Micromachining bulk glass by use of femtosecond laser pulses with nanojoule energy

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Using tightly focused femtosecond laser pulses of just 5 nJ, we produce optical breakdown and structural change in bulk transparent materials and demonstrate micromachining of transparent materials by use of unamplified lasers. We present measurements of the threshold for structural change in Corning 0211 glass as well as a study of the morphology of the structures produced by single and multiple laser pulses. At a high repetition rate, multiple pulses produce a structural change dominated by cumulative heating of the material by successive laser pulses. Using this cumulative heating effect, we write single-mode optical waveguides inside bulk glass, using only a laser oscillator.

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In recent years, optical breakdown induced in transparent materials by femtosecond laser pulses[1–3] and its application to micromachining[4–12] have received much attention. When a femtosecond laser pulse is focused inside the bulk of a transparent material, the intensity in the focal volume can become high enough to cause absorption through nonlinear processes, leading to optical breakdown in the material.[1–3] Because the absorption is strongly nonlinear, this breakdown can be localized in the focal volume inside the bulk of the material, leaving the surface unaffected.[5–8] The energy deposited in the material produces permanent structural changes in the sample,[2,4–8] which can be used to micromachine a three-dimensional structure inside the bulk of the glass. Recent demonstrations include three-dimensional binary data storage[5] and direct writing of optical waveguides[9–11] and waveguide splitters.[12] Until now, however, micromachining with femtosecond lasers has required amplified laser systems operating at kilohertz repetition rates, severely limiting the maximum processing speed in many applications.

In this Letter we demonstrate that one can produce optical breakdown and permanent structural change inside a transparent material by use of only an oscillator. First, we show that in Corning 0211, a borosilicate glass, structural change is produced with as little as 5 nJ of energy for laser pulses that are focused with a 1.4 numerical-aperture (NA) oil-immersion microscope objective. Next we examine the morphology of the structural changes produced by both single laser pulses and multiple pulses at a 25-MHz repetition rate. We find that single pulses produce structures with a diameter of less than 0.5 μm, whereas multiple pulses arriving at a high repetition rate act as a point source of heat inside the material, melting a volume of glass with a diameter that depends on the number of laser shots. Finally, we demonstrate that it is possible to directly write single-mode optical waveguides inside the bulk of a glass sample at a speed of 20 mm/s by use of an unamplified femtosecond laser oscillator.

To determine the energy threshold for permanent structural change we use a light-scattering technique.[7,8] A 100-fs pump pulse from a regeneratively amplified Ti:sapphire laser system is focused beneath the surface of the sample by a microscope objective with a NA of 0.25–1.4. We probe the pumped region of the sample with a He–Ne laser and block the directly transmitted He–Ne probe light. When breakdown occurs the resulting structural changes scatter some of the He–Ne probe light around the beam block into a detector. Figure 1 shows the measured energy threshold for permanent structural change in Corning 0211 as a function of the NA of the microscope objective.

To a first approximation, the energy, E_{th}, corresponding to the intensity threshold for optical breakdown, I_{th}, scales linearly with the laser spot size (i.e., $E_{\mathrm{th}} \propto I_{\mathrm{th}}/\mathrm{NA}^2$). Aberrations and self-focusing both cause a departure from this linear scaling. Aberrations play a role primarily at high NA ($\geq$0.5), at which the paraxial approximation breaks down.[13] We minimized aberrations in the experiments presented here.[14] Self-focusing, on the other hand, is more important at low NA ($\leq$0.65), because the amount of self-focusing depends on the ratio of the peak power of the laser pulse to the critical power for self-focusing for the material and because a higher peak power is needed to reach the breakdown intensity at low

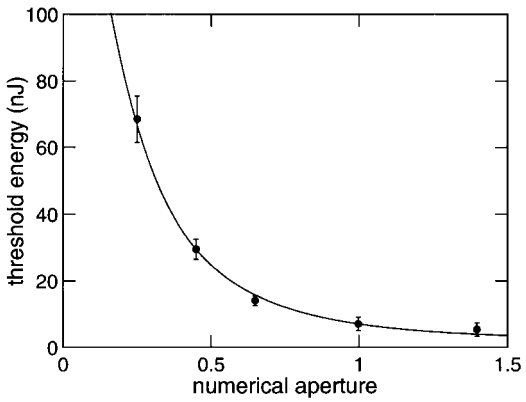

Fig. 1. Dependence of the threshold energy for producing permanent structural change on the NA of the focusing objective for 100-fs laser pulses in Corning 0211. The solid curve shows a fit of Eq. (1)to the data, yielding $I_{\mathrm{th}} = 2.8 \times 10^{17}$ W/m^2 and $P_{\mathrm{cr}} = 1.5$ MW.

