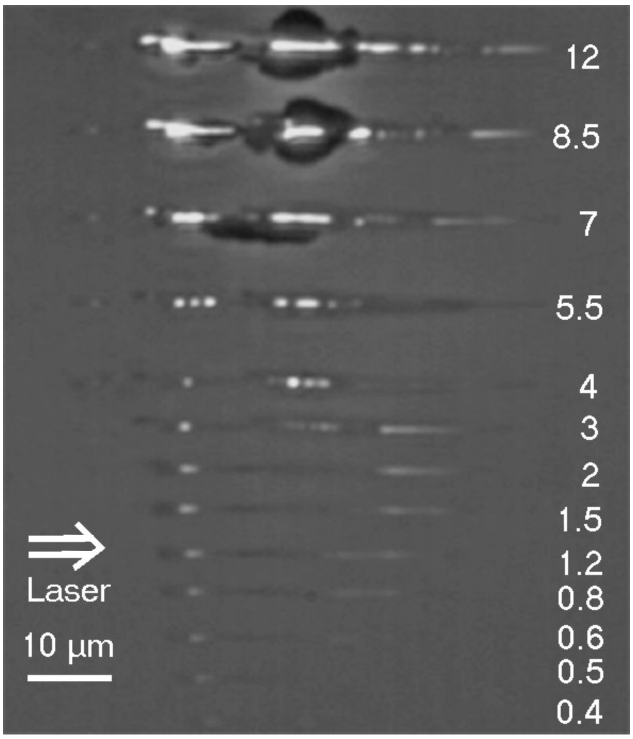

Fig. 3 Energy dependence of laser-induced traces for a single pulse interaction in bulk fused silica. Aberrations are enhanced by focusing 0.5 mm into the bulk. On the right column, input energies in µJ are specified.

paraxial rays have different optical paths due to refraction at the air-dielectric interface, the different spatial components of the beam focus in different planes perpendicularly to the axis of propagation. The interaction region appears more elongated and can extend over 50 µm for high energies. The axial energy density being weaker, energetic thresholds are different. The contrast is lower than before, showing that the Δn induced is not as large as in the case of a compensated propagation. The same features as we already mentioned can be identified. In the initial soft interaction regime, between 0.4 and 4 µJ, the white cylinder of lower refractive index moves clearly towards the focusing objective as the energy increases. This is an explicit effect of self focusing. Those observations bring additional comments about waveguide writing conditions. Two methods are commonly used to write guiding structures. One can either write the structure longitudinally [23], i.e. the sample moves parallel to the laser beam, or transversally, in a configuration where the sample moves perpendicularly with respect to the writing beam. In the first case, the main advantage is that one can obtain a symmetric waveguide, compared to the transversal configuration where the section of the guiding structure is elliptical. Nevertheless, the longitudinal setup has strong limitations. First, the maximal length of the guide is limited. Then, a comparison of Figs. 1 and 3 shows that the optical properties of the waveguide may change along the propagation. Obviously, it makes the interaction scheme more complicated to understand and to optimize. Therefore, the discussion will stay with transversal writing. We already pointed out that transversal writing induces an elliptical profile. This can be overcome by performing some beam spatial shaping before writing [24]. Practically, the guiding structure is written by translating the sample continuously and slowly, perpendicularly with respect to the beam, so that each region of the guide is receiving several pulses.

Figure 4 shows a phase contrast picture of laser induced modifications in fused silica where each track has been irradiated 1000 times. The difference with the previous picture is striking, putting forward the importance of the incubation phenomena in fused silica. The threshold is here substantially lower than in the single pulse experiment. It means that even if no modifications could be clearly detected below 0.4 µJ in the single pulse mode, the material has been already structurally modified and its optical properties altered. The repetition rate being 1 kHz, heat accumulation effects from pulse to pulse can be excluded. Some tracks suggest that after 1000 shots, the saturation is not reached yet, as the black tail still exhibits some white bubble-like features. This is notably the case for the high energies traces. Compared to the single shot irradiation, the tracks are more extended. At 1.2 µJ for instance, the track length is at least a factor of 2 larger. The induced Δn is also more intense as black regions appear darker than in the single shot set of measurements. Standard microscope observations reveal that the white region is also an amplitude object where optical transparency has been lowered. Therefore, for guiding structures writing, one should try to minimize the size of this white area. In our experimental conditions, this white microcylinder does not totally disappear. Nevertheless, the white region is less and less bright when the input energy decreases. Hence, we

