Writing waveguides in glass with a femtosecond laser

K. M. Davis, K. Miura, N. Sugimoto, and K. Hirao

Hirao Active Glass Project, Exploratory Research for Advanced Technology, Research Development Corporation of Japan,
15 Mori Moto-Cho, Shimogamo, Sakyo-Ku, Kyoto G06, Japan

Received May 6, 1996

With the goal of being able to create optical devices for the telecommunications industry, we investigated the effects of 810-nm, femtosecond laser radiation on various glasses. By focusing the laser beam through a microscope objective, we successfully wrote transparent, but visible, round-elliptical damage lines inside high-silica, borate, soda lime silicate, and fluorozirconate (ZBLAN) bulk glasses. Microellipsometer measurements of the damaged region in the pure and Ge-doped silica glasses showed a 0.01–0.035 refractive-index increase, depending on the radiation dose. The formation of several defects, including Si E' or Ge E' centers, nonbridging oxygen hole centers, and peroxy radicals, was also detected. These results suggest that multiphoton interactions occur in the glasses and that it may be possible to write three-dimensional optical circuits in bulk glasses with such a focused laser beam technique. © 1996 Optical Society of America

Since the 1970's, many investigations of the effects of UV radiation damage in high-silica glasses (especially Ge-doped silica glass) have been performed with the objective of producing optical devices (e.g., Bragg gratings) in fibers and thin films.[1] In contrast, laser damage by visible and IR laser light has received little attention owing to the low photon energy at these wavelengths. The development of high-energy-density femtosecond pulse lasers, however, has prompted us to investigate the unexplored potential for inducing multiphoton photochemical reactions and optical devices in glass by use of lasers of sub-UV photon energy. Our results show that stable, visible laser damage and photoinduced refractive-index changes can be achieved with a red visible femtosecond laser.

A regeneratively amplified 810-nm Ti:sapphire laser that emits 120-fs, 200-kHz, mode-locked pulses and delivers an average power of 975 mW was used for our study. The 5-mm-diameter beam was focused through 5–20× microscope objectives and injected into polished plates of dry and wet silica, Ge-doped silica, borate, soda lime silicate, and fluorozirconate (ZBLAN) glasses. The average power of the laser beam at the sample location was controlled between approximately 40 and 800 mW by neutral-density filters that were inserted between the laser and the microscope objective. With the help of an XYZ stage, the samples were translated at rates of 100–10,000 μm/s either parallel or perpendicular to the incident laser beam, thus creating visible damage lines inside the glasses.

For the silica and the Ge-doped silica glasses, we made cross sections of the damage lines by cutting the samples and polishing the exposed surfaces. Refractive-index profiles of the cross sections were measured with a microellipsometer. For UV and electron spin resonance (ESR) spectroscopic measurements, we took scans before and after we wrote identical damage line patterns in glass plates by using a 10× microscope objective and translating the samples perpendicular to the incident light at a rate of 100 μm/s. For these samples, the average laser power at the sample was ~470 mW. Assuming a square-wave pulse, a uniform beam intensity, and a diameter of the laser focal point that is equal to the thickness of the observed damage lines (~6 μm), we found that the samples experienced 12,000 pulses/spot, and each spot was subjected to a dose of 100 MJ/cm^2 during a single pass of the laser.

For all translation speeds and directions, structural changes were induced along the path traversed by the focal point of the laser, and colorless, transparent linear damage marks were produced inside every glass that was probed. These damage lines and their cross sections are visible when transmitted light optical microscopy is used (Fig. 1) and are stable at room temperature. When the laser beam was focused on the near surface of the samples, machining of the glass, most likely owing to thermal shock, occurred at high laser powers, but when the beam was focused on the interior of the samples, no cracking was detected in any of the glasses regardless of laser power. For the

