# Femtosecond laser micro-structuring and refractive index modification applied to laser and photonic devices

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#### **ABSTRACT**

Rapid progress has been made in the last few years in the development direct-write, femtosecond laser micro-structuring and waveguide writing techniques in various materials, particularly semiconductor and other photo-sensitive glasses. There is considerable potential for this becoming a disruptive technology in photonic device fabrication, perhaps even leading to the development of devices that are difficult to fabricate by any other technique. We will review these developments, and with an optimistic eye, offer some perspectives on the future of this technology for opto-electronic systems.

#### 1. Introduction

Femtosecond laser micromachining, materials processing and micro-structuring has become important in recent years for many fields including in micro-optics, micro-electronics, micro-biology and micro-chemistry. Laser ablation, because of its non-contact nature, allows the micromachining and surface patterning of materials with minimal mechanical and thermal deformation. It is now well known that for many of these applications the femtosecond regime offers advantages over the nanosecond regime. These advantages lie in its ability to deposit energy into a material in a very short time period, before thermal diffusion can occur. As a result, the heat-affected zone, where melting and solidification can occur, is significantly reduced, leading to structured features that are smaller in size, have higher aspect ratios, and have greater spatial precision. Moreover, at intensities below ablation thresholds, many materials exhibit non-linear absorption that leads to structural changes, either in the surface or in bulk material. It is now recognized that these processes and effects can be utilized to make a number of interesting and potentially useful micro-devices. This is particularly the case for micro-optical components. Laser waveguide-writing potentially can lead to the creation of 3-dimensional structures in transparent media, the possibility of writing active devices in waveguides, and perhaps in the not distant future, the possibility of creating complete optical systems on a single chip with laser writing techniques.

Another advantage of femtosecond laser micromachining is its versatility in so far as the range of materials that can be processed. Femtosecond laser micro-machining is applicable to metals, semiconductors, polymers, oxide ceramics, silica aerogels, optical glasses and crystals, and lends itself to a variety of processing that include the fabrication of photonic crystals<sup>1</sup>, data storage, fabrication of waveguides<sup>2</sup>, gratings and single mode couplers<sup>3</sup>.

#### 2. Femtosecond laser interaction

## 2.1 Femtosecond laser ablation.

The mechanisms that govern femtosecond laser ablation can be complex, depending on the regime being used. In general the following takes place<sup>4</sup>: Bound and free electrons at the surface layer are excited via multi-photon absorption. Hot electrons are generated, the material becomes ionized and a plasma forms at the surface of the material. The energy is then transferred to the lattice through bond breaking and material expansion (Fig. 1).



Fig.1. Schematic illustrating the difference between (a) conventional and (b) femtosecond micromachining

Since the processes described above occur on a picosecond time scale, thermal diffusion into the material is nearly negligible. The thermal relaxation is characterized by the thermal diffusion length D related to the pulse width  $\tau_p$  by  $D = \kappa \tau_p^{1/2}$ , where  $\kappa$  is the thermal diffusivity of the material<sup>5</sup>. If D is shorter than the absorption length, the ablation precedes the thermal diffusion and the material does not have time to melt and re-solidify. As a consequence higher precision in structural micro-machining can be achieved. In addition, in the nanosecond regime, it is generally accepted that ablation begins with the ionization of surface carriers, which are typically defects or impurities<sup>6</sup>. Due to the non-uniform distribution of surface carriers in dielectrics, experiments have demonstrated that no precisely-defined laser-induced damage threshold exists for laser pulses longer than 10 ps. By contrast, ultrashort laser pulses (<200 fs), with target intensities often in excess of  $10^{12}$  W/cm<sup>2</sup>, are capable of freeing bound electrons via Multi-Photon Ionization (MPI). Thus, experiments have shown that the laser-induced damage threshold of an ultrashort laser pulse has a precise value corresponding to the onset MPI, which is completely determined by the ionization band-gap energy of the target.

## 2.2 Femtosecond laser materials modification

In the laser regime in which materials modification occurs, the processes involves are much more materials-specific, and most probably, dependent also on the irradiation conditions. Much of our work has so far concentrated on the uses of chalcogenide glasses. These As-S and As-Se materials have bond structures that are amenable to re-orientation by femtosecond laser light.



