Femtosecond laser modification of fused silica: the effect of writing polarization on Si-O ring structure

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Abstract: A femtosecond laser with a 1 kHz repetition rate and two different polarization states was used to fabricate low-loss waveguides in fused silica. Investigations of chemically-mechanically polished waveguide regions using near-field scanning optical microscopy revealed the presence of modifications outside the glass regions directly exposed to a circularly polarized writing laser. These waveguides also exhibited refractive index contrast up to twice as large as that of waveguides written with linearly polarized radiation. The observed differences in refractive index were shown by Raman spectroscopy to correlate to an increased concentration of 3-member silicon-oxygen ring structures. We propose that the observed differences in material properties are due to the polarization dependence of photo-ionization rates in fused silica.

OCIS codes: (320.2250) Femtosecond Phenomena; (350.3390) Laser Materials Processing; (230.7370) Waveguides; (160.2750) Glass and other amorphous materials; (190.4180) Multiphoton processes

References and links

1. K. M. Davis, K. Miura, N. Sugimoto, and K. Hirao, "Writing Waveguides in Glass with a Femtosecond Laser," Opt. Lett. **21,** 1729-1731 (1996).
2. D. Homoelle, S. Wielandy, A. L. Gaeta, N. F. Borrelli, and C. Smith, "Infrared Photosensitivity in silica glasses exposed to femtosecond laser pulses," Opt. Lett. **24,** 1311-1313 (1999).
3. C. B. Schaffer, A. Brodeur, J. F. Garcia, and E. Mazur, "Micromachining bulk glass by use of femtosecond laser pulses with nanojoule energy," Opt. Lett. **26,** 93-95 (2001).
4. A. M. Streltsov and N. F. Borrelli, "Study of femtosecond-laser-written waveguides in glasses," J. Opt. Soc. Am. B **19,** 2496-2504 (2002).
5. M. Will, S. Nolte, B. N. Chichkov, and A. Tuennermann, "Optical properties of waveguides fabricated in fused silica by femtosecond laser pulses," Appl. Opt. **41,** 4360-4364 (2002).
6. G. D. Marshall, M. Ams, and M. J. Withford, "Direct laser written waveguide-Bragg gratings in bulk fused silica," Opt. Lett. **31,** 2690-2691 (2006).
7. J. W. Chan, T. R. Huser, S. H. Risbud, J. S. Hayden, and D. M. Krol, "Waveguide fabrication in phosphate glasses using femtosecond laser pulses," Appl. Phys. Lett. 82, 2371-2373 (2003).
8. M. Ams, G. D. Marshall, D. Spence, and M. J. Withford, "Slit beam shaping method for femtosecond laser direct-write fabrication of symmetric waveguides in bulk glasses," Opt. Express **13,** 5676-5681 (2005).
9. A. Zoubir, M. Richardson, C. Rivero, A. Schulte, C. Lopez, K. Richardson, N. Ho, and R. Vallee, "Direct femtosecond laser writing of waveguides in As2S3 thin films," Opt. Lett. **29,** 748-750 (2004).
10. R. R. Thomson, S. Campell, I. J. Blewett, A. K. Kar, D. T. Reid, S. Shen, and A. Jha, "Active waveguide fabrication in erbium-doped oxyfluoride silicate glass using femtosecond pulses," Appl. Phys. Lett. **87,** 121102 (2005).
11. G. D. Marshall, P. Dekker, M. Ams, J. A. Piper, and M. J. Withford, "Directly written monolithic waveguide laser incorporating a distributed feedback waveguide-Bragg grating," Opt. Lett. **33,** 956-958 (2008).
12. R. Osellame, S. Taccheo, G. Cerullo, M. Marangoni, D. Polli, R. Ramponi, P. Laporta, and S. De Silvestri, "Optical gain in Er-Yb doped waveguides fabricated by femtosecond laser pulses," Electron. Lett. **38,** 964-965 (2002).

