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Femtosecond Laser-Written Double Line Waveguides in Germanate and Tellurite glasses

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ABSTRACT

The authors report the fabrication and characterization of passive waveguides in GeO_2–PbO and TeO_2–ZnO glasses written with a femtosecond laser delivering pulses with 3μJ, 30μJ and 80fs at 4kHz repetition rate. Permanent refractive index change at the focus of the laser beam was obtained and waveguides were formed by two closely spaced laser written lines, where the light guiding occurs between them. The refractive index change at 632 nm is around 10^{-4}. The value of the propagation losses was around 2.0 dB/cm. The output mode profiles indicate multimodal guiding behavior. Raman measurements show structural modification of the glassy network. The results show that these materials are potential candidates for passive waveguides applications as low-loss optical components.

Keywords: double line waveguide, germanate, tellurite, femtosecond laser writing.

1. INTRODUCTION

Direct femtosecond (fs) laser writing of photonic components and circuits is currently a widely studied application in the area of processing transparent dielectrics[1-4]. The capacity to modify the internal structure and the refractive index of optical glasses and other dielectrics via nonlinear absorption mechanisms is generally pointed out as the responsible phenomena involved, but its mechanisms are still not completely explained and are an ongoing research topic for many kind of materials. Depending on the material properties, as well as on the characteristics of the laser used for inscribing, two types of waveguides can be obtained: the first type consists of a single line, where the modification of the material causes a refractive index increase, leading to light confinement[5]. The second type consists of stress-induced positive refractive index changes in the region adjacent to the writing[6], or negative refractive index changes in the laser focal region[7]. In the latter case, the light is guided between two or more written lines. Single line waveguides have the advantage of simpler and faster processing and have also been demonstrated in a variety of glasses including fused silica, borosilicate[2], chalcogenides[3] and BK7[4]. Double line waveguides have been demonstrated in a number of hosts, including crystals[6, 8-12]. This latter type of writing was the methodology used in this work after previous tests with both types of writing.

Heavy metal oxide glasses, like germanates and tellurites, are interesting materials for photonic applications due to properties such as their high linear refractive index (~2) that is responsible for a high nonlinear refractive index, their extensive transmission window from visible to near infrared, and a lower cutoff phonon energy (<700 cm^{-1}) when compared to silicate, borate, and phosphate glasses. Previously we reported on the fabrication and characterization of active waveguides in GeO_2–PbO–Ga_3O_3 glass samples doped with Er^{3+}, written by a femtosecond laser delivering pulses of 80 fs duration at 1 kHz repetition rate. Single line waveguides were formed under different laser pulse energies and scan velocities, and the passive and active optical properties of the waveguides were investigated[13].

This work presents, to the best of our knowledge, the first demonstration of waveguiding in germinate and tellurite glasses by double line writing by fs laser pulses.

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2. EXPERIMENTAL

2.1 Preparation of glasses

High purity starting oxide powers (99.999%) were used in a conventional melting and quenching method, where the compounds were fully mixed in platinum (TeO_2 –ZnO) or aluminum (GeO_2-PbO) crucibles, melted, and then poured into pre-heated brass molds in air, to avoid internal stress. The temperature and time duration for melting and annealing are presented in table 1 for each glass composition. After cooling, the samples were polished to acquire optical quality. Transparent and homogeneous glasses were produced.

Table 1. Parameters of the fabrication of the bulk glasses.