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NA.[15,16] As long as the peak power remains well below the critical power, the change in the laser spot size that is due to self-focusing can be accounted for.[18]

For diffraction-limited focusing[14] in the presence of weak self-focusing, the energy corresponding to breakdown intensity is related to the NA of the objective by

$$E_{\mathrm{th}} = \frac{I_{\mathrm{th}} \tau \lambda^2}{\pi (\mathrm{NA})^2 + I_{\mathrm{th}} \lambda^2 / P_{\mathrm{cr}}}, \qquad (1)$$

where P_{cr} is the critical power for self-focusing in the material, τ is the laser pulse duration, and λ is the wavelength of the laser.[17] With the threshold intensity and the critical power for self-focusing as free parameters, a fit of Eq. (1) to the data in Fig. 1 yields a threshold intensity of 2.8×10^{17} W/m^2 and a critical power of 1.5 MW.

Over the entire range of NA's used, breakdown is achieved below the critical power for self-focusing. Consequently, self-focusing is negligible for 1.0 and 1.4 NA, whereas for 0.65, 0.45, and 0.25 NA self-focusing results in 6%, 10%, and 20% decreases, respectively, in spot size. In previous work, self-focusing played a more dominant role,[16] complicating the determination of the threshold intensity for bulk optical breakdown.

To visualize the structural changes induced by the optical breakdown we made arrays of structures and imaged the arrays in a differential interference contrast microscope. Figure 2a shows an image of permanent structures produced in the bulk of Corning 0211 glass by single 100-fs, 10-nJ laser pulses focused by a 1.4-NA microscope objective. The propagation direction of the femtosecond laser beam is into the plane of the image. The structures have a 0.5-μm diameter, corresponding to the resolution limit of the optical microscope. The structures are cylindrical in shape and $\sim$2 μm long. Whereas the voids obtained at higher laser energy suggest an explosive mechanism,[6] the structural changes shown in Fig. 2a could be due to localized melting, to color center formation, or to a mechanism analogous to ultraviolet densification.[18] Details of the mechanism are currently under investigation by our and other research groups.

As Fig. 1 shows, a structural change in the material can be produced with only 5 nJ at 1.4 NA, making it possible to micromachine a material with an unamplified laser. We therefore constructed an extended cavity Ti:sapphire oscillator with a repetition rate of 25 MHz, a pulse energy of 15 nJ, and a 30-fs pulse duration.[19,20] Figure 2b shows bulk structures produced with this long-cavity oscillator by use of trains of 25,000 successive 5-nJ, sub-100-fs laser pulses.[21] The laser beam is incident perpendicularly to the plane of the image. The structures in Fig. 2b have a diameter of 10 μm, much larger than the single-shot structures in Fig. 2a. Several rings, indicating regions of different refractive index, are visible in the image.

At 25 MHz the time interval between successive laser pulses is much shorter than the 1-μs time scale for diffusion of heat out of the focal volume.[22] As a result, successive laser pulses deposit energy faster than it can diffuse away, raising the temperature of the material in the focal region. On a longer time scale, the heat deposited by a succession of thousands of pulses diffuses into the surrounding material and melts a region much larger than the focal volume. The glass resolidifies nonuniformly, providing the optical contrast observed in Fig. 2b. The diameter of the structures increases approximately as the number of incident laser pulses to the one-fourth power. The diameter saturates at $\sim$20 μm after $\sim 10^6$ pulses, most likely because the structure itself distorts the focus enough to lower the laser intensity below the breakdown threshold, thereby preventing further energy deposition. Similar cumulative heating effects have been seen in surface ablation of fused silica by use of high-repetition-rate, high-energy, ultrashort pulse trains.[23]

Figure 3 shows cylindrical structures written inside bulk Corning 0211 glass by translation of the sample at 20 mm/s perpendicularly to an incident 25-MHz train of 5-nJ, sub-100-fs laser pulses focused with a 1.4-NA objective. The cylinders form single-mode optical waveguides, indicating that the refractive index is larger at the center than in the surrounding material. This change in refractive index suggests that the glass at the center of the cylinder densifies as it cools. The inset in Fig. 3 shows an end view of one of these waveguides after cleaving of the glass to produce a clean face. The same ring structure as in Fig. 2b is evident in the end view.

