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Bulk heating of transparent materials using a high-repetition-rate femtosecond laser

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ABSTRACT Femtosecond laser pulses can locally induce structural and chemical changes in the bulk of transparent materials, opening the door to the three-dimensional fabrication of optical devices. We review the laser and focusing parameters that have been applied to induce these changes and discuss the different physical mechanisms that play a role in forming them. We then describe a new technique for inducing refractive-index changes in bulk material using a high-repetition-rate femtosecond oscillator. The changes are caused by a localized melting of the material, which results from an accumulation of thermal energy due to nonlinear absorption of the high-repetition-rate train of laser pulses.

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1 Introduction

The use of femtosecond lasers to locally alter the structure of bulk transparent materials has attracted much attention in recent years. Waveguides and other optical devices can be fabricated in three dimensions using the refractive-index changes produced in this way. Several applications have been demonstrated, including three-dimensional binary data storage [1–4] and the direct writing of single- [5–9] and multimode [6] optical waveguides, waveguide splitters [10–12], and a waveguide amplifier [13]. While each of these demonstrations relies on nonlinear absorption of femtosecond laser pulses by the material, a wide variety of laser and focusing parameters are used. The mechanism for producing a refractive-index change is likely to be different for different laser, focusing, and material parameters, and a systematic characterization of the refractive-index changes produced in transparent materials under different conditions is needed. Such a characterization provides a micromachining 'roadmap' that will guide users to the best laser and focusing parameters for their application, and will help uncover the fundamental mechanisms for producing refractive-index changes. To this end, we review the laser and focusing parameters that have been applied to date, discuss the refractive-index change produced by each, and mention possible mechanisms that lead to these changes. We then describe, in detail, a new mechanism for producing a refractive-index change in bulk transparent materials that relies on cumulative heating of the material around the focal volume by nonlinear absorption of a high-repetition-rate train of femtosecond laser pulses.

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2 Review of material changes and mechanisms

When a femtosecond laser pulse is focused inside a transparent material, the intensity in the focal volume can become high enough to cause nonlinear absorption of laser energy by the material. In most cases, multiphoton ionization provides seed electrons for avalanche ionization, which leads to the formation of an optically dense plasma [14, 15]. Because the absorption is nonlinear, it occurs only in the focal volume, where the laser intensity is high. If sufficient energy is deposited into the focal volume by this nonlinear absorption, permanent structural (and therefore refractive-index) changes are produced. The mechanism by which these permanent changes are induced by the nonlinearly deposited energy has not, however, been firmly established. There are several mechanisms which are likely to play a role depending on laser, focusing, and material parameters.

We first consider tightly focused pulses with energy well above the damage threshold. For example, when 300-nJ, 100 fs, 800 nm pulses are tightly focused inside a transparent material with a microscope objective with numerical aperture (NA) greater than about 0.4, sub-micrometer-sized regions with a large refractive-index change (up to 0.5) are formed in the material at the focus [1, 9, 16]. These regions, which consist of a hollow or less dense central region surrounded by a densified shell, are the result of an explosive expansion of the highly energetic electron–ion plasma formed by the laser pulse in the surrounding material [17]. Using structures produced in this way, three-dimensional binary data storage with capacities of up to 10^{16} bits/m^3 has been demonstrated [1–4].

For lower laser energy and similar focusing, or for slower focusing and similar laser energy, the refractive-index changes are much smaller. Under tight-focusing conditions and with laser energy up to a few times the threshold for producing an observable structural change (about 20 nJ for

0.65 NA focusing in fused silica), the plasma produced by the laser pulse is not energetic enough to drive an explosive expansion. Instead, a small refractive-index change ($\Delta n \approx 10^{-4}$ to 10^{-2}) is produced in the focal volume [9, 16]. Loose focusing (about 0.1 NA) of 1-μJ pulses produces a larger region with a similar refractive-index change compared to that produced with tight focusing of near-threshold pulses [5–7, 10, 18, 19]. The structures produced using microjoule pulses can be linked together by slowly translating the sample during irradiation with a train of pulses, yielding optical waveguides in bulk glass [4–6, 13, 20]. Because these structures can be used to form waveguides, the refractive-index change in the laser-irradiated volume must be positive, although the refractive-index profile can be complicated and appears to depend on laser parameters [11].