Glass	Composition (wt %)	Melting/ annealing temperature (C°)	Melting/ annealing time (min)
TeO_2-ZnO	TeO_2: 85.0 ZnO: 15.0	800/325	20/120
GeO_2-PbO	GeO_2: 40.3 PbO: 59.7	1200/420	60/60

2.2 Waveguide writing:

The femtosecond laser setup consists of a Ti:Sapphire laser system operating at λ = 800 nm, able to deliver up to 800 μJ of energy in a pulse with duration ranging from 25 to 200 fs, in a 4 kHz repetition rate pulse train (Femtopower Compact Pro HR/HP, from Femtolasers). Sample translation stages with 300 nm precision were controlled via an integrated CadCam system. The parameters used for writing the waveguides are shown in table 2. Several pairs of closely spaced, parallel waveguides were written using a 20× objective lens with N.A = 0.4, focal length = 10 mm and depth of focus = 200 μm, separated by a distance of 10 μm, using laser energies of 3μJ and 30 μJ. The waveguides were written 0.7 mm beneath the sample's surface. After the waveguide writing, the glasses were re-polished at the input and output facets, which were damaged during the femtosecond writing process. The final length of the obtained waveguides was 1.0 cm.

Table 2. Parameters used in the writing process

Writing speed (mm/min)	1.0
Laser spot size (μm)	5.0
Pulse duration (fs)	80
Wavelength (nm)	800
Polarization in writing direction	parallel
Repetition rate (kHz)	4
Depth of focus (μm)	200

2.3 Materials and Waveguide Characterization:

Absorbance measurements at the laser focus were obtained to analyze how the transparency of the material is affected in this region using a spectrometer (Cary 5000). Optical transmission microscopy was used to capture the images of the laser written structures. A CCD camera was used to analyze how the laser processing affects the surface of the glasses. Figure 1 shows the setup used to determine the near field profile (using a CCD camera), optical losses (using a

Powermeter) and beam quality factor M^2 (using a measurement system formed by a CMOS controlled by a LabVIEW™ program) of a coupled HeNe beam. The optical losses were determined using equation 1, where P_2 represents the output power measured for a sample of length Z_2, and P_1 the output power of a sample with reduced length Z_1[14],

$$\text{Optical losses} = -\frac{10\log\left(\frac{P_2}{P_1}\right)}{Z_2 - Z_1} \quad (1)$$

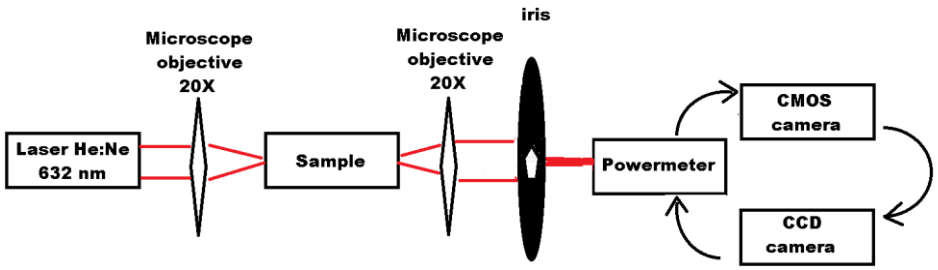


Figure 1. Basic schematic diagram used to determine the near field profile, optical losses and M^2 factor.

The refractive index contrast of the regions in the middle of the written lines and between two written lines was estimated by measuring the output signal of the waveguide at $1/e^2$ width, at a distance of several centimeters, and the numerical aperture (N.A) of the waveguides was calculated from the ratio between the distance and the mode radius. The refractive index change could be estimated by the measured N.A. of the waveguides, as described in ref [15] by the basic equation 2, where n_1 and n_2 represent the refractive index of the core and the cladding, respectively. Raman spectroscopic measurements were made in the focal region of the writing and in the bulk glass using a confocal Raman Microscope (WITEC, model: Alpha300) at λ= 532 nm, power of 45 mW, wavenumber within the range of 0 to 3793 cm^{-1}, a 50× objective and resolution of 0.464 μm.

$$N.A. = \sqrt{n_1^2 - n_2^2} \approx \sqrt{2n_2\Delta n} \quad (2)$$

3. RESULTS AND DISCUSSIONS

Figure 2 shows the absorption spectra of the TeO_2 –ZnO and GeO_2-PbO glasses taken from the bulk and also from the waveguides written by 30 μJ pulses. The spectra for the waveguides processed with lower energy pulse (3μJ) is not shown because there were no measurable differences between them and the bulk spectra. Comparing the results we observe a small change in the transmittance window, mainly for the GeO_2-PbO glasses. These changes may be attributed to nonlinear absorption of the fs laser beam that generates defects such as oxygen deficient centers and non-bridging oxygen hole centers[15, 16].