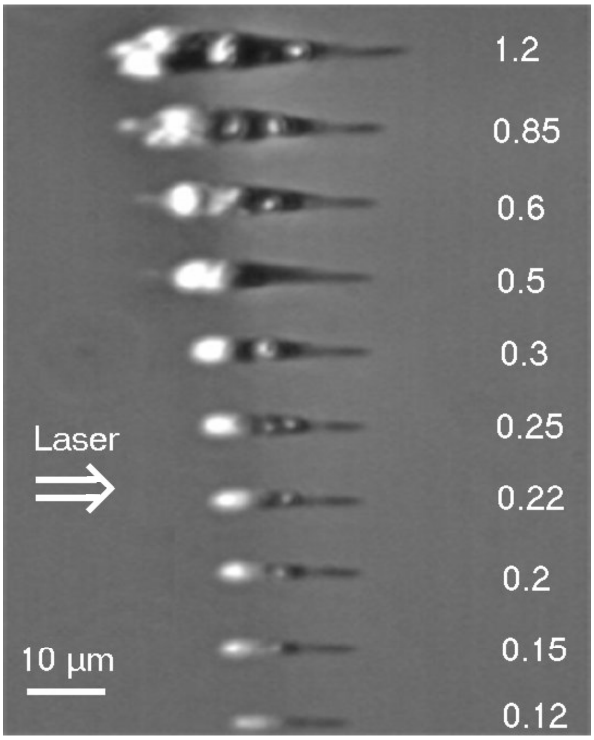

Fig. 4 Energy dependence for a 1000 shots interaction in bulk fused silica. Aberrations are minimized by focusing 200 µm into the bulk. On the right column, input energies in microjoules are specified

assume that it could be significantly reduced by irradiating the sample with lower energy, using an higher numerical aperture. From this picture we can also deduce that a high number of pulses at a weak energy is probably the most suitable for waveguide writing.

Figure 5 is a picture of a waveguide written transversally with the same numerical aperture as before (NA=0.45), low energy (about 120 nJ), and approximately 1000 shots per site, as the scanning speed was set at 1µm/s. Those experimental conditions are similar to those reported by Nolte et al. [4], with the difference that a higher number of pulse per site was employed. The report has shown that such structures were indeed guiding light. In phase contrast microscopy, a black stripe appears clearly. We assume that this corresponds to the guiding part, of stronger refractive index. This is confirmed by observations in Bk7, known as

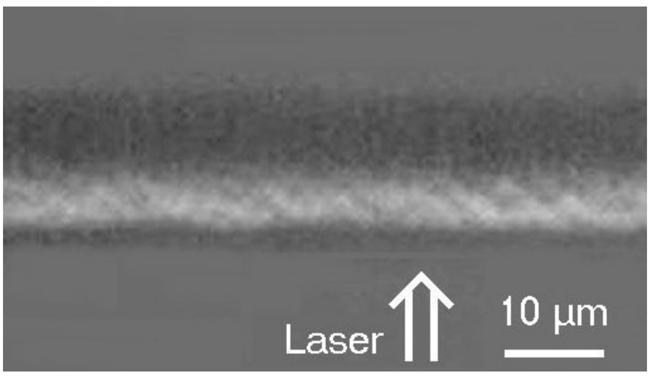

Fig. 5 Phase contrast picture of a waveguide written in fused silica. The focusing objective is aberration free for this 170 µm writing depth. The scanning velocity is 1µm/s.

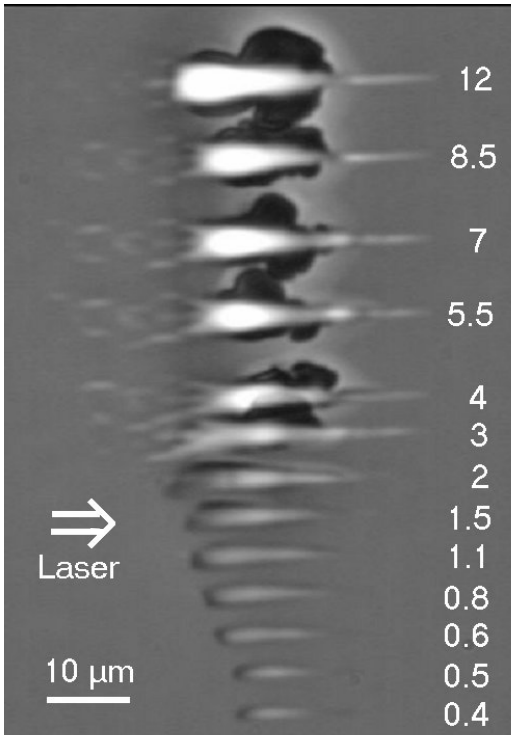

Fig. 6 Energy dependence for a single pulse interaction in Bk7. Aberrations are minimized by focusing 200 µm into the bulk. On the right column, input energies in microjoules are specified.