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13. L. Gui, B. Xu, and T. C. Chong, "Microstructure in lithium niobate by use of focused femtosecond laser pulses," IEEE Photon. Technol. Lett. **16,** 1337-1379 (2004).
14. J. Thomas, M. Heinrich, J. Burghoff, S. Nolte, A. Ancona, and A. Tuennermann, "Femtosecond laser-written quasi-phase-matched waveguides in lithium niobate," Appl. Phys. Lett. **91,** 151108 (2007).
15. A. H. Nejadmalayeri, P. R. Herman, J. Burghoff, M. Will, S. Nolte, and A. Tuennermann, "Inscription of optical waveguides in crystalline silicon by mid-infrared femtosecond laser pulses," Opt. Lett. **30,** 964-966 (2005).
16. Y. Shimotsuma, P. G. Kazansky, J. R. Qiu, and K. Hirao, "Self-organized nanogratings in glass irradiated by ultrashort light pulses," Phys. Rev. Lett. **91,** 247405 (2003).
17. C. Hnatovsky, R. S. Taylor, E. Simova, V. R. Bhardwaj, D. M. Rayner, and P. B. Corkum, "Polarization-selective etching in femtosecond laser-assisted microfluidic channel fabrication in fused silica," Opt. Lett. **30,** 1867-1869 (2005).
18. M. Ams, G. D. Marshall, and M. J. Withford, "Study of the influence of femtosecond laser polarisation on direct writing of waveguides," Opt. Express **14,** 13158-13163 (2006).
19. A. H. Nejadmalayeri and P. R. Herman, "Ultrafast laser waveguide writing: lithium niobate and the role of circular polarization and picosecond pulse width," Opt. Lett. **31,** 2987-2989 (2006).
20. J. W. Chan, T. R. Huser, S. H. Risbud, and D. M. Krol, "Structural changes in fused silica after exposure to focused femtosecond laser pulses," Opt. Lett. **26,** 1726-1728 (2001).
21. J. W. Chan, T. R. Huser, S. H. Risbud, and D. M. Krol, "Modification of the fused silica glass network associated with waveguide fabrication using femtosecond laser pulses," Appl. Phys. A **76,** 367-372 (2003).
22. A. Pasquarello and R. Car, "Identification of Raman Defect Lines as Signatures of Ring Structures in Vitreous Silica," Phys. Rev. Lett. **80,** 5145-5147 (1998).
23. A. E. Geissberger and F. L. Galeener, "Raman studies of vitreous SiO2 versus fictive temperature," Phys. Rev. B **28,** 3266-3271 (1983).
24. M. Okuno, B. Reynard, Y. Shimada, Y. Syono, and C. Willaime, "A Raman spectroscopic study of shock-wave densification of vitreous silica," Physics and Chemistry of Minerals **26,** 304-311 (1999).
25. V. V. Temnov, K. Sokolowski-Tinten, P. Zhou, A. El-Khamhawy, and D. von der Linde, "Multiphoton Ionization in Dielectrics: Comparison of Circular and Linear Polarization," Phys. Rev. Lett. **97,** 237403 (2006).
26. D. Liu, Y. Li, M. Liu, H. Yang and Q. Gong, "The polarization-dependence of femtosecond damage threshold inside fused silica," Appl. Phys. B **91**, 597-599 (2008).
27. S. M. Eaton, H. Zhang, M. L. Ng, J. Li, W. J. Chen, S. Ho, and P. R. Herman, "Transition from thermal diffusion to heat accumulation in high repetition rate femtosecond laser writing of buried optical waveguides.," Opt. Express **16,** 9443-9458 (2008).

1. Introduction

Since the first demonstration of femtosecond (fs)-laser induced refractive index modification by Davis *et al*. [1], optical waveguides have been fabricated in a range of media including fused silica [2-6], phosphate glass [7, 8], borosilicate glass [2,4], chalcogenide glass [9], active doped glasses [10-12], lithium niobate [13, 14], and silicon [15]. Fused silica is one of the most widely used substrates for fs-written devices because it permits the fabrication of low-loss waveguides, is transparent across a wide wavelength range, exhibits high mechanical, thermal and chemical stability, and is relatively inexpensive.

Many parametric studies have been performed on writing waveguides in fused silica, but it is only recently that attention has been given to the role that the polarization of the writing beam has on the waveguide properties. The influence of writing polarization on the modification was first observed in the form of nano-structure within laser modified regions, the morphology of which was observed to be strongly polarization dependent [16, 17]. Later, Ams *et al*. demonstrated that waveguides written with a circularly polarized beam exhibited significantly lower losses than waveguides written with a linearly polarized beam under the same conditions [18]. The polarization of the writing beam has also been reported to influence the optical properties of waveguides written in lithium niobate [19].