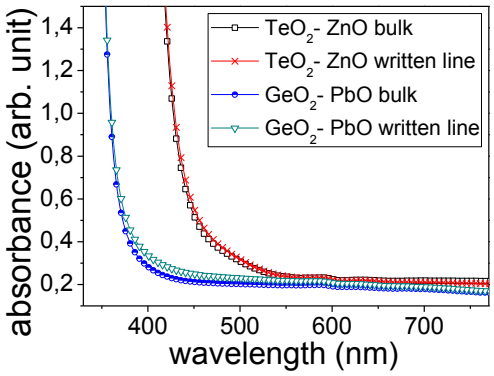
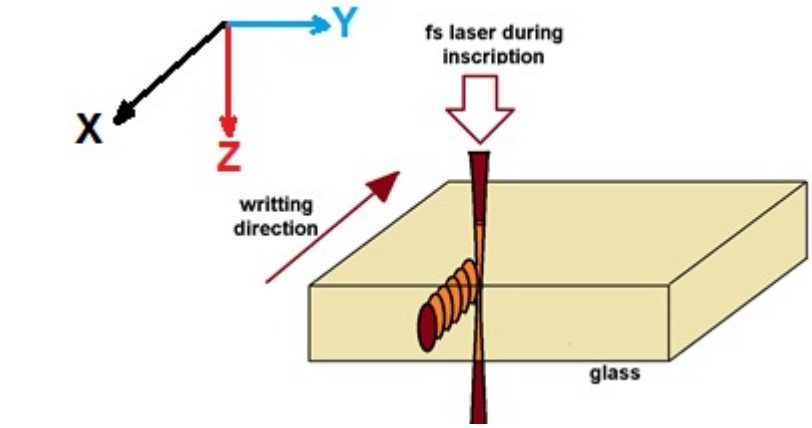
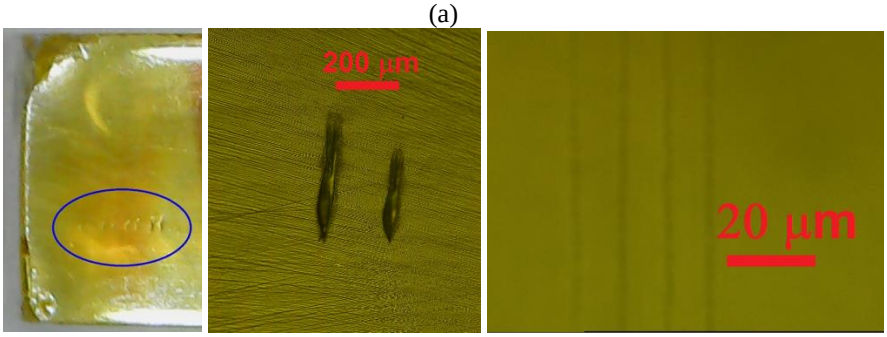


Figure 2. Absorption spectra of the samples (bulk and written area) in the TeO_2-ZnO and GeO_2-PbO glasses.

Figure 3(a) shows a schematic of the direction of the writing process. A picture captured of the output facet of the TeO_2-ZnO glass waveguide after two single-line writing tests is shown in figure 3(b). It is possible to observe the damage at the air/glass interface, making an additional polishing step at the input and output facets necessary after laser processing. Figure 3(c) shows a double line waveguide composed of two writings separated from each other by 10 μm, using a pulse energy of 30 μJ. Each dark line in Figure 3(c) corresponds to the border of a 6 μm wide region affected by the writing process. Guiding occurs in between the two centerlines.



