Micromachining and material change characterization using femtosecond laser oscillators

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ABSTRACT

We use third harmonic generation (THG) microscopy to image waveguides and single-shot structural modifications produced in bulk glass using femtosecond laser pulses. THG microscopy reveals the internal structure of waveguides written with a femtosecond laser oscillator, and gives a three-dimensional view of the complicated morphology of the structural changes produced with single, above-threshold femtosecond pulses. We find that THG microscopy is as sensitive to refractive index change as differential interference contrast microscopy, while also offering the three-dimensional sectioning capabilities of a nonlinear microscopy technique. It is now possible to micromachine three-dimensional optical devices and to image these structures in three dimensions, all with a single femtosecond laser oscillator.

Keywords: third harmonic generation, nonlinear microscopy, laser-induced breakdown, laser-induced damage, waveguide writing, laser micromachining.

1. INTRODUCTION

The use of femtosecond lasers for bulk micromachining of transparent materials has attracted much attention in recent years. These lasers provide a means to locally alter the refractive index of a transparent material in three-dimensions, potentially allowing complex three-dimensional optical devices to be directly written inside the sample. If adequately perfected, this method would offer significant advantages over the multi-step and inherently two-dimensional techniques, based on photolithography or ion diffusion, currently used to form waveguide structures.

When a femtosecond laser pulse is tightly focused inside a transparent material, the intensity in the focal volume can become high enough to initiate nonlinear absorption of the laser energy by the material through multiphoton, tunneling, and avalanche ionization [1-4]. Energy is only absorbed in the focal volume, where the laser intensity is high, leaving the surface and the material surrounding the focus unaffected [5, 6]. If enough energy is deposited into the focal volume by this nonlinear absorption, permanent material changes are left behind. Depending on laser and material parameters, several different material change morphologies are observed including small density variations [6-8], voids [5, 7, 9], color centers [10], and trapped stress [11, 12]. All of these morphologies lead to a permanent change in the refractive index of the material in the microscopic focal volume. These refractive index changes are the building blocks from which waveguides and other optical devices can be fabricated. To date, three-dimensional binary data storage [5, 13-15] and the fabrication of single- [6, 10, 16-19] and multi-mode [18] waveguides, waveguide splitters [16, 17, 20], and a waveguide amplifier [21] have been demonstrated.

A high laser intensity ($> 10^{13}$ W/cm^2 for most transparent materials) must be reached in order to micromachine the bulk of a transparent material [4]. Under typical focusing conditions used for micromachining (numerical aperture between 0.1 and 0.4) and for a 100-fs pulse, this intensity implies that laser energies of 100 nJ to 1 μJ are required. Reaching these energies requires amplified lasers, adding cost and complexity to the micromachining system. In addition, because the repetition rate of amplified lasers is low, the machining speed is greatly limited. Waveguides produced using 1-kHz amplified laser systems are typically written at a machining speed of only tens of micrometers per second [16].

Commercial and Biomedical Applications of Ultrafast and Free-Electron Lasers, Glenn S. Edwards, Joseph Neev, Andreas Ostendorf, John C. Sutherland, Editors, Proceedings of SPIE Vol. 4633 (2002) © 2002 SPIE · 0277-786X/02/$15.00

In recent work, researchers have shown that, if very tight focusing is used (numerical aperture of 1.2 to 1.4), the intensity necessary for micromachining can be reached with unamplified femtosecond laser oscillators [6, 7, 17]. In addition to offering a simpler and cheaper laser system, because of their higher repetition rate these lasers allow faster processing speeds compared to amplified systems. Waveguides have been directly written into bulk glass at speeds of 20 mm/s using an oscillator with a 25-MHz repetition rate [6].