While a number of groups, notably Chan *et al* have performed spectroscopic studies of fs-modified fused silica [20, 21], the influence of writing polarization on the modification of the silicon-oxygen network has not been investigated until now. In this paper, we demonstrate that waveguides written with circularly polarized radiation in fused silica exhibit increased densification and larger refractive index contrasts than waveguides written with linearly polarized radiation when the peak irradiance of the writing beam exceeds 42 TW/cm^2, and

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show for the first time how the effect of writing polarization can be understood by looking at the polarization-dependence of ionization mechanisms that drive the modification process.

2. Methodology

Optical waveguides were fabricated using a regeneratively amplified Ti:sapphire laser with pulses of < 120 fs at a wavelength of 800 nm and a repetition rate of 1 kHz. Laser pulses were passed through a 500 μm wide slit, ensuring the waveguide cross sections are circularly symmetric [8]. The laser pulses were focused inside the fused silica substrate using a 20× objective lens (Olympus UMPlanFL, NA 0.46). The beam at focus formed an ellipse with an $1/e^2$ beam diameter of 1 ± 0.1 μm parallel to the waveguide axis and 10 ± 0.5 μm perpendicular to the waveguide axis. The $1/e^2$ Rayleigh range was also 10 ± 0.5 μm.

Laser polarization was controlled using a Berek compensator (New focus model 5540) placed just before the slit. The substrate was translated using a computer-controlled XYZ stage at a speed of 25 μm/s. The fused silica sample was chemically-mechanically polished at each end after exposure with a Logitech PM4 lapping and polishing machine using Ultra-Sol 500s (Eminess technologies) colloidal silica polishing compound.

Three groups of waveguides were written, one group was written with circularly polarized radiation, one with linear polarization parallel to the waveguide axis and one with linear polarization perpendicular to the waveguide axis. Each group consisted of three waveguides written at pulse energies of 1.5 μJ, 3.0 μJ and 4.5 μJ, pulse energies which are known to yield good quality waveguides for our writing geometry. Pulse energies larger than 4.5 μJ exceed the damage threshold for fused silica and form relatively high loss waveguides. The optical properties of waveguides written with different linear polarizations were found to be almost identical within experimental uncertainties, and so detailed comparisons between linear polarizations were not pursued.

Refractive index profiles were collected for each waveguide (at a wavelength of 633 nm) using a refractive index profilometer from Rinck Electronik.

Raman spectra were collected using a Renishaw Ramascope. Samples were illuminated using a 442 nm HeCd laser with a ~ 1 μm spot size focused using an OFR 40×-NUV objective.

Optical near-field and shear-force images were obtained simultaneously using a near-field scanning optical microscope (NTegra Solaris) from NT-MDT. Waveguides were illuminated using an 8 mW HeNe laser source operating at 633 nm coupled in to a single-mode fiber pigtailed to the waveguide input. The optical signal was detected using a photo-multiplier tube.

3. Results

3.1 Near-field scanning optical microscopy

Near-field Scanning Optical Microscopy (NSOM) enabled the simultaneous measurement of both the near-field optical output and the corresponding surface profile at the output facet of each waveguide. Examining the surface topology of the output facet revealed insights into the sensitivity of the glass to the chemical-mechanical polishing process used to prepare the end facet. In particular, the region of glass directly irradiated by the femtosecond laser writing beam exhibited increased sensitivity compared to the bulk glass, resulting in faster polish rates that present as depressions under the NSOM when monitoring the shear-force signal, as shown in Fig. 1. The depressions are smooth and to the 100 nm resolution of the NSOM, are absent of any microstructure, which is in contrast to studies done using hydrofluoric acid (HF) to etch fused silica [16, 17]. In addition, the regions of glass in the lateral plane and outside of the irradiated region exhibited reduced etch rates compared to the bulk glass, resulting in the presence of slight surface elevations, appearing to the left and right of the central waveguide region shown in Fig. 1.

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The polarization of the writing beam was observed to change the polish rate of the fused silica relative to the unexposed glass. For example, for pulse energies of 3.0 μJ and 4.5 μJ a variation in the polish rate was observed for glass regions irradiated by circularly polarized radiation, but no variation in the polish rate was observed for glass regions irradiated by linearly polarized radiation. At relatively low writing pulse energies of 1.5 μJ there was no discernible change in the polish rate for either the linear or the circular polarizations.

