Morphology of femtosecond laser-induced structural changes in bulk transparent materials

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Using optical and electron microscopy, we analyze the energy and focusing angle dependence of structural changes induced in bulk glass by tightly focused femtosecond laser pulses. We observe a transition from small density variations in the material to void formation with increasing laser energy. At energies close to the threshold for producing a structural change, the shape of the structurally changed region is determined by the focal volume of the objective used to focus the femtosecond pulse, while at higher energies, the structural change takes on a conical shape. From these morphological observations, we infer the role of various mechanisms for structural change. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1650876]

In recent years, femtosecond lasers have been used for a multitude of micromachining tasks. For large bandgap materials, in which laser machining relies on nonlinear absorption of high-intensity pulses for energy deposition, structurally altered regions with micrometer size can be produced in the bulk of the material.[1,2] These microscopic material changes are the building blocks from which more complex, three-dimensional devices can be micromachined. Recent demonstrations include three-dimensional binary data storage,[1,3,4] and the direct writing of optical waveguides,[2,5–9] waveguide splitters,[7,10,11] and waveguide optical amplifiers.[12] The growing interest in femtosecond laser micromachining of bulk transparent materials makes it important to uncover the mechanisms responsible for producing permanent structural change.

To micromachine a transparent material in three dimensions, a femtosecond laser pulse is tightly focused into the bulk of the material. High laser intensity in the focal volume induces nonlinear absorption of laser energy by the material via multiphoton, tunneling, and avalanche ionization.[13–16] If enough laser energy is deposited, permanent structural changes are produced in the material at the location of the laser focus. Depending on laser, focusing, and material parameters, different mechanisms may play a role in producing these changes and lead to different morphologies—from small density and refractive index variations, to color centers, to voids.

In this letter, we present a systematic study of the morphology of structural changes produced in the bulk of a borosilicate glass (Corning 0211) by single, tightly focused, 110-fs, 800-nm laser pulses.[17] Using differential interference contrast (DIC) optical microscopy and scanning electron microscopy (SEM), we investigate the dependence of the morphology of structural changes on laser energy and the numerical aperture (NA) of the focusing objective. Our observations show a transition, as the laser energy is increased, from a structural change mechanism dominated by localized melting or densification to one dominated by an explosive expansion.

The threshold energy for producing a structural change is 30 nJ when focusing with a 0.45-NA microscope objective and 4.5 nJ with a 1.4-NA oil-immersion objective. We find that for laser energies less than a few times larger than these thresholds, the shape and extent of the structural change is determined by the focal volume of the objective and the changes in refractive index are small. Structures produced by 50-nJ pulses focused at 0.45 NA are cylindrical and are about 3-μm long [Fig. 1(a)]. Structures like these can be linked together to form single-[5–7,9] and multimode[6,7,9] optical waveguides. With 15-nJ pulses focused at 1.4 NA, the structures are only slightly elongated along the beam propagation direction and are 1-μm long [Fig. 1(b)]. The width of the structures for both 0.45- and 1.4-NA focusing is near the 0.5-μm resolution limit of the optical microscope, and the structures are difficult to visualize without a contrast enhancing microscopy technique such as DIC, indicating that the refractive index change is small.

For laser energies that exceed the threshold energy by

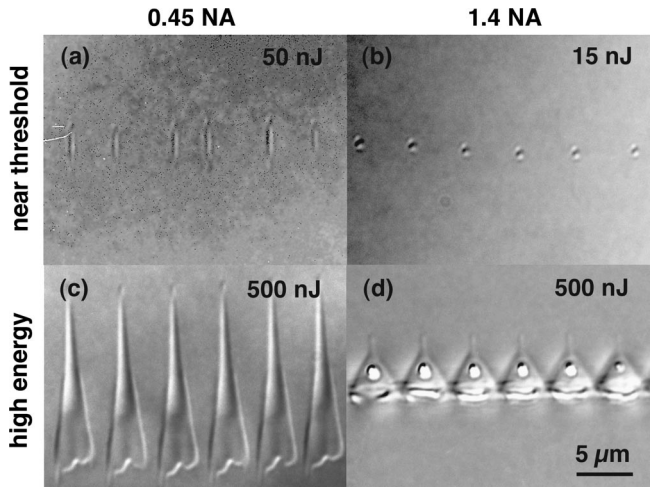

FIG. 1. Side-view DIC optical images of structures produced in bulk glass using single, 110-fs laser pulses with various laser energies and focusing conditions: (a) 50 nJ, 0.45 NA; (b) 15 nJ, 1.4 NA; (c) 500 nJ, 0.45 NA; and (d) 500 nJ, 1.4 NA. The laser pulse is incident from the bottom of the figure.