Fig. 2. Femtoseconds laser bond re-structuring in As<sub>2</sub>S<sub>3</sub>

Details of the material re-structuring processes can in principal be obtained by a number of techniques, though the difficulty of spatially localizing the diagnostic to the size of the waveguide (typically  $10 \mu m$ ) limits its sensitivity. Perhaps the diagnostic that shows the strongest promise is space-resolves Raman spectroscopy which provides spectral signatures of the bond structure in the modified region. From this diagnostic, with waveguides written in thin films of  $As_2S_3$ , there are strong indications that the structural changes are of the type where As-S bonds re-structure to form As-As and S-S bonds thereby changing the material density and refractive index (Fig. 2).

## 3. Femtosecond micro-ablation and micro-structuring

Femtosecond lasers appear to be a promising tool for the micro-structuring of optical materials. It was demonstrated that simple femtosecond oscillator lasers, without incorporating chirp-pulse amplification (CPA) could produce optical breakdown and structural change in bulk transparent materials using tightly focused pulses of just 5 nJ<sup>7</sup>. This appears a promising approach to take, and we have begun to explore this regime.

Using an extended cavity unamplified Kerr-lens modelocked Ti:Sapphire laser, relief and volume gratings with a 20 µm period were fabricated on a 1.66 µm thick, As<sub>2</sub>S<sub>3</sub> thin film. The laser emission has a spectral bandwidth of approximately 40 nm (FWHM) centered at 800 nm and a repetition rate of 25 MHz. An interferometric autocorrelation measured sub-50 fs pulse duration. The system has an average output power of 0.55 W and produces energies up to 20 nJ per pulse. The output of the laser was focused by a 15x, 0.28NA reflective objective onto a target attached to a 3D motorized translation system. The sample was processed in two regimes: Firstly, the intensity was kept below the ablation threshold, generating a volume grating resulting from photo-darkening. This process is accompanied by a slight photo-expansion, as observed through an interferometric microscope. In the second regime, intensities above the ablation threshold produced a relief grating with grooves of 2-µm depth (Fig. 3).

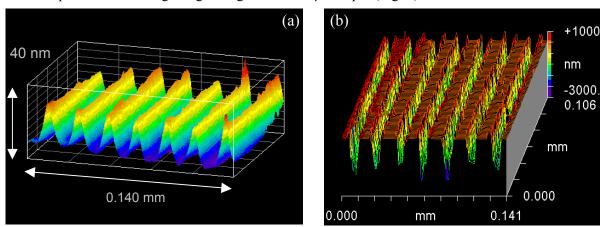


Figure 3 – Surface profile of (a) the phase and (b) relief grating on the As<sub>2</sub>S<sub>3</sub> film produced with sub-50 fs laser pulses from the extended cavity Ti:Sapphire oscillator

These regimes show promise for the fabrication of much finer grating structures. There are advantages to using both phase and relief gratings.

#### 4. Materials modification with femtosecond lasers

A number of research groups have now demonstrated structural modifications in optical glasses with femtosecond lasers<sup>8-13</sup>. Many of the advantages of femtosecond lasers over conventional lasers, and their potential for the fabrication of photonic devices<sup>8,9</sup> stem from their highly localized and deterministic energy

deposition, devoid of most thermally induced effects<sup>14</sup>. In this regime, the focused intensity within the material must reach a certain minimum value, depending on the material, for a structural change to occur. Initially these intensities could only be met with ~100 fs Ti:Sapphire lasers employing chirped pulse amplification (CPA) in conjunction with a regenerative or multipass amplifier, producing pulses at kHz repetition rates, with microJoule energies<sup>8,9</sup>. In these investigations, each micro-element of the material experiences the effect of only a few successive laser pulses. However, if the required intensities can be achieved, there are advantages to be gained from operating at higher repetition rates with lower pulses energies. Recent studies have demonstrated the fabrication of photonic devices using simple femtosecond laser oscillators<sup>11-13</sup>. More deterministic energy deposition control and greater processing speeds can thus be achieved. Moreover, the development of lasers operating in this regime can be simpler in design and avoid the use of separate amplification.