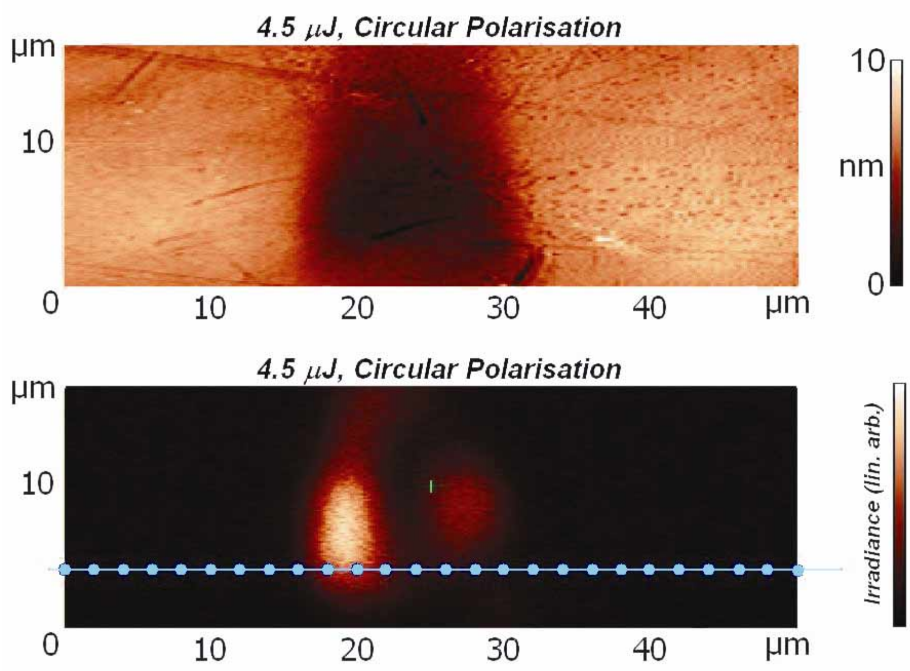

Fig. 1. Shear-force (above) image of the surface topology of the output facet of a waveguide written with circularly polarized radiation and 4.5 μJ pulse energy. The corresponding optical near-field (below) confirms the presence of waveguiding, the non-Gaussian output profile is due to the waveguide being highly multi-mode at 633 nm. The writing laser was directed along the Y-axis coming from the top of the image. Spots indicate points where Raman spectra were collected.

The depths of the depressions were measured to be 1.75 ± 0.25 nm for the 3.0 μJ case and 4.00 ± 0.25 nm for the 4.5 μJ case. The heights of the elevations flanking the central depression were measured to be 1.25 ± 0.25 nm for the 3.0 μJ case and 2.00 ± 0.25 nm for the 4.5 μJ case.

The variation in the chemical-mechanical properties alludes to a change in the atomic structure in the fs-laser exposed regions of glass. This change in atomic structure was further investigated using Raman spectroscopy.

The elevated regions flanking the central depressions are noteworthy as they hint at modification of the fused silica outside the region directly exposed to the writing beam. No change in refractive index or Raman spectra, relative to the surrounding unmodified regions of glass, was detected in these regions. The nature of these modifications is unknown, however it is possible that these regions are a result of stress induced by the waveguide on the surrounding regions of glass.

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3.2 Raman microscopy

To characterise the glass bonding structure in the waveguides, a Raman spectrum was obtained every 2 μm along a cross section across each waveguide perpendicular to the direction of the writing laser, as shown in Fig. 1.

A typical Raman spectrum obtained when examining fused silica is shown below in Fig. 2. The so-called “defect” lines, labeled D_1 and D_2 arise due to the presence of 4-member and 3-member silicon-oxygen ring structures respectively [22]. To allow comparison of the data, the intensity of the D_2 peak was measured relative to the intensity of the ω_3 peak (which arises due to vibrations in the random network of Si-O bonds), because the intensity of the ω_3 peak with respect to the overall Raman intensity is known to be relatively constant [23].