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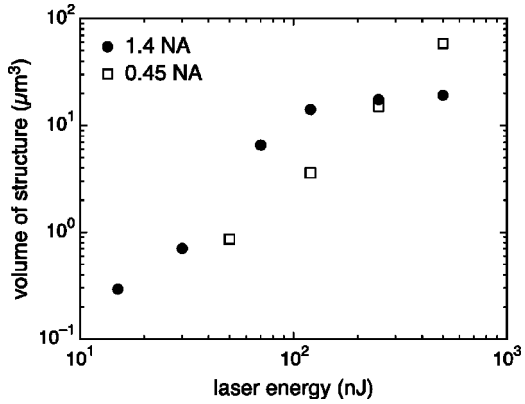

FIG. 2. Plot of the volume of the structure produced as a function of the energy of the laser pulse for 1.4-NA (solid circles) and 0.45-NA (open squares) focusing. The volume of the structures produced with 1.4-NA focusing saturates above 140 nJ, where void formation occurs.

about a factor of 10 or more, the structures show a more complicated morphology. Structures produced with 500-nJ pulses focused at 0.45 NA are much larger than the focal volume, and have a conical shape pointing along the direction of propagation [Fig. 1(c)]. The structures are formed closer to the laser than the focal plane of the objective due to self-focusing.[18] With 500-nJ pulses focused at 1.4 NA, the conical structures have a larger cone angle than with 0.45-NA focusing, and the material at the base of the cone is cracked [Fig. 1(d)]. Also visible is a high-contrast, spherically shaped region in the center of each cone. For 1.4-NA focusing, these spherical regions are produced with laser pulse energies above about 100 nJ, and are visible under standard white-light transmission microscopy, indicating that the refractive index change is larger in these regions than for the rest of the structure.

From images such as those shown in Fig. 1, we calculate the volume over which the structural change extends as a function of laser energy, assuming cylindrical symmetry about the laser propagation direction. For 1.4-NA focusing, the volume increases quickly with increasing laser energy up to about 100 nJ, then increases more slowly, while with 0.45-NA focusing, the volume does not saturate up to an energy of 500 nJ (Fig. 2).

To view the structural changes just shown in greater detail, we used electron microscopy. Because SEM is a surface imaging tool, it is necessary to expose the bulk structural modifications before imaging. Previously, we brought the bulk structures to the surface by polishing the sample down to the level of the structures.[1] Here, we prepare SEM samples by fracturing a thin piece of glass that has been filled with a large number of structures, each produced with a single laser pulse. Some of the structures are bisected by the fracture plane, allowing them to be imaged. After fracturing, the sample is coated with 5 to 10 nm of graphite to make it conducting. In addition to preserving small-scale features that would be smoothed out by polishing, this technique provides side-view images, which are hard to obtain by polishing because of the small diameter of the structures. Recently, this fracture technique has been used to obtain SEM images of structures produced in bulk glass by irradiation with multiple femtosecond laser pulses on one spot in the sample.[19,20] For structures produced by single pulses focused at 1.4 NA, we observe a transition in morphology as the laser energy is increased, from small surface relief [Fig. 3(a)], to a void [Fig. 3(b)], to extensive cracking of the material [Fig. 3(c)]. Voids like those in Fig. 3(b) are responsible for the high-contrast spherical structures seen in Fig. 1(d).