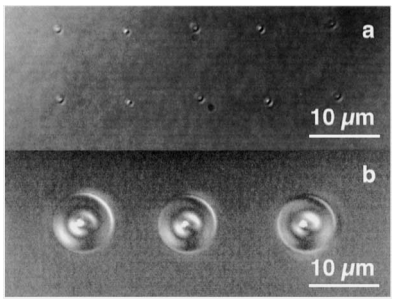

Fig. 2. Optical microscope images of structures produced in bulk Corning 0211 by a, single 10-nJ, 100-fs laser pulses and b, 25,000 5-nJ, sub-100-fs pulses at a 25-MHz repetition rate. In both cases the pulses were focused by a 1.4-NA microscope objective.

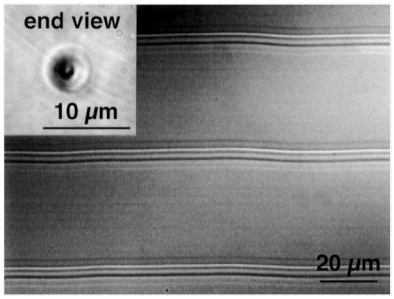

Fig. 3. Optical microscope image of waveguides written inside bulk glass by a 25-MHz train of 5-nJ, sub-100-fs pulses focused by a 1.4-NA microscope objective. The sample was translated at 20 mm/s perpendicularly to the incident direction of the laser beam. The inset shows one end face of a waveguide.

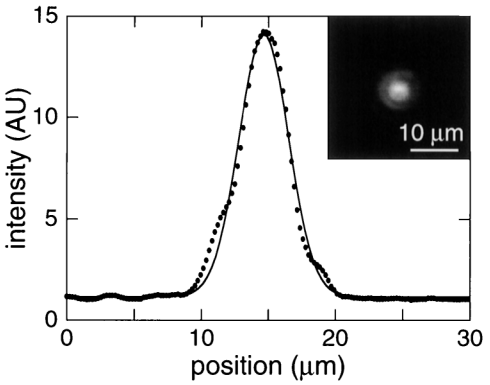

Fig. 4. Waveguide output profile at 633 nm. The points correspond to a cross section of the near-field mode, and the curve represents a best-fit Gaussian. The inset shows a CCD image of the near-field mode.

Figure 4 shows the near-field output profile of one of these waveguides for 633-nm laser light. As the data show, the waveguide has a single-mode, near-Gaussian output profile for visible wavelengths. From the divergence of the waveguide output we find that the refractive-index change between the core of the waveguide and the surrounding material is approximately 3×10^{-4}.

In conclusion, we have shown that by tightly focusing femtosecond laser pulses one can produce optical breakdown and structural change in glass with pulse energies that can be obtained without amplification. Near the 5-nJ threshold a single pulse produces a submicrometer-sized structurally altered region inside the material. At a high repetition rate, the laser acts as a point source of heat inside the bulk of the material, and the morphology of the resulting structural change is dominated by thermal effects. We also demonstrated micromachining of single-mode optical waveguides inside bulk glass at a writing speed of 20 mm/s. This technique makes it possible to directly write waveguides in three dimensions into bulk optical materials and opens the door to the fabrication of a wide variety of passive and active optical devices for the telecommunications industry.