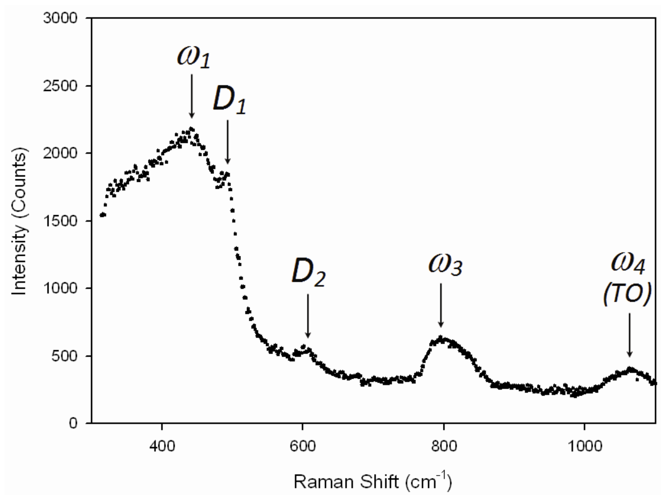

Fig. 2. A typical Raman spectrum of fused silica, showing the ω_1, D_1, D_2, ω_3 and ω_4 (TO) peaks at 440 cm^{-1}, 490 cm^{-1}, 605 cm^{-1}, 800 cm^{-1} and 1060 cm^{-1} respectively.

The waveguides written with 4.5 μJ pulse energy were characterized by measuring the ratios between the intensities of the D_2 and ω_3 peaks as a function of position along the cross-section of the waveguides. Waveguides written with both circularly and linearly polarized radiation were characterized in this manner, the data are shown in Fig. 3.

The intensity of the D_2 peak is observed to increase relative to the intensity of the ω_3 peak for both waveguides, indicating that the concentration of 3-member silicon-oxygen ring structures has increased in these regions in comparison to the unmodified glass, a characteristic associated with increased densification of the fused silica [24] and in agreement with what has been previously reported by Chan *et al.* [20, 21]. In this instance however, it can be seen that the waveguide written with circularly polarized radiation has a significantly higher population of 3-member Silicon-Oxygen ring structures than the waveguide written with linearly polarized radiation, and is therefore denser. This increased density was found to directly correspond to regions of increased refractive index, reported in the following section.

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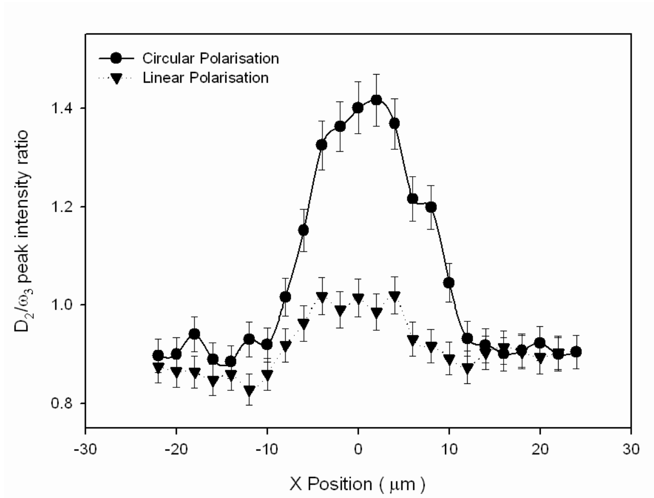

Fig. 3. Ratio of the D_2 and ω_3 peak intensities as a function of waveguide position for waveguides written with 4.5 μJ, circularly polarized and 4.5 μJ, linearly polarized radiation. The waveguide is centered at X = 0 in both instances. Un-modified glass possessed a D_2/ω_3 ratio of 0.9 ± 0.02.

3.3 Refractive index profilometry

The refractive index change induced by the waveguide writing process was strongly dependent on both the pulse energy and the polarization of the femtosecond laser beam. Table 1 summarizes the measured maximum refractive index changes for the investigated range of waveguide writing conditions.

Table 1. Laser-induced peak refractive index change for each set of writing conditions. The uncertainty in all cases is $\pm 0.1 \times 10^{-3}$.

Writing Pulse Energy	Δn - Linear Polarization	Δn - Circular Polarization
1.5 μJ	0.8×10^{-3}	1.25×10^{-3}
3.0 μJ	1.0×10^{-3}	2.3×10^{-3}
4.5 μJ	1.8×10^{-3}	2.4×10^{-3}

Waveguides written with circularly polarized radiation exhibited a greater refractive index change than those written using linearly polarized radiation for the same pulse energy. The greatest difference was exhibited for waveguides written with a 3.0 μJ pulse energy, where the refractive index contrast was more than a factor of 2 larger for the waveguides written with circularly polarized radiation relative to those written with linearly polarized radiation.

