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Femtosecond laser-pulse-induced birefringence in optically isotropic glass

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We used a regeneratively amplified Ti:sapphire femtosecond laser to create optical birefringence in an isotropic glass medium. Between two crossed polarizers, regions modified by the femtosecond laser show bright transmission with respect to the dark background of the isotropic glass. This observation immediately suggests that these regions possess optical birefringence. The angular dependence of transmission through the laser-modified region is consistent with that of an optically birefringent material. Laser-induced birefringence is demonstrated in different glasses, including fused silica and borosilicate glass. Experimental results indicate that the optical axes of laser-induced birefringence can be controlled by the polarization direction of the femtosecond laser. The amount of laser-induced birefringence depends on the pulse energy level and number of accumulated pulses. © 2004 American Institute of Physics. [DOI: 10.1063/1.1707231]

I. INTRODUCTION

When an intense femtosecond pulsed laser is focused inside a bulk transparent material, the intensity in the local volume can become high enough to cause absorption through nonlinear processes, leading to optical breakdown or photo-damage. Slightly below the optical breakdown threshold, this nonlinear, multiphoton absorption process can locally alter the structure of bulk transparent material and cause a refractive index change.¹⁻⁵ In addition to the fundamental interest in the processes that lead to permanent refractive index change, there is great interest in using this approach to create three-dimensional waveguides and integrated optics in bulk glass. Recent work^{6,7} has indicated that when a femtosecond laser focuses into a bulk glass, optical anisotropy can be created. However, the effect of laser processing conditions and quantitative measurements on the amount of birefringence induced by the laser pulses have not been systematically studied and are the subject of our investigation. The capability of creating optical birefringence in an amorphous silicon dioxide material provides a pathway to control the polarization of light in silicon dioxide optical waveguides and will have a significant impact on integrated optics.

II. EXPERIMENTAL PROCEDURE

The laser used in this experiment was a regeneratively amplified Ti:sapphire femtosecond pulsed laser (Spectra-Physics, Hurricane). The femtosecond laser pulses had a 125-fs pulse width [full width at half maximum (FWHM)] and 1-kHz pulse repetition rate. The energy of the pulse could be varied by the combination of a half-wave plate and a thin-film polarizer. Further energy reduction was accomplished by the use of neutral-density filters to avoid photo-damage to the polished glass plates. The average laser power used in this investigation was between 450 and 900 μW , corresponding to a pulse energy between 0.45 and 0.9 μJ . The laser was focused to a 3- μm -diam spot inside the glass, using a long-working-distance microscope objective (Mitu-

toyo, 20 \times , NA=0.40). The calculated peak intensity of the femtosecond pulse was $9.8 \times 10^{20} \text{ W/m}^2$ for a pulse energy of 0.9 μJ .

In this investigation, a thermally treated chemical-vapor-deposited (CVD) amorphous silica plate (Quartz International, Albuquerque, NM) was used. The CVD amorphous glass was optically isotropic, and there was no birefringence observed before laser writing. The glass plate (polished on both surfaces) was mounted on a motorized x - y translation stage with linearly controlled motion along the z direction to adjust the focus of the laser inside the glass. The laser was focused approximately 600 μm below the surface of the glass plates. In addition to these linear motion controls, the sample could also be rotated in the x - y plane on a rotational stage. In the writing process, the polarization direction of the beam was fixed, and the sample was rotated with respect to the polarization direction of the laser beam.

After laser writing, the samples were evaluated using a transmission microscope (Olympus BH-2). Samples were illuminated using a monochromatic light filtered from a halogen lamp. The filter has a center wavelength of 671 nm and a bandwidth FWHM of 10 nm. The samples were put between two crossed polarizers on the rotating stage of the microscope. The angular rotation of the sample with respect to the polarizer was accurately measured by the stage goniometer. The transmitted light through the samples was captured by a charge-coupled-device (CCD) digital camera attached to the transmission microscope viewer. Optical transmission signals were evaluated using a National Instruments PCI-1424 digital image capture card. The CCD camera is operated in the linear regime. The maximum intensity of the transmitted light for the laser-modified region was determined by scanning the image array for the maximum value in each of ten images. The values were then averaged and recorded.

