ARTICLE IN PRESS

Available online at www.sciencedirect.com

ScienceDirect

Journal of Non-Crystalline Solids xxx (2007) xxx–xxx

JOURNAL OF
NON-CRYSTALLINE SOLIDS

www.elsevier.com/locate/jnoncrysol

The structural and refractive index changes in the waveguides written by femtosecond laser in Er-doped silicate glasses

Yigang Li, Zian He, Hengsheng Tang, Liying Liu, Lei Xu *, Wencheng Wang

State Key Lab for Advanced Photonic Materials and Devices, Department of Optical Science and Engineering,
School of Information Science and Engineering, Fudan University, Shanghai 200433, China

---

Abstract

Waveguides with low propagation losses were written in erbium-doped silicate glasses by tightly focused femtosecond laser pulses. Raman spectra measurements and 2-D refractive index mapping in the micro-areas near the focus were carried out. The results supported that the refractive index change induced by fs laser pulses comes from the increase of the low rank ring structures in the silicate network.
© 2007 Elsevier B.V. All rights reserved.

PACS: 42.70.Ce; 42.82.Cr; 78.20.Ci; 78.30.−j

Keywords: Planar waveguides; Laser–matter interactions; Photoinduced effects; Raman spectroscopy; Silicates; Rare-earths in glasses; Medium-range order

---

1. Introduction

Waveguides fabrication by femtosecond (fs) laser pulses has attracted much attention recently [1–3]. It was demonstrated that focused fs laser pulses can induce local increase of refractive index inside glass networks without creating obvious damage [2]. A combination of multiphoton absorption and avalanche ionization allows energy to be deposited in a small volume around the focus [4]. An advantage of this technique is that it can work in a wide variety of glasses, compared to other traditional techniques [1]. In addition, fs laser pulses give the potential to directly write complex circuits and three-dimensional optical waveguide structures inside transparent materials [3]. However, the mechanism of how the fs laser pulses induce the refractive index changes in glasses is still not clear. There are two competitive theories. One is the color-center model which attributes the refractive indices changes to the color-centers produced by the fs laser irradiation. These color-centers are in sufficient numbers and strength to alter the refractive indices through a Kramers–Kronig mechanism [5]. The other is the structure-change model which supposes that the deposition of the fs laser pulses energy introduced some sort of the structural changes around the focus that alter the refractive indices. The later theory was supported by the result of Raman spectra changes which indicates a low rank ring structures increase in the fused silica after irradiated by fs laser pulses [6].

Er:Yb-active waveguide amplifiers have very important applications in optical communication systems [7]. Traditional manufacturing techniques such as chemical vapor deposition with subsequent reactive ion etching or ion exchange method have limits in the flexibility in choices of materials or fabrication processes. The direct realization of optical structures by the fs laser pulses in erbium-doped glasses may provide reliable and cheap active devices [8].

In this work, we report on the optical properties in erbium-doped silicate glass waveguides fabricated by fs laser pulses. Results from different writing conditions are compared and discussed. In order to explore the mechanism of the photoinduced refractive index changes in the silicate glass sample, Raman spectra and 2-D refractive index mapping in the micro-areas near the focus of fs laser

---

* Corresponding author. Tel.: +86 21 65642144; fax: +86 21 65641344.
E-mail address: leixu@fudan.ac.cn (L. Xu).

0022-3093/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnoncrysol.2006.12.128

Please cite this article in press as: Y. Li et al., J. Non-Cryst. Solids (2007), doi:10.1016/j.jnoncrysol.2006.12.128

pulses were measured. It was found that the refractive index changes and the abundance of low rank ring structures in irradiated glass increased in the same way. These results give new evidence to support the structural-change mechanism.

2. Experiments

The waveguides were written inside Er:Yb doped silicate glass by fs laser pulses that are from a regenerative amplified Ti:Sapphire laser. This laser system provides 120 fs pulses at a 1 kHz repetition rate and 800 nm wavelength. The laser pulse energy was controlled in a range of 1–5 μJ by a Glan–Taylor prism. The laser beam was focused by a 4x, 0.1 N.A. micro-objective inside a piece of erbium-doped glass. The glass was mounted on a three-dimensional computer-controlled translational stage. All the glass surfaces were optically polished. When the sample moved at rates of 0.5–30 μm/s parallel to the incident laser beam, waveguides were formed along the paths.

The Raman spectra in the glass samples were recorded by a Dilor Labram-1B Raman spectrometer, equipped with a 6.4 mW, 632.8 nm He–Ne laser. The probe laser was focused 5 μm under the surface of the sample. A 100x, 0.8 N.A. micro-objective was used to obtain a lateral spatial resolution of 0.7 μm.