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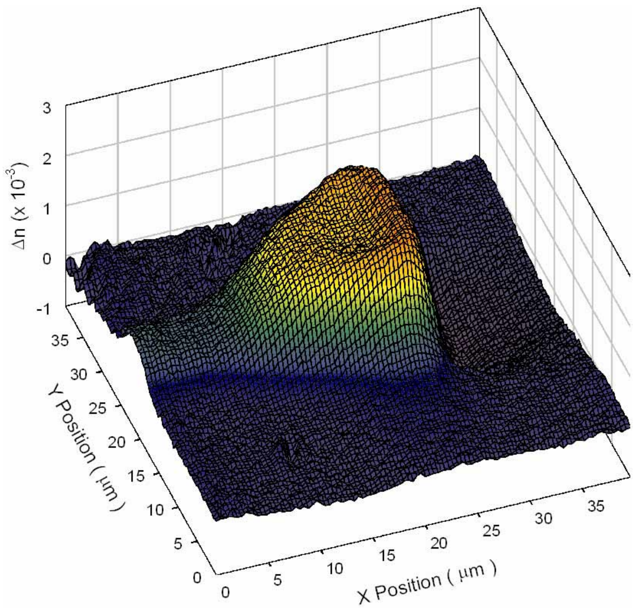

Fig. 4. Refractive index profile for a waveguide written with a pulse energy of 4.5 μJ and circularly polarized radiation. The writing laser propagates in the positive X direction.

The refractive index profile of the waveguide written with circularly polarized radiation and 4.5 μJ pulse energy is shown in Fig. 4. The diameter of both waveguides studied in the previous section is measured to be 17.6 ± 0.4 μm for the waveguide written with circularly polarized radiation and 16.4 ± 0.4 μm for the waveguide written with linearly polarized light in the Y-direction (direction perpendicular to the writing laser). These values correlate to the diameter of the modified region observed under the Raman microscope (18 ± 2 μm and 15 ± 2 μm for waveguides written with circularly polarized and linearly polarized radiation respectively).

Originally the lower transmission losses observed for waveguides written with circularly polarized light by Ams *et al.* were attributed to the morphology of the waveguide. We now attribute lower transmission loss to the higher refractive index exhibited by these waveguides, as losses due to waveguide inhomogeneities such as random curvature and diameter variations are known to be less for higher refractive index contrasts.

4. Discussion

The observed difference in properties between waveguides written with circularly polarized radiation and linearly polarized radiation is perhaps surprising given that fused silica is isotropic. To understand the effect of laser polarization on the waveguide writing process, it is important to consider the underlying physical mechanisms that result in modification of fused silica.

Modification of fused silica is driven by the absorption of optical energy from the writing beam, initially due to photo-ionization (which includes multi-photon ionization and tunneling ionization processes), then as the free-carriers acquire enough energy, impact (or avalanche) ionization. The degree to which fused silica is modified is therefore expected to depend on the irradiance of the writing beam and polarization-dependent free-carrier generation rates.

The polarization-dependence of photo-ionization was investigated experimentally by Temnov *et al*, who showed that for comparatively low irradiances, (< 15 TW/cm^2) the photo-ionization rates were approximately 3-4 times greater in fused silica for linearly polarized radiation than for circularly polarized radiation and 6 times greater in sapphire [25]. For

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higher irradiances (30-35 TW/cm^2), Temnov *et al* reported that the photo-ionization rates for each polarization became comparable, due to shift away from a purely multi-photon ionization regime into a regime with significant contributions from both multi-photon and tunneling ionization mechanisms. For irradiances greater than 35 TW/cm^2 circularly polarized radiation yielded up to 30% higher photo-ionization rates in sapphire than linearly polarized radiation, however, Temnov *et al.* did not explore whether circularly polarized radiation yielded higher photo-ionization rates in fused silica above 35 TW/cm^2 due to the onset of surface breakdown.