Micromachining with a high repetition-rate oscillator also enables a new, thermal, mechanism for altering the refractive index of a transparent material [7]. Pulses from a typical laser oscillator arrive at a rate faster than the rate at which energy deposited by one pulse can diffuse out of the focal volume. As a result, multiple pulses irradiating one spot in the sample leads to an accumulation of thermal energy around the focal volume, eventually melting the material around the focal volume. The subsequent resolidification dynamics lead to density variations, and therefore refractive index variations, in the glass that was melted. Waveguides [6, 17] and waveguide splitters [17] have been fabricated in bulk glass using this femtosecond thermal micromachining technique.

With the new capability for fabricating three-dimensional optical devices offered by femtosecond lasers comes the need for an imaging tool to visualize these structures. To date, white light and differential interference contrast (DIC) microscopy have been used to image waveguides written with femtosecond lasers. These techniques do not offer three-dimensional sectioning capability, and are therefore of limited use for evaluating truly three-dimensional structures. What is needed is a microscopy technique that is sensitive to small refractive index changes, has three-dimensional sectioning capabilities, and is intrinsic (no contrast agents can be added to structures machined in the bulk of a sample). Previously, third harmonic generation (THG) microscopy [22-24] has been proposed for imaging laser-induced damage in glass [25]. Here, we further explore how well THG microscopy can image waveguides and other structures written in bulk glass with femtosecond laser pulses.

2. THIRD HARMONIC GENERATION MICROSCOPY OF MICROMACHINED GLASS

Under tight-focusing conditions in a material with normal dispersion, THG is suppressed in a uniform bulk sample because of a phase mismatch [26]. Whenever an interface is located at the focus, however, THG can be efficiently produced [22-24]. Furthermore, because THG is a nonlinear process, third harmonic radiation will only be generated in the focal volume, thus providing natural optical sectioning capabilities. Collecting the third harmonic light as a function of the position of the focus inside a sample therefore allows one to map out interfaces in the sample in three dimensions.

Figure 1 shows a schematic diagram of the THG microscope used in these studies. A passively modelocked Nd:glass laser produces 150-fs, 1064-nm pulses that are focused by a high numerical aperture (NA) microscope objective into the sample. The third harmonic light is collected in the forward direction by a 0.75-NA microscope objective and is focused onto a photomultiplier tube. A short-pass filter and an interference filter block the fundamental wavelength, allowing only the third harmonic radiation to reach the detector. To position the laser focus within the sample, a pair of scan mirrors is used to raster scan the laser beam in the plane perpendicular to the laser propagation direction, and the sample is translated along the laser propagation direction with a calibrated piezoelectric stage. The amount of THG is recorded as a function of the position of the laser focus inside the sample, building up a three-dimensional image of THG efficiency.

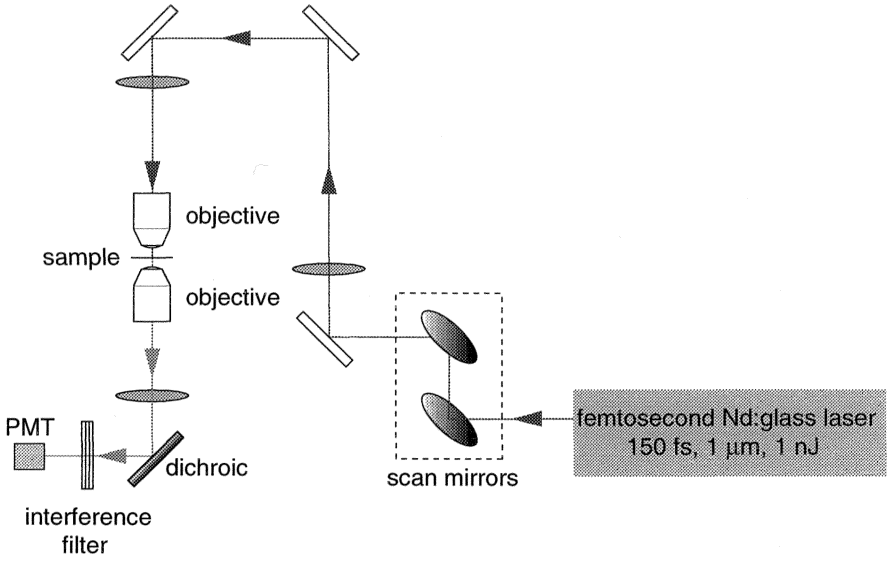

Figure 1. Third harmonic generation microscope. The third harmonic light is collected as a function of the position of the focused femtosecond pulses inside the sample.