(a)



(b) (c) (d)

Figure 3(a) Schematic picture of the direction of the writing process. (b) Input/output interface of several writing tests of single lines in TeO_2-ZnO glass.(c) Close up photo of the output facet of the TeO_2-ZnO sample showing the effect of the fs laser writing at 30μJ (left) and 3μJ (right) laser pulse energy. (c) Top view microscope image of two written double line waveguides in the GeO_2 – PbO fabricated with 30μJ laser pulse energy.

Figure 4. (a) and (b) images of the TeO_2-ZnO and GeO_2-PbO output faces respectively, showing waveguiding between the double lines for the two waveguides, TeO_2-ZnO and GeO_2 – PbO, written with 30μJ, captured with a CCD at the waveguide exit facet.

To determine the optical losses using equation 1, a powermeter was used to measure the output power, P_2 and P_1 of the samples with total width of Z_2 =1.0 cm, and a reduced width of Z_1=0.6 cm, respectively. The results are shown in table 3 where we can notice that the lowest optical losses were obtained for 30 μJ.

Table 3. Optical loss results for the samples written with different laser energies

Glass	Optical loss (dB/cm) Pulse energy = 30 μJ	Optical loss (dB/cm) Pulse energy = 3 μJ
TeO_2-ZnO	1.25	9.95
GeO_2-PbO	1.75	6.25

The near field profiles at the output of the waveguides are shown in figure 5. Comparing the results for TeO_2 – ZnO *and* GeO_2 – PbO glasses, varying writing speed and energy, we observe improvement of the near field profiles. Figure 5 shows the first obtained waveguide exit beams (writing speed: 6 mm/min, energy pulse=3μJ). However, it was not possible characterize those beams because of low confinement and high scattering of the signal. The other pictures are of lower loss waveguides obtained with writing speed of 1 mm/min and pulse energies of 3 and 30μJ, respectively.

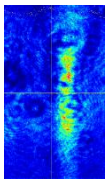
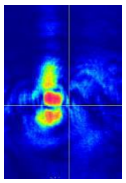
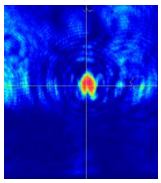
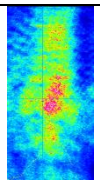
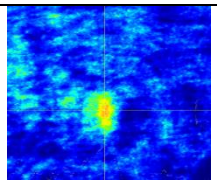
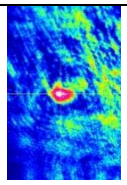
Glass	writing speed: 6 mm/min Energy pulse: 3 μJ	writing speed: 1 mm/min Energy pulse: 3 μJ	writing speed: 1 mm/min Energy pulse: 30 μJ
TeO_2-ZnO			
GeO_2-PbO			

Figure 5. Near field mode profile (at 632 nm) of the beams transmitted by the waveguides.

The refractive index changes (Δn) were determined for the waveguides with the lowest optical losses, fabricated with 30 μJ of pulse energy and 1mm/min writing speed. At λ=632 nm we calculated $\Delta n = 1 \cdot 10^{-4}$ and $\Delta n = 8 \cdot 10^{-4}$ for the TeO_2 – ZnO and GeO_2 – PbO glasses, respectively.

A standard procedure was used to determine the M^2 of the output signal of the waveguides[17], were the heights and widths of the transverse intensity distributions were measured at the $1/e^2$-level to obtain beam propagation factors along perpendicular axes (horizontal x and vertical y). The obtained results are listed in table 4.