III. RESULTS AND DISCUSSION

Figure 1(a) shows a transmission image (through z or the laser beam direction) of femtosecond laser modified regions

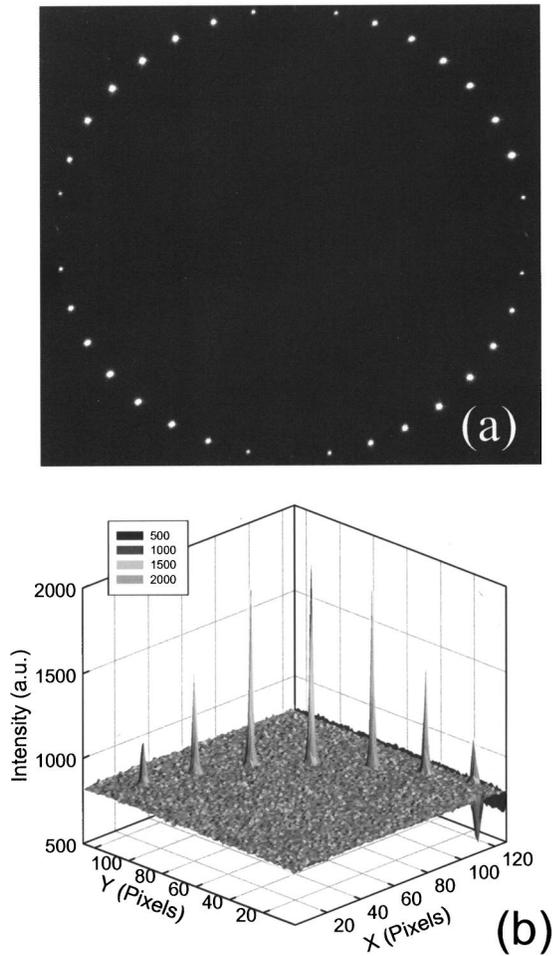


FIG. 1. (a) Transmission image of a laser-modified glass plate between two crossed polarizers, with the polarization direction of the writing laser rotated 10° between two adjacent spots, and (b) the transmitted light intensity in the first quarter.

in a CVD quartz plate between two crossed polarizers. Each modified region was exposed to 60 000 pulses (1 min dwell time) parallel to the beam direction with an average pulse energy of 0.9 μJ. The glass plate was rotated 10° after each laser irradiation. It was found that when the pulse energy was greater than 0.9 μJ, stress birefringence and even microcracks could be observed near laser-modified regions. Since the amorphous quartz plate was optically isotropic, the bulk glass was dark when the two polarizers were crossed. The laser-modified regions (shown by the ring of dots), however, were bright, indicating the creation of birefringence in these regions. Figure 1(b) illustrates the transmission variation of these modified spots in the first quarter of the ring structure. The results indicated that the intensity of transmission changed sinusoidally. It is well known that transmission through the second polarization analyzer depends on the amount of phase retardation $[\Delta\varphi = 2\pi(n_e - n_o)d/\lambda = 2\pi\Delta nd/\lambda]$, where d is the thickness of the birefringent medium and the orientation angle (θ) of the birefringent optical axes [see Eq. (1)]. Since the amount of phase retardation created by the femtosecond laser pulses remained the same for these spots, the sinusoidal change in the transmission from spot to spot indicated that the optical axes of the

birefringent medium was rotated by the polarization change of the writing laser. Since the polarization direction of the laser beam is rotated 10° from spot to spot, there should have been a direct correlation between the polarization direction of the writing laser and the orientation of optical axes of the laser-modified birefringent medium. This correlation was immediately confirmed when the transmitted light became extinct [see Fig. 1(a)] every 90°. These observations prove that the optical axis of the femtosecond laser-induced birefringence in bulk glass can be controlled by the polarization direction of the femtosecond writing laser.