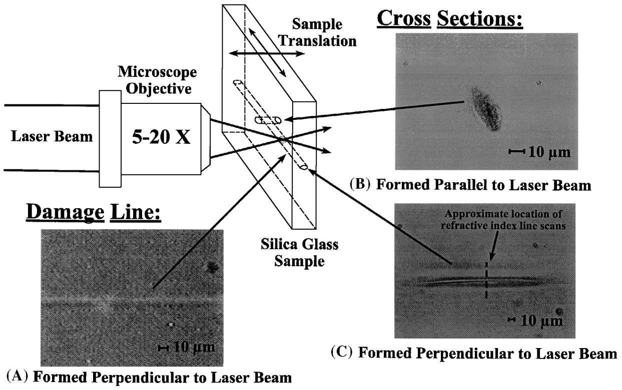

Fig. 1. Laser damage process with images of (A) a laser damage line, (B) the cross section of a line written by translation of the sample parallel to the incident laser beam, and (C) the cross section of a line written by translation of the sample perpendicular to the incident laser beam by a 5× microscope objective. The dashed line in (C) represents the path traversed during the microellipsometer measurements.

high-silica glasses, the damage lines produced when the samples were translated parallel to the axis of the laser beam were roughly cylindrical, with diameters of approximately 12 to 5 μm for the 5–20× microscope objectives. The cross sections of the lines created when the samples were translated perpendicular to the laser axis were elliptical because of the depths of focus of the lenses, and the long dimension of the cross sections ranged from approximately 275 to 100 μm for the 5–20× lenses. For the high-silica glasses, the size of the damage was fairly constant with changing translation speeds. For the soda lime silicate, ZBLAN, and borate glasses, however, the dimensions of the damage area changed significantly with translation speed, and when these glasses were held motionless in the laser beam the damage spread radially from the focal point of the laser with time as if the glass were being slowly melted.

Because of the potential telecommunications applications for the pure and the Ge-doped silica glasses, the properties of the damage in these glasses were examined first. Using a microellipsometer, we measured refractive-index profiles across the cross sections of damage lines that were formed perpendicular to the laser beam [see Fig. 1(C)]. For the pure and the Ge-doped silica glasses, a single pass of the laser produced refractive-index increases of approximately 0.015 and 0.01, respectively, at the center of the damage region (Fig. 2). After 10 passes of the laser, the refractive index at the center of the damage was ~ 0.035 higher than that of the surrounding glass in the Ge-doped silica. The refractive indices of the as-received glasses vary by less than ±0.0005 across the probed region according to the manufacturers' specifications, and the error in the ellipsometer measurements is less than ±0.01. Thus the refractive-index increase in curve (A) of Fig. 2, at least, is unquestionably real.

According to ESR measurements[2] (Fig. 3), the pure and the Ge-doped silica glasses showed increases in the concentrations of Si E' and Ge E' centers, respectively, and the formation of peroxy radicals and nonbridging oxygen hole centers was observed in both glasses. The UV difference spectra (Fig. 4) are in agreement with these assignments[3–5] but also show a possible decrease in the concentration of neutral-oxygen monovacancies (NOMV's) in the Ge-doped glass.[6] In addition, unidentified broad absorption bands running from 320 to 700 nm are formed in both glasses.

The creation of defects in the high-silica glasses by 810-nm radiation suggests that the damage mechanism involves a multiphoton process. To our knowledge, such damage at this wavelength has not been reported, and it is suspected that the high pulse energy of the femtosecond laser is responsible for the effect. The photoinduced refractive-index increase in pure-silica glass also appears to be a new observation.