Table 4.M^2 measurements

Glass	M_x^2	M_y^2
TeO_2-ZnO	4.62	6.04
GeO_2-PbO	7.24	6.82

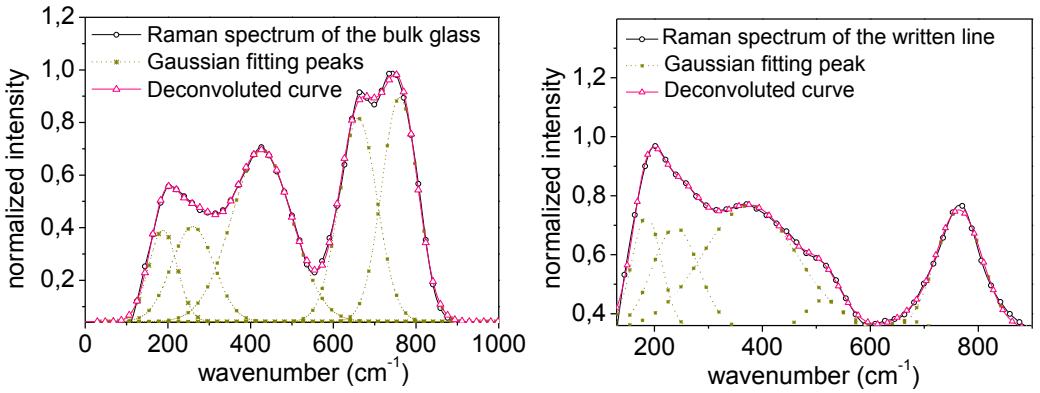
The Raman spectroscopy was crucial in the comprehension on how the fs laser affects the glassy network of the samples. The measurements in the focal region of the written line and between two written lines are shown in figure 6, where the spectra were deconvoluted into Gaussian peaks. In a first analysis we observed that the results in the region between the double lines are similar to the bulk glass, which indicates that no structural change is caused by the fs laser writing technique at the locus of waveguiding.

Some observations in the results of the TeO_2 – ZnO are:

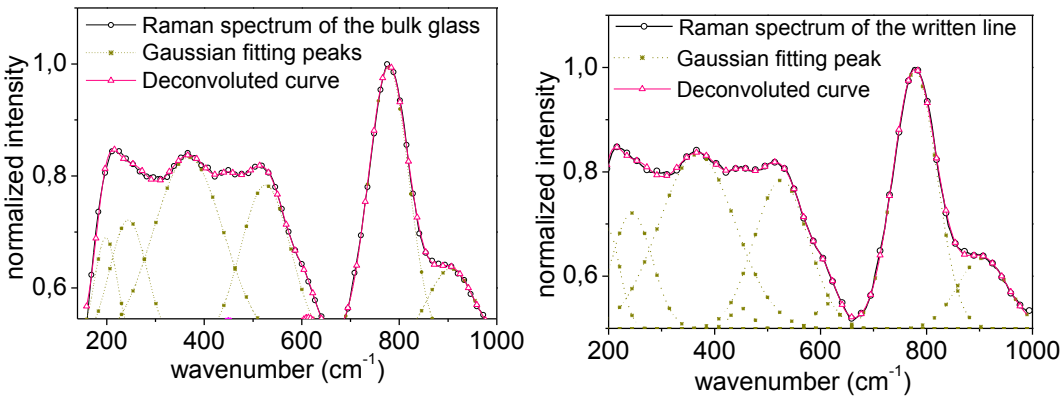
- The boson peak around 200 cm^{-1} (related to low frequency-vibrations and characteristic of amorphous materials) is the highest one in the fs laser written region (fig. 6(b)). Its amplitude is inversely proportional to the structural order of the sample[18], which suggests the formation of different basic glassy units or a less connected structural glassy matrix.
- The peak at 250 cm^{-1} (related to ZnO_4 bending vibrations of Zn-O bonds in ZnO_4 tetrahedrons[19]) is the second highest in the fs laser written region (fig. 6(b)), a behavior not observed in the bulk glass, which suggest that the fs laser written affects the ZnO bonding vibrations of the glass modifier.
- The peak at 360 cm^{-1} (related to Te-O bonds of $-\alpha TeO_2$ [20]) is detectable only on the written region, which suggests changes in the network basic units relation. This peak is related to a stable crystalline phase, created by the laser writing process.
- The peak at 425 cm^{-1} is related to bending vibrations of Zn-O and Te-O-Te linkages formed by corner sharing of (TeO_4), (TeO_{3+1}) polyhedrons and TeO_3 units and is visible in the bulk glass. In the written region this peak quenches and shifts to 450 cm^{-1}, which indicates that the writing processes leads to breaking of the bonds of Te-O-Te and Te -O- Zn[21].
- The peak at 665 cm^{-1}, related to antisymmetrical TeO_4 vibrations that generally assumes large amplitudes in tellurite glasses[21-24], is not visible in the written region, which suggest that antisymmetrical vibrations of TeO_4 do not exist in the written region. This is another evidence of structural changes caused by the laser writing process.
- The peak at 745 cm^{-1}, related to pyramidal TeO_3 units, shifts to 763 cm^{-1} in the written region. This can be attributed to a lesser ordination of this kind of unit, which is a trigonal basic unit of tellurite glasses formed by broken bonds of Te-O[25].