To map the refractive index changes in the micro-area near the focus, the micro-reflection method was employed [9]. The experiment setup is shown in Fig. 1. A He–Ne laser beam was introduced and split into two branches. One was sent to a detector as the reference and the other was perpendicularly focused onto the sample surface. The reflected light from the front surface of the sample was detected. According to the Fresnel formula, the refractive index difference between two positions on the sample can be calculated from the corrected reflectivity accurately:

$$\Delta n = n_2 - n_1 = \frac{1 + \sqrt{(I_2/I_1) \times 0.04}}{1 - \sqrt{(I_2/I_1) \times 0.04}} - 1.5, \tag{1}$$

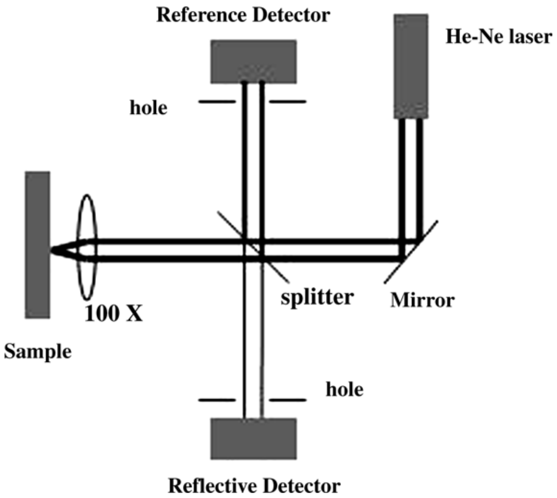

Fig. 1. The setup of the micro-reflection method.

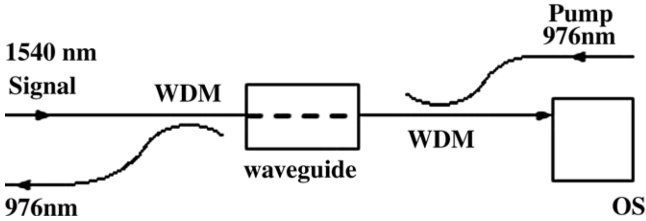

Fig. 2. The experimental setup for measuring optical gain.

where I_1 and I_2 are measured reflected light intensities corrected to the reference arm. The refractive index of our sample is near 1.5, so a reference reflectivity $R = 0.04$ was used in Eq. (1). The Δn measurement error using this method was about 0.001. The sample was mounted on 3-D precise motorized translational stages that were controlled by a computer. The spatial resolution of this setup was 0.5 μm.

The propagating property of the fabricated waveguides was also measured. The 632.8 nm light from a He–Ne laser was coupled into the waveguides and the propagation losses were obtained according to the propagation lines, assuming that the scattered light was proportional to the signal intensity.

The optical gains of these waveguides were evaluated using a standard optical amplifier configuration which is shown in Fig. 2. The probe signal light was from a narrow band ASE source, its central wavelength is 1540.4 nm and the FWHM is 11.6 nm. The total probe power was kept constant at −30 dBm to avoid gain saturation. A 976 nm pump laser from a single mode pigtailed laser source was coupled into the waveguide through a WDM from the reverse direction. The output signal from the waveguide was sent into an AQ6315 Optical Spectrum Analyzer (OSA). The OSA monitored the change of 1540 nm signal with a resolution of 2 nm. The measurement error is about 0.2 dB.

3. Results and discussion

Fig. 3 shows a photograph of three written waveguides with the lengths of 5 mm. Their fabrication conditions were (a) 3 μJ at 1 μm/s, (b) 4 μJ at 1 μm/s and (c) 4 μJ at 0.5 μm/s. The propagation losses of waveguides (b) and (c) were 0.8 dB and 1.0 dB, respectively. However, the decline of scattering light from waveguide (a) was not discerned in our CCD camera, which means that its propagation loss was too low to be measured by this technique.

The enhancements of signal under 200 mW, 976 nm pumping were (a) 1.0 dB, (b) 0.9 dB and (c) 1.0 dB. ASE signal at 200 mW pump power was also measured by disconnecting the probe light. It was only −75.9 dBm (25.7 pW), much lower than the signal difference, about 2.5 nW. So the ASE intensity can be neglected in our case.