Several mechanisms have been proposed for creating these small refractive-index changes. One of them is thermal; energy deposited by the laser melts the material in the focal volume, and the subsequent resolidification dynamics lead to density (and therefore refractive-index) variations in the focal region [9, 16]. This model is supported by recent measurements of the change in the Raman spectrum from fused silica damaged with femtosecond pulses [21]. Pure fused silica exhibits a maximum in density at a temperature of 1800 K [22]. If the glass is rapidly cooled from this temperature, the higher-density (and therefore higher refractive index) structural arrangement is frozen in [23, 24]. The Raman spectrum indicates the presence of this higher-density structural arrangement of the glass after femtosecond irradiation [21], consistent with the observation that the refractive index increases in fused silica after femtosecond laser irradiation. In other glasses, however, the density decreases with increasing temperature [22], yet waveguides are still formed by femtosecond irradiation. In these other glasses, the positive refractive-index change in the laser-irradiated region cannot be explained by the freezing in of a high-temperature structural arrangement.

Several other mechanisms have been proposed that may play a role in producing a refractive-index change. Electron spin resonance [4, 5], ultraviolet absorption [4, 5], and photoluminescence [21] measurements show that femtosecond laser exposure produces defects and color centers. These defects and color centers could, in principle, produce a significant change in refractive index. The color centers, however, can be annealed away by heating the glass to about 600 K [25], while the refractive index does not revert to its original value until the temperature reaches 1100 K [10], indicating that the color centers cannot be responsible for all of the refractive-index change that is produced [5, 25]. It has also been suggested that femtosecond laser pulses directly drive a densification of the glass by breaking bonds in the material through multiphoton ionization [10]. This mechanism is analogous to the mechanism proposed for ultraviolet-induced densification of fused silica [26].

It has recently been shown that structures produced with loosely focused, 1-μJ pulses are birefringent, with the birefringence axis determined by the direction of the laser polarization [18, 19]. Trapped stress in the material around the focal volume is the likely cause of this birefringence. Because thermal effects are spherically symmetric, the dependence of the birefringence on laser polarization cannot be accounted for by a mechanism based on melting. Instead, the observations suggest that the refractive-index changes are produced by a structural rearrangement that is directly driven by the laser field, such as the bond breaking by nonlinear ionization mechanism discussed above.

Two-photon absorption of loosely focused (0.28 NA), 400-nm, 25-fs pulses in low-band-gap glass (where optical breakdown is not likely to occur) has also been used for micromachining [11]. The structures produced using this technique are birefringent with the axis along the polarization of the incident laser beam and exhibit a small positive refractive-index change ($\Delta n \approx 10^{-3}$) [11]. Because the intensity required to initiate two-photon absorption is low, one can write waveguides into bulk glass using an unamplified laser system using this method, greatly simplifying the laser apparatus required [11]. A photo-induced structural transition and defect formation likely play a role in producing the refractive-index change.

In many micromachining applications, multiple pulses are incident on the same spot inside the sample and cumulative effects must be considered. Indeed, in most of the waveguide-writing demonstrations to date, each region of glass in the waveguide is exposed to thousands of laser pulses. Several reports indicate that the magnitude of the refractive-index change grows as more pulses are incident on one spot in the sample [5, 10, 11, 27]. There have also been reports of incubation effects at the surface (the damage threshold is lower for multiple pulses compared to single pulses) [28, 29], but incubation effects have not been studied in depth for bulk material [27].

In addition to the number of pulses incident on one spot in the sample, one must also consider the rate at which these pulses arrive. In particular, the time between successive pulses should be compared to the characteristic time for thermal diffusion out of the focal volume. If the time between pulses is shorter than the thermal diffusion time there is an accumulation of thermal energy in the focal volume, and the material around the laser focus heats up.

3 Bulk heating with high-repetition-rate femtosecond lasers

We recently demonstrated that we can reach the threshold for optical breakdown in glass using a long-cavity femtosecond oscillator [30, 31] and 1.4 NA focusing optics [9, 16]. The focal volume for 800-nm light and 1.4 NA focusing is less than 0.3 μm^3, yielding a characteristic thermal diffusion time in glass of about 1 μs [32]. The time between pulses from the long-cavity femtosecond oscillator is 40 ns, allowing laser energy to be deposited through nonlinear absorption at a rate that is much faster than the rate at which it is carried away from the focal volume by thermal diffusion. While the mechanism that leads to a refractive-index change is thermal, the energy-absorption mechanism is still nonlinear, and the intensity threshold for optical breakdown must be reached for laser energy to be deposited. To achieve this high intensity with a high-repetition-rate laser, a femtosecond or high-energy picosecond laser source is required. We recently used this cumulative heating effect to write single-mode opti-

cal waveguides in bulk glass [9, 16]. Here we explore the bulk heating mechanism in more detail.