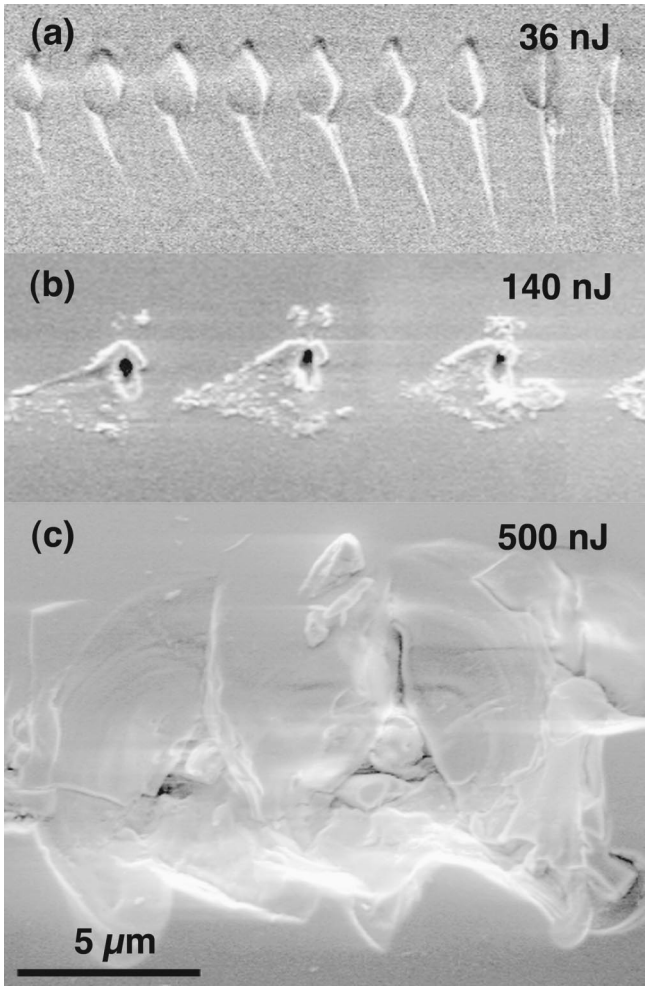

FIG. 3. Side-view SEM images of structures produced in bulk glass using single, 110-fs laser pulses focused by a 1.4-NA oil-immersion microscope objective. The laser energies are: (a) 36 nJ, (b) 140 nJ, and (c) 500 nJ. The laser pulse is incident from the bottom of the figure.

The cone-shaped structures in Figs. 1(c) and 1(d) are the result of different temporal slices of an above-threshold pulse producing breakdown at different positions in the material. The leading edge of the pulse has only enough energy to exceed the intensity threshold for breakdown at the laser focus, and therefore forms the tip of the cone. Subsequent time slices of the pulse have enough energy to produce breakdown upstream from the laser focus, where the laser beam diameter is bigger. Breakdown occurs further and further ahead of the focus, leading to the formation of a conical structure reflecting the shape of the focusing laser beam. The peak of the pulse produces breakdown the farthest upstream from the laser focus, forming the base of the cone. The second half of the laser pulse is absorbed by the plasma formed by the peak of the pulse, leading to the extensive structural changes at the base of the cone seen in Figs. 1(d) and 3(c). This leads to a structure that extends over a region much larger than, and ahead of, the focal volume of the objective, and spans a wide range of material modifications, from small

density and refractive index changes near the tip of the cone, to voids inside the cone, to cracking at the base of the cone.

The voids visible in Fig. 3(b) suggest an explosive mechanism. After laser excitation, hot electrons and ions explosively expand out of the focal region into the surrounding material, leaving a void or less dense central region surrounded by a denser halo.[21,22] The void is not necessarily formed at the laser focus, but rather where the energy density is high enough to drive the expansion [Fig. 1(d)].

For laser energies less than about 100 nJ with 1.4-NA focusing, the energy density is not high enough to drive the explosive expansion just described. The surface relief observed in Fig. 3(a) is most likely caused by fracturing either just above or just below material with a different density.[22] A density change also explains the small refractive index change of the structures shown in Figs. 1(a) and 1(b). These density changes may result from localized melting of material by the laser pulse that, because of strong gradients in temperature and pressure, is followed by nonuniform resolidification.[23,24] Another possibility is that the laser pulse drives a structural transition by directly (i.e., nonthermally) breaking bonds in the material.[7] Silica glasses undergo densification when exposed to ultraviolet light due to ionization in the glass,[25] and femtosecond pulses may ionize the same bonds through a high-order nonlinear process.

In conclusion, using optical and electron microscopy, we characterized the morphology of the structural change produced in bulk glass by tightly focused femtosecond laser pulses with various laser energies and under various focusing conditions. We find that near the threshold for permanent structural change, the structures produced by single pulses consist of small density and refractive index changes with a shape that reflects the focal volume of the focusing objective [Figs. 1(a) and 1(b)]. With very tight focusing, the threshold for structural change is less than 5 nJ, allowing micromachining with unamplified lasers.[2,11] At higher laser energies, the shape of the structure is conical with the tip of the cone oriented along the laser propagation direction [Figs. 1(c) and 1(d)], and voids appear in the material [Fig. 3(b)].

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