Fig. 4 shows the Raman spectra at the centers of five waveguides written using different pulse energies. The

Please cite this article in press as: Y. Li et al., J. Non-Cryst. Solids (2007), doi:10.1016/j.jnoncrysol.2006.12.128

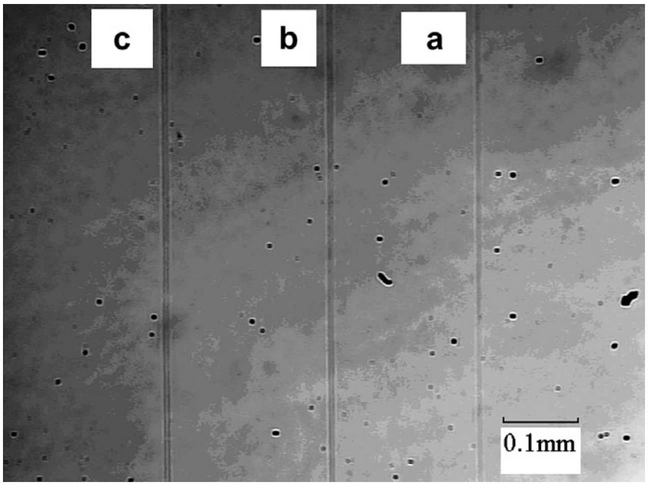

Fig. 3. Waveguides written in Er:Yb co-doped silicate glass. The condition of each waveguides is: (a) 3 μJ at 1 μm/s, (b) 4 μJ at 1 μm/s and (c) 4 μJ at 0.5 μm/s.

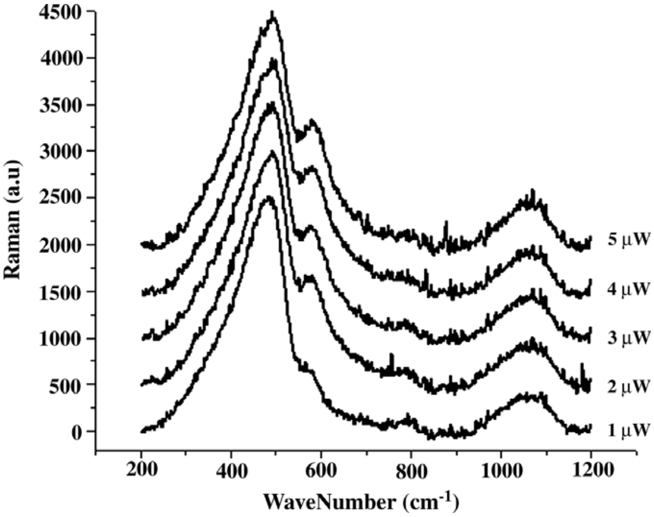

Fig. 4. The Raman spectra at the centers of the waveguides written by different pulse energies. Note that the lines were vertically shifted for better viewing.

strong peak at 450 cm^{-1} is due to the bending vibration of the Si–O–Si bond. The peaks at 800 cm^{-1} and 1060 cm^{-1} come from the symmetrical and anti-symmetrical stretching vibration of Si–O–Si bond, respectively [10]. A shoulder at 580 cm^{-1} was assigned as the breathing mode of the 3-membered ring structure. In fused silica, it is known that the breathing modes of 4-membered and 3-membered ring structures are at 450 cm^{-1} and 605 cm^{-1}, respectively [6]. However, in our glasses, both of the modes were red shifted, most probably due to the glass composition or glass preparing method difference. Moreover, the mode of the 4-membered ring structure totally overlapped with the Si–O–Si vibration mode and is thus hardly recognized. Being normalized by the 1060 cm^{-1} peak, it is clear in Fig. 4 that the peak associated with 3-membered rings grows with the increase of the pulse energy. The increment of this peak intensity reveals that the 3-membered ring structures accumulated in the silicate glass.

The 2-D profile of refractive index changes (Δn) after exposure to the fs laser pulses was measured by the micro-reflection method mentioned above. Fig. 5 shows the distribution of Δn in a 10×10 μm area surrounding waveguide (b) (Fig. 3). Fig. 6 shows the Δn profiles across waveguides generated at different energies. It's obvious that the refractive index changes at the centers of the waveguides also increased as the fs laser pulse energy increases.