Traditionally it has been suggested that the refractive-index increase resulting from UV irradiation of Ge-doped silica glasses arises from the bleaching of the NOMV band at 5.06 eV (245 nm) owing to the reaction of these sites to form Ge E' centers.[7,8] More recently, however, it was shown that this bleaching mechanism can account for only a small portion of the refractive-index changes,[9] and a model based on densification and strain in the glass has been proposed.[10] The observed refractive-index increases

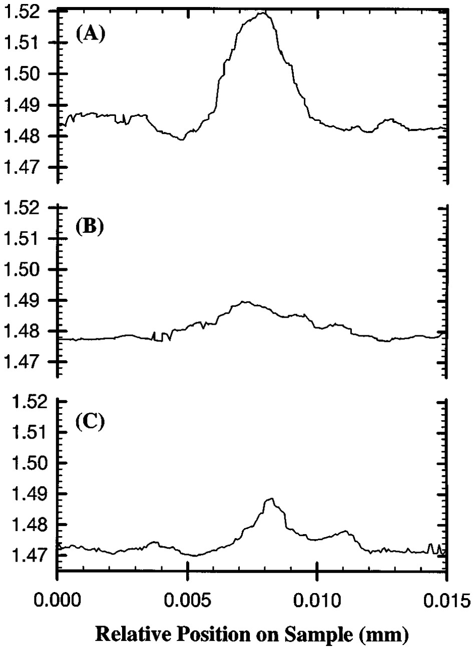

Fig. 2. Refractive-index mappings across the cross sections of damage lines created by translation of the samples perpendicular to the laser beam [see Fig. 1(C)] with a 10× lens, an average laser power of 470 mW, and a sample translation rate of 100 μm/s for (A) $3GeO_2$ $97SiO_2$ after 10 passes of the laser along an identical route, (B) $3GeO_2$ $97SiO_2$ after a single pass of the laser, and (C) pure silica after a single pass of the laser. The visible dimensions of the damage cross sections are approximately 6 μm × 180 μm for all the damage marks, so the outer limits of the mappings represent the refractive index of the surrounding glass.

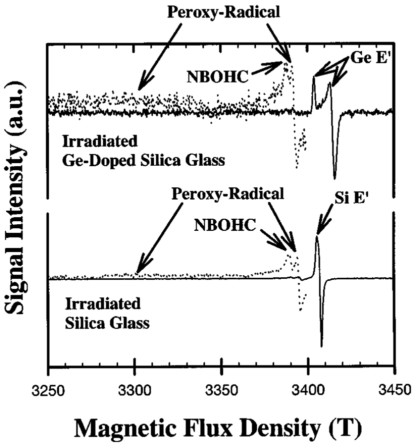

Fig. 3. ESR spectra of SiO_2 and $3GeO_2$ $97SiO_2$ glasses after laser irradiation. The solid and the dotted curves indicate ESR spectra collected with 1- and 50-mW microwave powers, respectively. The as-received pure-silica glass contained immeasurably small defect concentrations, and the as-received Ge-doped silica glass contained minute concentrations of ESR-detectable defects compared with the damaged glass. NBOHC's, Nonbridging oxygen hole centers.

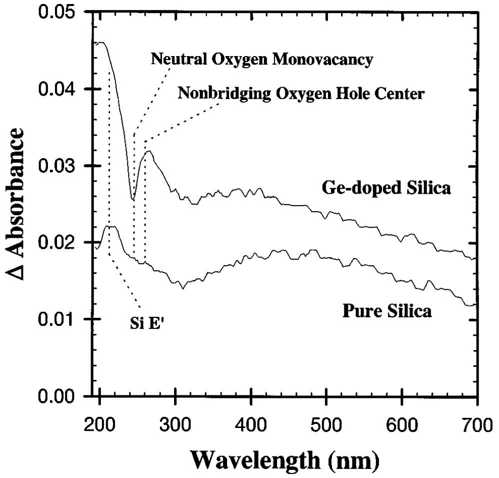

Fig. 4. UV difference spectra of the high-silica glasses before and after they were laser irradiated. The Ge-doped silica spectrum is vertically offset by +0.01 for clarity. The noise in these spectra is due to the damaged region, which comprises less than 5 vol. % of the glass probed by the spectrometer beam.