To test whether fused silica mimics the behaviour of sapphire at irradiances greater than 35 TW/cm^2, we fabricated a new set of eight waveguides. A spherical aberration corrected 40× objective (effective NA = 0.5) was used to minimise beam distortion, allowing the peak irradiance of the writing beam to be accurately determined. The induced refractive index change for each waveguide was measured using refractive-index profilometry and the resultant plot is shown in Fig. 5. At lower irradiances (38 TW/cm^2) linearly polarized radiation is observed to induce a greater refractive index change than circularly polarized radiation, however at irradiances greater than 42 TW/cm^2 the situation is reversed and circularly polarized radiation induces a greater refractive index change.

We can infer the relative photo-ionization rates of each polarization from the refractive index change that is induced, as higher photo-ionization rates result in more energy from the writing beam being transferred to the fused silica. Fused silica does indeed exhibit a regime shift whereby the polarization that yields the highest photo-ionization rates (and thus the highest refractive index change) shifts from linearly polarized to circularly polarized radiation as the irradiance increases.

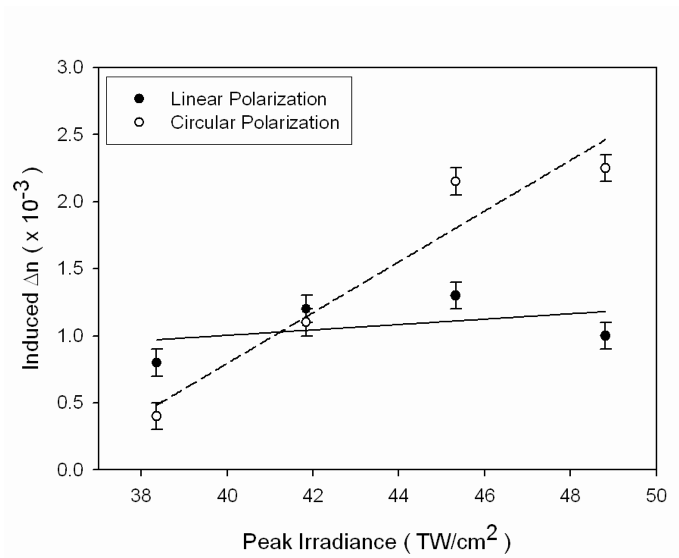

Fig. 5. Induced Refractive Index as a function of the peak irradiance of the writing laser.

We therefore conclude that the greater modification induced by circularly polarized radiation observed throughout this study is due to higher photo-ionization rates for circularly polarized radiation than linearly polarized radiation. We reconcile this result with the work of Temnov *et al*. by showing that higher photo-ionization rates are only obtained using circularly polarized radiation when the peak irradiance exceeds approximately 42 TW/cm^2.

Previous studies have shown that at low NAs (< 0.4) nonlinear effects have the potential to strongly distort a focused femtosecond laser beam through Kerr self-focusing and filamentation, because the critical power for self-focusing is less than the power threshold for damage [26]. It is not clear whether our hypothesis holds in the case where strong nonlinear beam distortion is present, however this is not a concern in waveguide writing regimes, since

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the absence of strong nonlinear distortion is a prerequisite for writing good quality waveguides.

It is interesting to note that Eaton *et al*. performed a recent study on the effect of polarisation in EAGLE2000 borosilicate glass and found that the polarisation of the writing beam did not play a significant role in determining the optical properties of the waveguides [27]. The waveguides studied by Eaton *et al*. were written using high (> 200 kHz) repetition rates. Waveguide formation in this regime is known to rely on thermal accumulation and not on plasma formation [3], and as a result no polarization dependence would be expected in that case.

5. Conclusion

We have experimentally demonstrated that the material properties of fused silica are significantly different after exposure to femtosecond laser radiation for different laser polarizations. Waveguides written with circularly polarized light were shown using NSOM to exhibit greater polish rates than unmodified glass, where waveguides written with linearly polarized light exhibited no such contrast. Using Raman microscopy and refractive index profilometry we observed waveguides written with circularly polarized radiation have a greater concentration of 3 member Si-O ring structures, resulting in densification and a higher refractive index than those written with linearly polarized radiation.

We conclude that the increased degree to which fused silica is modified using circularly polarized radiation can be explained by increased photo-ionization rates when using circularly polarized radiation when compared to linearly polarized radiation for irradiances greater than 42 TW/cm^2. This study shows the importance of optimizing the polarization of the writing laser when fabricating waveguides in isotropic media and shows for the first time how the effect of writing polarization can be understood by looking at the polarization-dependence of ionization mechanisms that drive the modification process.

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