Fig. 7 shows the Raman intensities of 3-membered ring structure and the refractive index change versus fs pulse energy in the same plot. As the pump energy increases, both Δn and Raman intensity grow similarly and reach plateaus eventually. This result indicated that Δn certainly has

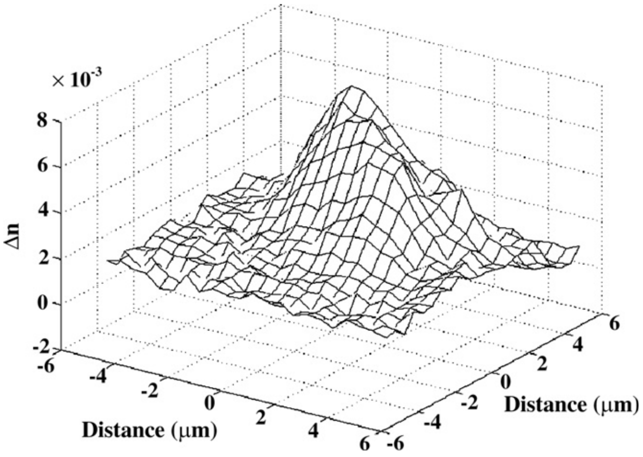

Fig. 5. The three-dimensional mesh figure exhibiting the distribution of refractive index changes in a 10×10 μm square which contains waveguide B. The units of the X–Y axes are μm and the unit of Z axis is 1×10^{-3}.

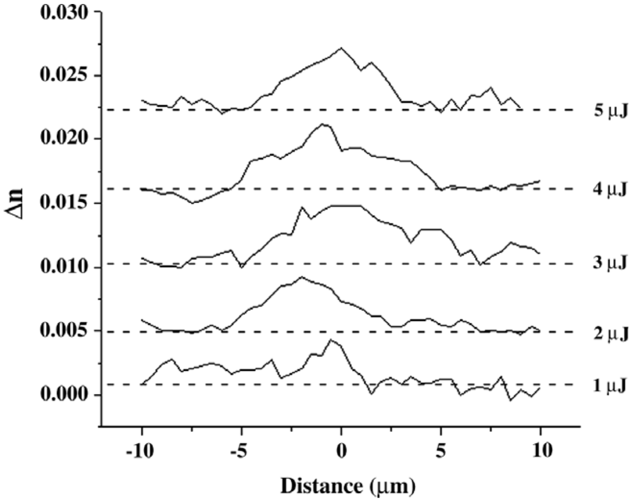

Fig. 6. The refractive index changes along the diameters of the waveguides written by different pulse energies. Note the base lines of each curve were lifted for better viewing.

Please cite this article in press as: Y. Li et al., J. Non-Cryst. Solids (2007), doi:10.1016/j.jnoncrysol.2006.12.128

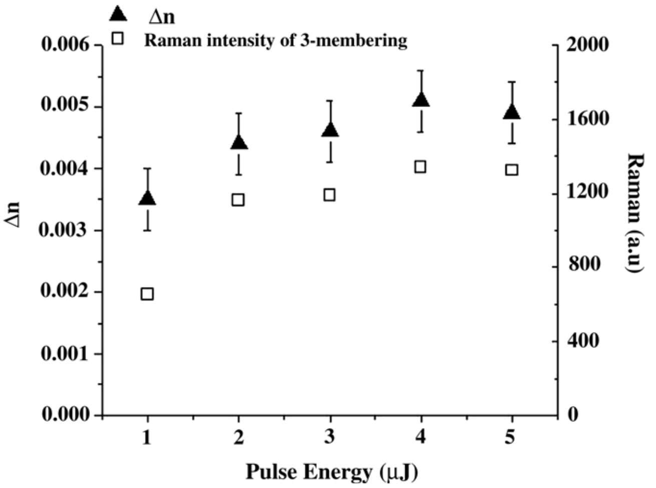

Fig. 7. The Raman intensities of the 3-membered rings and the refractive index changes in five waveguides written by different pulse energies.

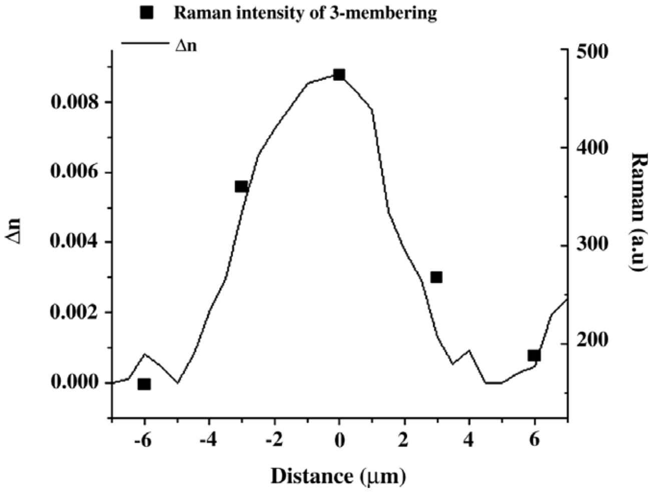

Fig. 8. The refractive index changes and the Raman intensities of the 3-membered rings along the diameter of waveguide B.

a strong positive relationship with the low rank ring structures in silicate glass irradiated by the fs laser pulses.