Some observations for the results of the GeO_2 – PbO Raman spectra are:

- A bigger similarity between the results of bulk and laser written areas is observed (figs 6(c) and 6(d)).
- The peak at 509 cm^{-1}, related to symmetric stretching vibrations along the Ge-O-Ge chain[26], shifts to 517 cm^{-1} in the written region. This can be related to a smaller density of these type of bonds in the written region.
- The other peaks at 610, 780 and 900 cm^{-1} are similar for the bulk and the fs-written regions, indicating no influence of the laser writing process. These peaks are related to vibrations of Ge-O bonds in GeO_6 octahedral units and Ge-O- and GeO-Ge symmetric stretching, respectively.



(a) (b)



(c) (d)

Figure 6. Deconvoluted Raman Spectra of TeO_2–ZnO glass: (a) Bulk area, (b) written area and deconvoluted Raman Spectra of GeO2–PbO glass: (c) Bulk area, (d) written area.

Some works in the literature link structural aspects of similar glassy materials with the refractive index. N. Parveen et al demonstrated that in samples with glassy networks[27], formed by a mixture of TeO_4/TeO_{3+1} and GeO_4 structural units, the increase of TeO_{3+1}/TeO_3 units leads to a decrease of the refractive index. A decrease in the TeO_4 content in the written region indicates a transformation of this basic unit into TeO_3, causing a refractive index modification, while the intermediate region between the two lines of writing keeps the bulk properties, having positive refractive index when compared to the adjacent written region.

For the GeO_2-PbO sample, the only evidences for structural modification are the Raman shift of the 509 cm^{-1} peak (observed in the bulk) to 517 cm^{-1} (observed in the written region) and the change of the transmittance window, as shown in figure 2.

It is important to notice that the thermo-mechanical effects caused by the laser irradiation in the fs regime are a complex event, and the conclusion about the phenomena caused by this laser irradiation depends strongly on the process parameters and materials evolved. A list of similar works that aggregate knowledge to this work is available in [28].

In a next step it would be of interest to include doping into the glasses and measure gain[29]. Also, for nonlinear optical devices, it would be of interest to experiment with heavy metal oxide glasses[30,31]. Especially ytterbium and erbium co-doping is of interest there because well-known telecommunication applications[32,33].

4. CONCLUSIONS

Double line waveguides have been written with fs laser irradiation for the first time in TeO_2 – ZnO and GeO_2 – PbO glasses. The mechanisms by which guiding is obtained have been related to structural changes in the glassy matrix, characterized by Raman spectroscopy. The main result caused by this structural modification is a negative index change in the center of the fs laser written lines that causes a relative positive index increase between the two lines causing, thereby, the possibility of light guiding. The measured beam quality factors indicate low-order, multi-mode guiding and the losses of the waveguides are between 1-2 dB/cm.

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