# 4.1. Optical waveguides in Chalcogenide glasses

Most studies using femtosecond direct laser writing have investigated waveguide fabrication in oxide glasses<sup>11-13</sup>. However, the chalcogenide glass (ChG) family exhibits several interesting properties that can be exploited for the fabrication of photonic devices. In particular, their excellent infrared transparency, large nonlinear refractive index, and low phonon energies<sup>15</sup> make ChG films good candidates for the fabrication of all-optical switches<sup>16</sup> and integrated optical elements<sup>17</sup>. Optical waveguides in As-S-Se-based chalcogenide glass have been fabricated by several techniques including photolithography, ion implantation and laser beam writing<sup>18</sup>. The material investigated in this study is Arsenic trisulfide (As<sub>2</sub>S<sub>3</sub>) which, like other chalcogenide glasses, has a semiconductor band structure. Despite its optical bandgap  $E_g$  at 2.35 eV ( $\lambda$ ~517 nm), photosensitive properties have been demonstrated over a broad energy range, from γ-radiation down to infrared, below the bandgap energy. Sub-bandgap irradiation has already been successfully exploited to write holographic gratings and self-written channel waveguides through two-photon-induced processes at 800  $nm^{18}$ .

We have used a 25 MHz Ti; Sapphire femtosecond laser producing 20-nJ pulses with 30-fs duration, a ~40-nm spectral bandwidth centered at 800 nm and 4.5% pulse-to-pulse stability for these studies. The Gaussian laser output beam, with an  $M^2$  of  $\sim 2.6$ , is focused to a 10- $\mu$ m spot with a 2.8 NA, all-reflective Schwarzschild microscope objective. The latter avoids spectral and temporal dispersive effects occurring from common refractive focusing elements. The writing studies were made with thin films of As<sub>2</sub>S<sub>3</sub> thermally evaporated from bulk glass starting materials onto Si/SiO<sub>2</sub> wafers<sup>19</sup>.

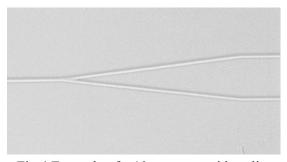


Fig.4 Example of a 10µm waveguide splitter

An example of a typical waveguide structure written in these films is shown in Fig.4). Coupling into  $As_2S_3$  thin films is limited by the high refractive index of the material (n=2.45) which produces high Fresnel losses at both ends of the waveguide. However, the waveguides themselves have a high transmission; more than 70% of the light exiting the film end face (channel and film) was confined in the channel waveguide.

Only a small fraction of the light either did not coupled due to mode mismatch, or was decoupled due to scattering, appearing on either side of the coupled mode.

### 4.2 Optical waveguide structures in polymers: Ring-structure waveguides in PMMA

Controlling the local change of the refractive index of a polymer such as PMMA with a laser would permit the manufacturing of integrated-optical waveguides in polymer integrated devices. One of the advantages of optical polymer technologies is their low cost of production and ease of processing and fabrication. Polymeric materials may be easily engineered to obtain the desired optical parameters such as high nonlinear coefficient values, high electro-optic coefficient values, and enhanced photosensitivity for specific photonic applications<sup>20</sup>. In particular, polymethylmethacrylate (PMMA) is an inexpensive and widely used polymer for the cores of communications grade polymer optical fibers. Moreover, doping species such as conjugated chromophores or rare-earth ions<sup>21</sup> can be incorporated into its polymer matrix for polymer optoelectronics applications.