According to the micro-explosion theory, the high energy of the laser pulse is deposited in a small volume around the focus, and creates a local extreme environment with very high temperature ($\sim 10^6$ K) and pressure (>200 GPa) just like the condition in an explosion [11]. The silicate network in glass will be changed and densified under this extreme condition. As a result, the refractive index increases. Chan reported that the low rank ring structures in fused silica increased with the fs pulse energy exposure and the increment of low rank ring structures in silica will induce densification. His finding was an important support to the structural-change mechanism and micro-explosion theory [6]. However, up to now, no direct relation between the low rank ring structures and the refractive index of the material was reported. Our experimental results give direct solid evidence to support the structural-change model. It's worth noting also that our results were from low loss written waveguides, while Chan's results were from irradiated lines on fused silica samples [6].

Suppose that the focused fs laser has a Gaussian profile, the refractive index distribution of the written waveguide should also have a bell shape. Moreover, if the increment of refractive index is caused by the structural changes, the abundance of the low rank ring structures should have a similar distribution profile.

In Fig. 8, the refractive index profile and the Raman intensities of the 3-member rings across the waveguide (b) (Fig. 3) are plotted. It is shown in this figure that the Raman intensities of the 3-membered rings and refractive index changes both showed a Gaussian-like distribution and each match quite well. The other waveguides exhibited similar results. It's the first report of the comparable Raman spectra and refractive index changes in waveguides fabricated by fs laser pulses. Under the condition of near 1 μJ pulse energy and in silicate glasses, structural changes could be an important mechanism for waveguide formation by focused fs laser pulses.

4. Conclusion

Waveguides with low propagation losses were written in erbium-doped silicate glass by fs laser pulses. The relationships between the refractive index changes and Raman intensities of 3-membered rings at the centers of the waveguides written by different laser pulse energies were studied. The experimental results revealed a close relation between these 3-membered ring structures and the refractive index changes induced by fs laser irradiation. These results support the structural-change model that relates the laser-induced refractive index changes in the silicate glass to the amount of low rank ring structures in the silicate network. Our results provide solid evidence in understanding the mechanism.

Acknowledgments

This work was supported in part by National Natural Science Foundation of China (#60378034, 10474015, 60478005) and Shanghai Science and Technology Commission (#02JD14001, 03DJ14001, 04DZ14001).

References

[1] K. Miura, J. Qiu, H. Inouye, T. Mitsuyu, K. Hirao, Appl. Phys. Lett. 71 (23) (1997) 3329.
[2] C. Florea, K.A. Winick, J. Lightwave Technol. 21 (1) (2003) 246.
[3] S. Nolte, M. Will, J. Burghoff, A. Tuennermann, Appl. Phys. A 77 (2003) 109.
[4] C.B. Schaffer, A. Brodeur, E. Mazur, Meas. Sci. Technol. 12 (2001) 1784.
[5] Alexander M. Streltsov, Nicholas F. Borrelli, J. Opt. Soc. Am. B 19 (10) (2002) 2496.
[6] J.W. Chan, T.R. Huser, S.H. Risbud, D.M. Krol, Appl. Phys. A 76 (2003) 367.

[7] G.C. Righini, S. Pelli, M. Ferrari, C. Armellini, L. Zampedri, C. Tosello, S. Ronchin, R. Rolli, E. Moser, M. Montagna, A. Chiasera, S.J.L. Ribeiro, Opt. Quant. Electron. 34 (2002) 1151.
[8] R. Osellame, S. Taccheo, M. Marangoni, R. Ramponi, P. Laporta, D. Polli, De S. Silvestri, G. Cerullo, J. Opt. Soc. Am. B 20 (7) (2003) 1559.
[9] R. Taylor, C. Hnatovsky, Proc. SPIE 4833 (2002) 811.
[10] F. Gan, Optical and Spectroscopic Properties of Glass, Springer, Berlin, 1992, p. 38.
[11] E.N. Glezer, E. Mazura, Appl. Phys. Lett. 71 (7) (1997) 882.

Please cite this article in press as: Y. Li et al., J. Non-Cryst. Solids (2007), doi:10.1016/j.jnoncrysol.2006.12.128