We first used THG microscopy to image waveguides written using a femtosecond laser oscillator. To produce the waveguides, a 25-MHz train of 30-fs, 800-nm pulses from a long-cavity Ti:Sapphire laser oscillator [27-29] were focused into a borosilicate glass (Corning 0211) sample with a 1.4-NA oil-immersion microscope objective. The sample was translated perpendicular to the incident direction of the laser beam at a speed of 14 mm/s. Due to the thermal mechanism described above, waveguides with a diameter of about 10 μm are formed [6]. Figure 2 shows rendered three-dimensional THG images of a section of this waveguide taken on the THG microscope. In Figure 2a (2b), the image plane is perpendicular (parallel) to the waveguide axis. In Figure 2c, the waveguide axis projects up and to the left. The imaging laser beam was focused by a 0.65-NA microscope objective to take this data. Note that the core of the waveguide shows more efficient THG than the surrounding material. No post-processing was performed on these images, or any other in this paper, except for simple thresholding to remove the background.

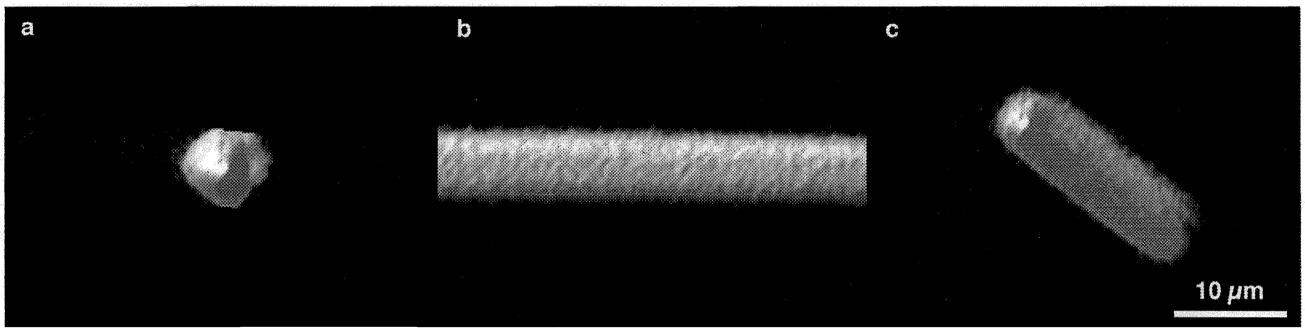

Figure 2. Rendered THG images of a waveguide written using a femtosecond laser oscillator. The image plane is perpendicular (parallel) to the waveguide axis in 2a (2b). In 2c, the waveguide axis projects up and to the left. The THG images were taken with the imaging laser beam focused by a 0.65-NA microscope objective.

Figure 3 shows rendered THG images of the same waveguide as in Figure 2 taken with the imaging laser beam focused by a 1.4-NA oil-immersion microscope objective. The orientations in the three images are the same as in Figure 2. Because the confocal parameter with a 1.4-NA objective is shorter than the diameter of the core of the waveguide, we resolve the interface between the central, core region and the outer ring of the waveguide. This interface is formed during the cooling of the glass that was melted by the femtosecond laser, and likely represents the transition from a

region of material with reduced refractive index (outside) to a region with increased refractive index (inside). In Figure 3a, the THG imaging laser beam is incident from the top of the image. Note that the sides of the interface between the core and outer ring do not show up well, while the top and bottom interface shows significantly enhanced THG. This emphasis of the top and bottom interfaces is caused by the fact that THG microscopy is most sensitive to interfaces that are perpendicular to the propagation direction of the light [24].