in both Ge-doped and pure-silica glasses support the idea that the changes are not caused primarily by the NOMV bleaching mechanism, as no NOMV's are found in the UV spectrum of the pure silica. From microscopic observation of the damage region during irradiation, the structural changes appear similar to what might be expected for localized melting, and it is suspected that the local structural rearrangement is significant. However, we do not yet have direct evidence of densification. It is also not known whether true melting and rapid quenching are occurring, whether the glass is experiencing a combination of defect formation and structural relaxation toward metastable equilibrium, or whether the structural changes are of a different nature. In addition, it is unclear what effect the unidentified absorption bands in Fig. 4 have on the refractive index.

The refractive-index increases observed here for the high-silica glasses are large enough for the creation of optical devices and waveguides in bulk glasses, and the ability to restrict the refractive-index changes to the region scanned by the focal point of the laser allows one to write complex, three-dimensional patterns in the glass. Writing damage lines perpendicular to the incident light beam provides the most flexibility for writing planar patterns and allows one to create multiple pattern layers by simply changing the focus depth of the beam. Although the lines written perpendicular to the laser are elliptical, it should be possible for one to produce cylindrical damage lines that can serve as light conduits by varying parameters such as the aperture of the microscope lens, the laser wavelength, and the sample thermal history.

The ability to create photoinduced refractive-index changes in glass by focused, femtosecond laser irradiation at sub-UV wavelengths has been demonstrated. This is important because it is easy to design optical systems that can manipulate this low-photon-energy light without causing laser damage except at the desired focal point of the beam. Moreover, it was possible to create visible but transparent laser damage without cracks in every glass examined. With improvements in the shape of the damage, damage lines formed by this process may one day be used as waveguides in optical circuits.

The assistance and advice of Junji Nishii of Osaka National Research Institute with regard to the ESR measurements is greatly appreciated. The silica glasses and the Ge-doped silica glass were kindly donated by Shin Etsu and Showa Electric Wire & Cable Co., Ltd., respectively.

References

1. K. O. Hill, Y. Fujii, D. C. Johnson, and B. S. Kawasaki, Appl. Phys. Lett. **32,** 647 (1978).
2. D. L. Griscom, in *Glass Science and Technology*, Vol. 4B of Advances in Structural Analysis, D. R. Uhlmann and N. J. Kreidl, eds. (Academic, New York, 1990), pp. 197–223.
3. R. A. Weeks and E. Sonder, in *Proceedings of the Symposium on Paramagnetic Resonance* (Academic, New York, 1963), Vol. 2, p. 869.
4. C. M. Nelson and R. A. Weeks, J. Am. Ceram. Soc. **43,** 396 (1960).
5. R. A. B. Devine, C. Fiori, and J. Robertson, in *Defects in Glasses*, F. L. Galeener, D. L. Griscom, and M. J. Weber, eds. (Materials Research Society, Pittsburgh, Pa., 1986), p. 177 .
6. R. Tohmon, H. Mizuno, Y. Ohki, K. Sasagane, K. Nagawawa, and Y. Hama, Phys. Rev. B **39,** 1337 (1989).
7. D. P. Hand and P. St. J. Russell, Opt. Lett. **15,** 102 (1990).
8. H. Hosono, Y. Abe, D. Kinser, R. Weeks, K. Muta, and K. Kawazoe, Phys. Rev. B **46,** 11445 (1992).
9. D. L. Williams, S. T. Davey, R. Kashyap, J. R. Armitage, and B. J. Ainslie, Appl. Phys. Lett. **59,** 762 (1991).
10. B. Poumellec, P. Niay, M. Douay, and J. F. Bayon, in *Photosensitivity and Quadratic Nonlinearity in Glass Waveguides: Fundamentals and Applications*, Vol. 22 of 1995 OSA Technical Digest Series (Optical Society of America, Washington, D.C., 1995), p. 112.