a less suitable candidate for light guiding [25], where no black stripe can be seen in similar experimental conditions.

Further investigations in Bk7 optical glass are presented in Figs. 6 and 7. One can still observe the onset of a strong regime at high energies, but this time the transition threshold between the two regimes is lower. This is presumably due to the lower bandgap of Bk7 compared to a-SiO_2. Nevertheless, there is a striking similitude between black stains observed in a-SiO_2 and in Bk7. In phase contrast microscopy, tracks appear in white colors, meaning that laser exposure induces a negative index contribution. This is in good agreement with the negative Δn recently reported by Bhardwaj et al. [25] in such glasses. The material in the vicinity of the head of the track has experienced an important temperature increase and has probably been softened. In the focal region, heating is even more important. It may trigger a thermal expansion associated with a phase transition. The expansion of the overheated material being facilitated towards the exterior a small black zone appears at the front of the trace. The origin of the complex structure composed of small white dots appearing at the back of the tracks (Fig. 6) is still unclear. Obviously, those dots indicate that a modification threshold has been reached. The difficulty is in understanding why the energy repartition is not homogenous in this region. It could possibly come from a reflection of the tail of the pulse on the plasma induced by the head of the pulse followed by an interference phenomena. A surprising feature of Bk7 is the very weak influence of the number of pulse on the morphology of the induced traces. Figure 7 shows the appearance of the laser made structures as a function of the number of shots at a

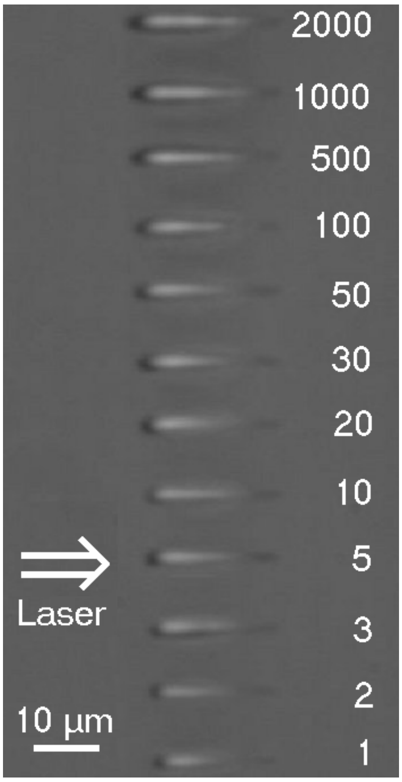

Fig. 7 Aspect of fs laser-induced traces in Bk7 for different number of shots per site. Aberrations are minimized by focusing 200 µm into the bulk. On the right column, the number of shot per site is specified.

constant energy (400 nJ). The size of the interaction zone barely changes and the contrast difference between a single shot and a multishot track is not significant enough to conclude to a progressive shot to shot modification of the material similar to what was observed in fused silica.

4. Conclusion

In conclusion, we have demonstrated the advantages of phase contrast microscopy to investigate laser induced modifications in glassy materials. We took the examples of fused silica and BK7 borosilicate crown glass. The response of those two materials to femtosecond irradiation is very different. In a-SiO_2 we report an increase of the refractive index accompanied by a low index microcavity. In Bk7, laser exposure leads to a decrease of the index of refraction, making us consider Bk7 as an inappropriate candidate for writing light guiding elements. The sensitivity of this method enables the study of the influence of fundamental parameters such as input energy or number of shots per site on the morphology of the interaction. Bk7 has been found to be insensitive to incubation while the number of shots seems to be a crucial factor in a-SiO_2 processing. Moreover, a comparison of the phase-contrast microscopy pictures with numerical simulation results helped us to build a plausible scenario for the interaction of ultrashort pulses with bulk a-SiO_2.

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