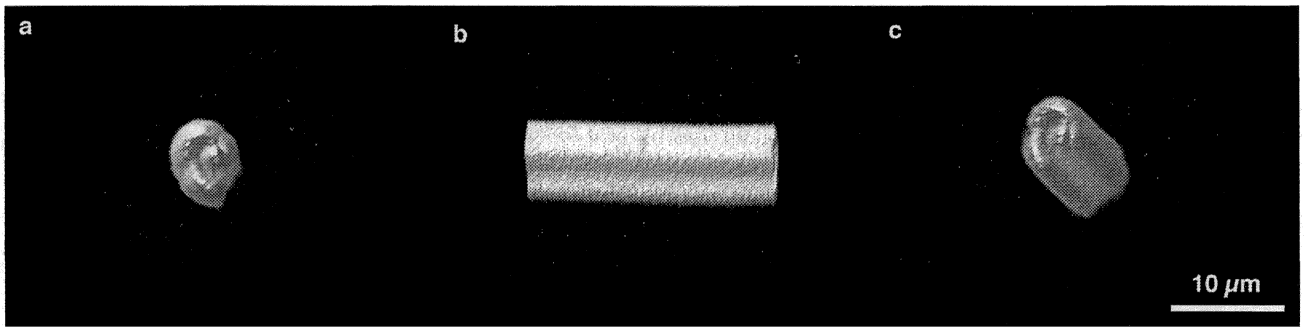

Figure 3. Rendered THG images of a waveguide written using a femtosecond laser oscillator. The image plane is perpendicular (parallel) to the waveguide axis in 3a (3b). In 3c, the waveguide axis projects up and to the left. The THG images were taken with the imaging laser beam focused by a 1.4-NA microscope objective.

Next, we imaged single-shot structural modifications produced by femtosecond laser pulses. Figure 4 shows THG images of structural modifications produced with different laser parameters taken with 1.4-NA focusing of the imaging laser beam. In all images, both the laser which produced the structural modification and the THG imaging laser are incident from the top of the image. Figure 4a shows structures produced in borosilicate glass with 1-μJ, 100-fs, 800-nm pulses from an amplified Ti:Sapphire laser system focused with a 0.45-NA microscope objective. The threshold for permanent structural change under these laser and focusing conditions is about 30 nJ [6], so the energy used here significantly exceeds the threshold. These conditions are comparable to those used to write waveguides with amplified lasers. The conical shape of the structure is due to different time slices of the above-threshold laser pulse producing optical breakdown and therefore damage at different locations upstream from the laser focus [7].

Figure 4b shows structures produced in borosilicate glass using 520-nJ, 100-fs, 800-nm pulses focused by a 1.4-NA microscope objective. For 1.4-NA focusing, the threshold for producing permanent structural change is only 5 nJ [6]. The structures in Figure 4b are conical in shape, but with a steeper cone angle compared to the structures in Figure 4a. The contrast is also higher, indicating that larger refractive index changes are produced with the more tightly-focused pulses. The complex morphology of the structures shown in Figures 4a and 4b requires a three-dimensional imaging technique, such as THG microscopy, to properly image.

In Figure 4c, structures produced with 25-nJ, 100-fs, 800-nm pulses focused by a 1.4-NA objective are shown. At this lower energy, the shape of the structure reflects the focal volume of the microscope objective. Using THG microscopy, we were able to image structures produced with as little as 10 nJ of laser energy. We observed a similar sensitivity using DIC microscopy, one of the most sensitive techniques for imaging refractive index changes. THG microscopy, however, provides three-dimensional sectioning capabilities, an important advantage for looking at complex three-dimensional optical devices or other structures.