The current techniques used for polymeric waveguide fabrication include photolithography and reactive ion etching<sup>22,23</sup>, photobleaching<sup>24,25</sup> and high energy ion implantation<sup>26</sup>. These methods are based on conventional planar technologies, and require numerous processing steps and/or involve the design and fabrication of a mask before waveguides can be fabricated. By contrast, direct-write techniques such as laser direct-writing<sup>27</sup> electron beam lithography<sup>28,29</sup> and proton beam writing<sup>30,31</sup> have the advantage of being maskless, allowing one-step fabrication processes. Femtosecond laser direct-writing has recently become an effective method in the fabrication of photonic structures in glasses<sup>2,12,13</sup>. Bends, Y-branches and directional couplers have been demonstrated in PMMA using this technique<sup>27</sup>. However, the photonic devices fabricated by the direct-write techniques were obtained in PMMA thin films, and are therefore restricted to planar configurations. In a companion publication we present the fabrication of three-dimensional ring-structure waveguides in bulk PMMA<sup>32</sup>. Mode profile evaluation of the fabricated structures will be performed by measuring the near-field intensity distribution of the modes.

The use of a simple MHz femtosecond laser oscillators allows greater processing speeds than low-repetition-rate amplified lasers conventionally used in these types of studies. A special Ti:Sapphire oscillator, described in 12 producing 20-nJ pulses, 30 fs in duration, at a 25-MHz repetition rate, is used as the writing laser. The beam is focused by a 10X 0.25-NA microscope objective and the PMMA sample is translated longitudinally with respect to the focal spot. Fig.5 shows an image of the fabricated waveguides observed with a differential interference contrast (DIC) optical microscope.

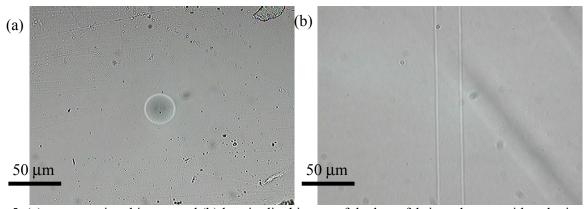


Fig. 5. (a) cross-sectional image and (b) longitudinal image of the laser fabricated waveguides obtained with a differential interference contrast microscope.

Although a positive refractive index change is observed in many materials such as fused silica<sup>10,33</sup> and chalcogenide glass<sup>12,19</sup>, Femtosecond exposure in PMMA produces a negative refractive index change at the

center of the exposed region, which appears darker in the DIC micrograph (Fig. 5). However, a positive index change is observed in the region surrounding the focus spot, which appears lighter in the DIC micrograph. This results in an annular refractive index distribution consistent with what has been observed in some materials such as phosphate glass (Schott IOG-1)<sup>34</sup>. In the case of PMMA, this feature may be caused by the thermal expansion in the focus, leading to an increased optical density in the surrounding region by thermally induced stress. Although no waveguiding was observed in the center of the waveguide, light can be guided in this higher index ring by total internal reflection. Fig. 6 shows the near-field intensity distribution at the output of the waveguide, captured by a Spiricon CCD camera.

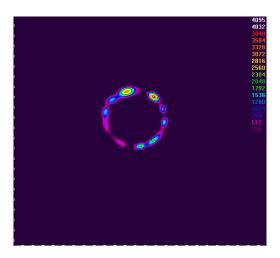


Fig. 6. Near-field intensity distribution at the output of the waveguide captured by a CCD camera

# 4.3 Longitudinal writing in oxide glasses

While planar technology remains the most dominant way of fabricating optical waveguides at the industrial level, laser writing techniques have been proposed that overcome some of the limitations associated with planar technologies. The fabricated structures are no longer confined in a 2-D space, and open up the possibility of fabricating 3-D integrated optical circuits. Moreover, laser writing techniques are in essence simpler than most existing planar technologies, as no mask needs to be designed and fabricated and waveguide writing becomes a single-step process. Although 3-D writing requires a translation of the beam with respect to the sample in both the longitudinal (along the optical axis) and transverse (perpendicularly to the optical axis) directions, it has been reported that the refractive index distribution in the transverse case suffers from high ellipticity<sup>35</sup>. However, in the longitudinal case only, very circular and homogeneous index profiles should be obtained due to the circular symmetry of the system.