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References

1. D. Du, X. Liu, G. Korn, J. Squier, and G. Mourou, Appl. Phys. Lett. **64,** 3071 (1994).
2. B. C. Stuart, M. D. Feit, S. Herman, A. M. Rubenchik, B. W. Shore, and M. D. Perry, J. Opt. Soc. Am. B **13,** 459 (1996).
3. M. Lenzner, J. Kruger, S. Sartania, Z. Cheng, Ch. Spielmann, G. Mourou, W. Kautek, and F. Krausz, Phys. Rev. Lett. **80,** 4076 (1998).
4. H. Varel, D. Ashkenasi, A. Rosenfeld, M. Wahmer, and E. E. B. Campbell, Appl. Phys. A **65,** 367 (1997).
5. E. N. Glezer, M. Milosavljevic, L. Huang, R. J. Finlay, T.-H. Her, J. P. Callan, and E. Mazur, Opt. Lett. **21,** 2023 (1996).
6. E. N. Glezer and E. Mazur, Appl. Phys. Lett. **71,** 882 (1997).
7. C. B. Schaffer, A. Brodeur, N. Nishimura, and E. Mazur, Proc SPIE **3616,** 143 (1999).
8. C. B. Schaffer, A. Brodeur, N. Nishimura, and E. Mazur, in *Digest of Quantum Electronics and Laser Science Conference*, 1999 OSA Technical Digest Series (Optical Society of America, Washington, D.C., 1999), p. 232.
9. K. M. Davis, K. Miura, N. Sugimoto, and K. Hirao, Opt. Lett. **21,** 1729 (1996).
10. K. Miura, J. Qui, H. Inouye, T. Mitsuyu, and K. Hirao, Appl. Phys. Lett. **71,** 3329 (1997).
11. C. B. Schaffer, A. Brodeur, J. F. Garcia, W. A. Leight, and E. Mazur, in *Digest of Conference on Lasers and Electro Optics*, 2000 OSA Technical Digest Series (Optical Society of America, Washington, D.C., 2000), p. 375.
12. D. Homoelle, S. Wielandy, A. L. Gaeta, N. F. Borrelli, and C. Smith, Opt. Lett. **24,** 1311 (1999).
13. A. E. Siegman, *Lasers* (University Science, Mill Valley, Calif., 1986), p. 630.
14. We minimize focusing aberrations by (1) using microscope objectives (Zeiss, Inc.) that are compensated for aberrations in the Corning 0211 glass, (2) focusing at the minimum aberration depth in the sample (170 μm), and (3) uniformly filling the full aperture of the objective. For NA > 0.65, the threshold energies shown in Fig. 1 decrease as $1/\mathrm{NA}^2$, indicating that the focusing is diffraction limited.
15. N. Bloembergen, IEEE J. Quantum Electron. **QE-10,** 375 (1974).
16. M. J. Soileau, W. E. Williams, M. Mansour, and E. W. Van Stryland, Opt. Eng. **28,** 1133 (1989).
17. J. H. Marburger, Prog. Quantum Electron. **4,** 35 (1975).
18. M. Douay, W. X. Xie, T. Taunay, P. Bernage, P. Niay, P. Cordier, B. Poumellec, L. Dong, J. F. Bayon, H. Poignang, and E. Delevaque, J. Lightwave Technol. **15,** 1329 (1997).
19. S. H. Cho, U. Morgner, F. X. Kartner, E. P. Ippen, J. G. Fujimoto, J. E. Cunningham, and W. H. Knox, in *Digest of Conference on Lasers and Electro-Optics*, 1999 OSA Technical Digest Series (Optical Society of America, Washington, D.C., 1999), p. 470.
20. A. R. Libertun, R. Shelton, H. C. Kapteyn, and M. M. Murnane, in *Digest of Conference on Lasers and Electro Optics*, 1999 OSA Technical Digest Series (Optical Society of America, Washington, D.C., 1999), p. 469.
21. A prism compressor is used to compensate for the dispersion in the microscope objective. The duration of the oscillator pulse at the focus was not measured, but the optical breakdown threshold was slightly lower than that for a pulse known to have a 100-fs duration at the focus. With only a simple prism compressor for dispersion compensation, a pulse duration as short as 15 fs has been achieved at the focus of high-NA objectives; see D. N. Fittinghoff, J. A. Squier, C. P. J. Barty, J. N. Sweetser, R. Trebino, and M. Müller, Opt. Lett. **23,** 1046 (1998).
22. H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids* (Oxford University, London, 1959), p. 256.
23. K. P. Chen, A. Oettl, P. R. Herman, and R. S. Marjoribanks, in *Digest of Conference on Lasers and Electro-Optics*, 1999 OSA Technical Digest Series (Optical Society of America, Washington, D.C., 1999), p. 357.