Using a 25-MHz femtosecond laser oscillator, we produced arrays of structures in bulk Corning 0211 (a zinc-doped borosilicate glass) using different numbers of incident pulses. We analyzed these structures using differential interference contrast (DIC) optical microscopy. Figure 1 shows an optical micrograph of structures made with 30-fs, 5-nJ pulse trains focused by a 1.4 NA objective. The number of incident pulses increases, by factors of 10, from 10^2 on the left to 10^5 on the right. The size of the structures shown in Fig. 1 is much larger than either the focal spot size (approximately 0.5 μm) or the size of the structures produced with single pulses [9]. Furthermore, the size of the structures increases with increasing number of laser pulses, and side-view microscopy reveals that they are spherical in shape. A series of rings is evident in the structures on the right in Fig. 1, suggesting regions in the structure with different refractive index.

The structures shown in Fig. 1 are produced by a cumulative heating of the material around the laser focus followed by nonuniform resolidification [9, 16]. Over many laser pulses a volume of material much larger than the focal volume is heated above the melting temperature for the glass. The larger the number of incident laser pulses, the larger the radius out to which the glass melts. After the pulse train is turned off, the material cools and, because of the temperature gradients, resolidifies nonuniformly, leading to the optical contrast shown in Fig. 1. The size of the structures stops growing after about 10^7 pulses. At that point, the structure distorts the laser beam in front of the focus, causing the intensity to drop below the threshold intensity and therefore preventing further energy deposition.

Only a thermal mechanism can account for the structures shown in Fig. 1, because only the sub-micrometer-sized focal volume at the center is directly irradiated, whereas the structures extend up to 10 μm from the focal spot. The spherical shape and increasing size of the structures with increasing number of pulses further points to a thermal melting mechanism.

We modeled the cumulative heating due to successive pulses using a thermal diffusion equation, and calculated the maximum radius out to which the temperature exceeds the melting temperature for different numbers of incident laser pulses. Energy deposition by the laser is modeled as a series of heat sources that are delta functions in time and are spherical in shape with a volume equal to the focal volume for a 1.4 NA objective. The only free parameter is the fraction of each laser pulse that is absorbed by the material. The model best fits the data with 30% absorption, which is consistent with the transmission of the laser pulse slightly above the threshold for permanent structural change [16, 27].

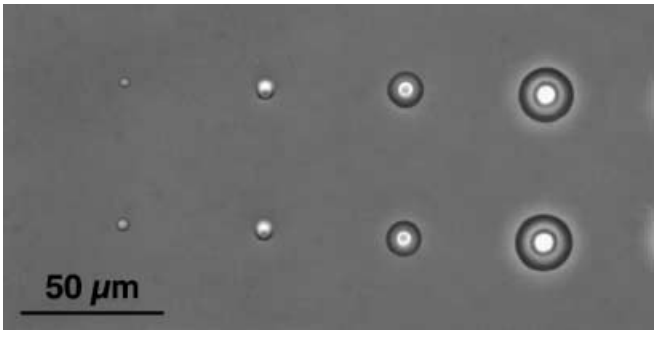

FIGURE 1 Optical microscope image of structures produced with multiple, 5-nJ, 30-fs laser pulses from a 25-MHz oscillator focused by a 1.4 NA objective. The laser pulses are incident perpendicular to the plane of the image, and the number of pulses incident on the sample increases, by factors of 10, from 10^2 on the *left* to 10^5 on the *right*