There has been much attention focused on waveguides in fused silica, due to its important role in the field of optical telecommunications, and the maturity associated with this material. The prospect of extending direct laser writing techniques to the fabrication of photonic devices in fused silica has led to the fabrication of Y-couplers<sup>19</sup>, single-mode<sup>3</sup> directional couplers and fiber Bragg gratings<sup>36</sup>. Figure 7 shows Differential Interference Contrast (DIC) optical microscope images of different waveguides fabricated in fused silica with different pulse energies. It can be seen that sufficient pulse energies are required in order to induce a refractive index change high enough to allow guiding. However, as the pulse energy is increased, voids and other inhomogeneities start forming within the waveguide core, preventing light from being guided. As a result, the working window of the writing beam must be carefully chosen.

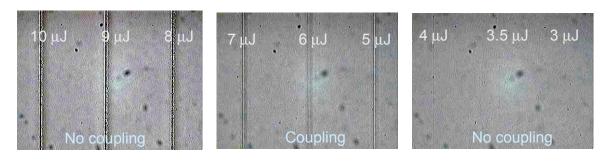


Fig. 7. Longitudinal DIC images of waveguides written with different pulse energies

Although the importance of fused silica is evident, other oxide glasses can be of importance for several applications. For instance, high nonlinearities can be desirable for switching applications, which are currently not viable in fused silica due to its low nonlinear index. We investigate the response of various oxide glasses (obtained from Ohara, Inc.) to fs radiation at 800 nm. The change in the transmission spectrum was measured from 275 nm to 3000 nm and is shown in Fig. 8.

It is important to note that the response varied depending on the composition of the oxide glass. While PBH71, NPH2 and TIH53 show no or little change in their transmission spectrum, LAH51, BSM25 and PHM53 exhibit a significant increase in the absorption at the high frequencies (275 – 600 nm). This result suggests that not all materials have the same potential for direct laser writing of waveguides. Further studies will focus on determining the advantages offered by each composition and evaluate their potential for the direct-write technique.

# 5. Summary

As more experience is gained in the interaction of femtosecond laser emission with transparent optical materials, several factors become evident. At intensities above the ablation threshold, to the first order, most materials are ablated in the same manner. Although ablation rates may vary from one material to another, the basic mechanism is the same for all. However, below the ablation threshold rests an array of fascinating complex processes that are highly variable dependent on material and irradiation conditions. The precise understanding of most of these structural changes remains to be understood, but already some tantalizing possibilities exist. If we can control the refractive index with wavelength-scale precision in three dimensions then there are many unique optical waveguide components can be created within transparent media.

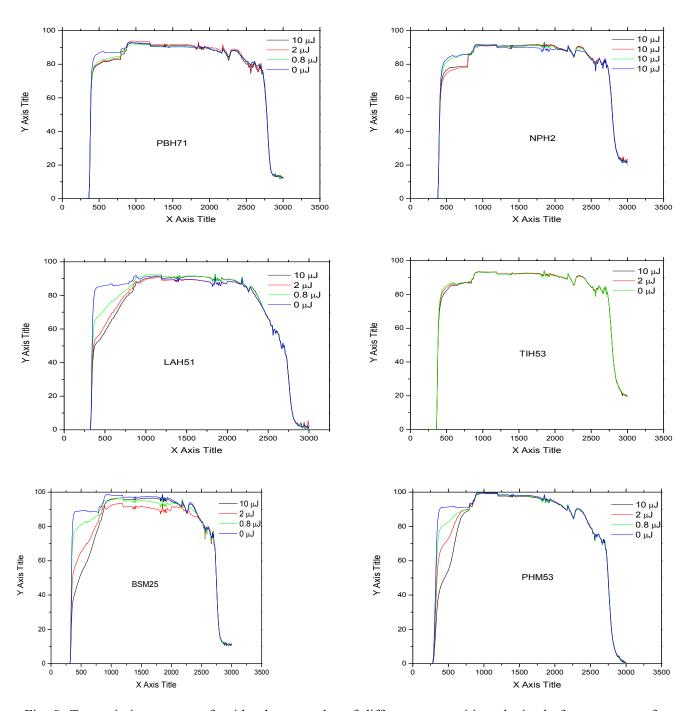


Fig. 8. Transmission spectra of oxide glass samples of different composition obtained after exposure of a 1mm x 1mm region with 0.8  $\mu$ J, 2  $\mu$ J and 10  $\mu$ J

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