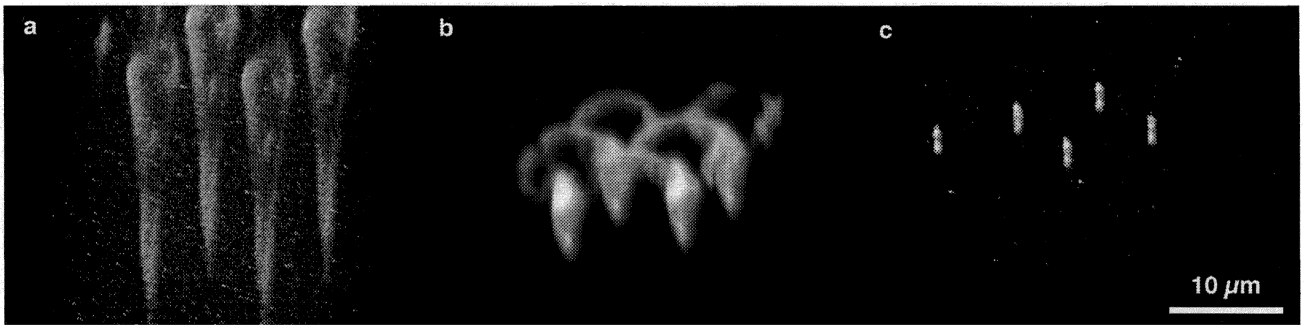

Figure 4. THG images of structural modifications produced by single, 100-fs, 800-nm laser pulses. The THG images were taken with 1.4-NA focusing of the imaging beam. The laser beam producing the structures and the imaging beam were both incident from the top of the image. Parameters of the laser which produced the structures: a) 1 μJ, 0.45-NA focusing; b) 520 nJ, 1.4-NA focusing; c) 25 nJ, 1.4-NA focusing.

3. CONCLUSIONS

Femtosecond oscillators provide both a means to produce refractive index changes in bulk glass and a means to image these changes in three-dimensions with high sensitivity. THG microscopy is an ideal technique for imaging structures written in bulk glass. It is as sensitive to refractive index change as DIC microscopy, it is intrinsic, and it provides three-dimensional sectioning capabilities. Micromachining with a femtosecond laser oscillator provides a faster material processing rate with a simpler laser system, and also enables a new, thermal, mechanism for producing a structural change in the material. Commercially available femtosecond oscillators are adequate for both micromachining and THG imaging, making this technology accessible to researchers without extensive laser expertise and to industry.

There are, however, some disadvantages to micromachining and THG imaging with commercially available femtosecond oscillators. Because the energy from these systems is only a few times the threshold energy for producing a structural change, and because of significant losses in typical high-NA microscope objectives, optical breakdown and therefore energy deposition can only be achieved with very tight external focusing (typically oil-immersion microscope objectives). This tight focusing requirement implies a reduced working depth compared to moderate NA focusing and an increased sensitivity to spherical aberration. The solution to this difficulty is to build femtosecond laser oscillators with higher pulse energy. There have been several promising developments in high-energy oscillators in the last few years, including long-cavity oscillators [27-29], fiber oscillators [30], and thin-disk lasers [31]. Further development of these sources will hopefully allow oscillator-only machining with moderate NA microscope objectives, thereby eliminating the obstacles imposed by oil-immersion objectives. For THG imaging, it is desirable to use longer wavelength femtosecond lasers than Ti:Sapphire-based systems. A longer fundamental wavelength prevents the third harmonic light from being too deep in the ultra-violet and therefore keeps the third harmonic away from the absorption edge of the material being imaged, and makes optical manipulation and detection of the third harmonic easier. Femtosecond lasers with a wavelength of about 1 μm are nearly ideal for THG imaging of structures in transparent materials such as glass, making development of Nd- and Yb-doped laser systems an important priority.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the assistance of Tim Foley in rendering the THG images. This work is partially supported by the National Science Foundation through grant DBI-9987257.

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