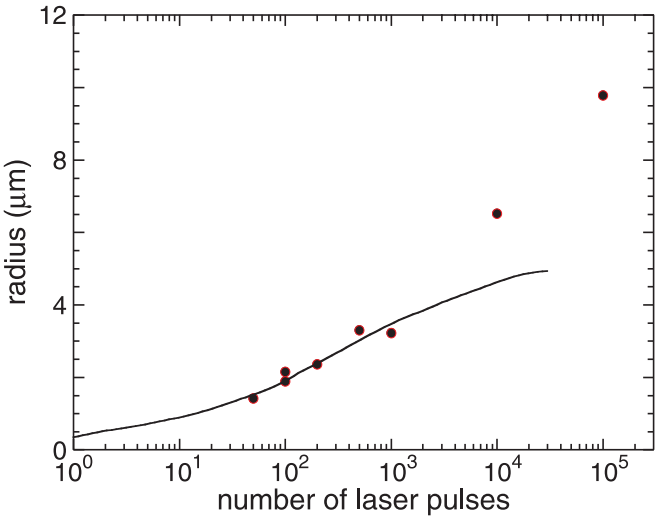

FIGURE 2 Radius of the structure produced with a train of 30-fs, 5-nJ laser pulses incident at 25-MHz and focused by a 1.4 NA objective as a function of the number of incident laser pulses. The *curve* represents the calculated radius of the region where the temperature exceeds the melting temperature of the glass

The circles in Fig. 2 show the radius of the structures measured from DIC images as a function of the number of incident laser pulses. The curve in Fig. 2 shows that the model fits the data well up to about 1000 laser pulses, after which it underestimates the size of the structures. This discrepancy can be attributed to a decrease in the thermal conductivity of the glass as the temperature approaches and exceeds the melting temperature [33] that is not taken into account in the model. As the number of pulses increases, the thermal energy deposited by subsequent pulses must diffuse out through a larger volume of heated material. Because the thermal conductivity is lower in this region, the thermal energy is not carried away as effectively, resulting in more heating around the focal region than would be expected based on the room-temperature value for the thermal conductivity. Consequently a larger volume melts, resulting in a larger structure.

The variations in refractive index shown in Fig. 1 are the result of localized melting and resolidification of the glass. The refractive index at the center must be higher than that at the edges of the structure because we know that a cylindrical structure formed by translating the sample during irradiation forms an optical waveguide. How, then, does the cooling and resolidification lead to such a refractive-index profile? After irradiation, the material melts out to a radius where the temperature equals the melting temperature of the glass. Molten material just inside this maximum radius then quickly quenches and solidifies into a lower-density structural arrangement of the glass. (Unlike fused silica, the density of Corning 0211 decreases when it is quenched as opposed

to slowly cooled.) This quenching continues radially inward as the glass continues to cool. Because there is no free surface that can expand to take up the extra volume occupied by the less-dense glass formed by this quenching, the material near the focal region is put under pressure. As a result of this pressure, the material near the focal region solidifies into a higher-density phase, leading to the higher refractive index at the core of the structures.

4 Applications

The cumulative heating technique described above offers new possibilities for the micromachining of transparent materials. In effect, the laser serves as a point source of heat that can be moved through the bulk of a material to produce a refractive-index change by localized melting and resolidification. Furthermore, by adjusting the number of laser pulses incident on one spot, the amount of thermal energy deposited per unit volume can be controlled with nanojoule precision. No other technique allows such precise deposition of thermal energy in micrometer-sized volumes in bulk material.

This technique has been used to directly write diffraction gratings, single-mode optical waveguides, and waveguide splitters inside bulk glass [9, 12, 16, 34]. By scanning the laser focus inside the sample, the refractive index can be changed in regions of any desired three-dimensional shape. For example, a three-dimensional waveguide splitter can be manufactured by intersecting three waveguides inside a transparent material. Some of the light launched into one of the waveguides couples to the other two at the intersection, making a waveguide beam splitter. Because the three waveguides do not all lie in the same plane, it would be very difficult to fabricate such a splitter using conventional, photolithographic or ion-implantation techniques.

In addition to providing a new mechanism for bulk micromachining, the bulk heating technique described above greatly simplifies the laser requirements and increases the processing speed for laser micromachining. Until now micromachining of transparent materials required amplified laser systems. The technique described here requires only an unamplified laser oscillator, greatly reducing cost and complexity. Furthermore, because of the high repetition rate of unamplified lasers, the micromachining speed is two to three orders of magnitude larger than what can be accomplished with amplified laser systems. We have already demonstrated writing speeds of up to 20 mm/s [9]. This technique could also be used to induce thermally driven chemical changes inside the bulk of a sample. Such changes could yield larger refractive-index changes or, if the solubility properties of the material were changed by the chemical reaction, a freestanding three-dimensional structure could be fabricated.

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