For a transparent optically birefringent medium between two crossed polarizers, the optical transmission (T) is

$$T = \frac{1}{2} \sin^2(2\theta)[1 - \cos(\Delta nkd)], \quad (1)$$

where $\Delta n = n_e - n_o$, $k = 2\pi/\lambda$, and d is the thickness of the birefringent region through which the light travels. As a first-order approximation, we assumed that the laser-induced birefringence is proportional to the intensity of the light.⁸ Therefore, the induced birefringence along the z axis ($x, y = 0$) for a focused Gaussian beam is

$$\Delta n = \frac{\Delta n_0}{1 + \left(\frac{z}{z_0}\right)^2}, \quad (2)$$

where z_0 is the Rayleigh length and Δn_0 is the peak value of the birefringence.

The total optical path delay (or the total retardation Δ) for two polarizations is

$$\Delta nd = \int_{-\infty}^{\infty} \frac{\Delta n_0}{1 + \left(\frac{z}{z_0}\right)^2} dz = \pi \Delta n_0 z_0, \quad (3)$$

where $z_0 = \pi w_0^2/\lambda$, λ is the wavelength in glass, and w_0 is the waist of the Gaussian beam. Substituting Eq. (3) into Eq. (1), the transmission through the sample and the second polarization analyzer can be determined by

$$T = \frac{1}{2} \sin^2(2\theta)[1 - \cos(\pi \Delta n_0 k z_0)]. \quad (4)$$

Based on Eq. (4), the transmission through a birefringent medium between two crossed polarizers depends on the value of the birefringence, the wavelength of the probing light, the thickness of the birefringent material, and the angle between the ordinary direction of the birefringent material and the polarization axis of the first polarizer. Figure 2 illustrates the variation of transmission as a function of sample rotation angle between two crossed polarizers. The open and solid circles represent the changes of transmitted light intensity for regions exposed to the 0.68- and 0.90-μJ pulses for 10 min under white-light illumination, respectively. The dotted and solid lines are the curve fitting results based on Eq. (4). The results indicate that the laser-induced birefringence is greater for the region exposed to the higher-energy pulses than for the region exposed to the lower-energy pulses. The average intensity and standard deviation from four measurements collected for monochromatic light (at 671 nm) are

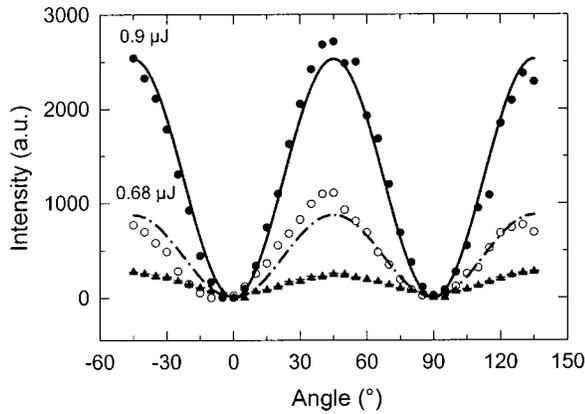


FIG. 2. Variation of the transmitted light intensity as a function of glass orientation.

indicated by the triangles. The measurement was performed on a region where the glass was exposed to a 0.9- μJ pulse for 4 min. The introduction of the bandpass filter significantly reduced the total light intensity through the bottom polarizer and resulted in a lower intensity variation. However, the general behavior was consistent with the predicted behavior of an optically birefringent medium. Similar results are also observed in fused silica and borosilicate glass.

Direct observation using an optical microscope under two crossed polarizers gave the spot size of the laser-modified region in the bulk glass. The diameter of the laser-modified spot was about 3 μm , and the average thickness of these birefringence regions was about 25 μm . The calculated thickness of the modified regions, estimated by twice of the Rayleigh length ($2z_0 = 2\pi w_0^2/\lambda_i$, $\lambda_i = 548.7$ nm in amorphous quartz) was about 25.8 μm , which is consistent with experimental observation. Furthermore, it was found that perpendicular to the propagation direction of the writing laser, these modified regions also showed the extinction behavior between two crossed polarizers for every 90° of rotation, suggesting that the laser-modified regions possessed a biaxial optical birefringence.

From Eq. (4), the laser-induced birefringence (the peak at the center of the spot) can be obtained by measuring the transmission at the condition of $\theta = 45^\circ$ with the assumption that the laser-modified birefringent region has a fixed depth of twice the Rayleigh length (25.8 μm). Figure 3 shows the maximum change of birefringence (Δn_0) induced by the laser as a function of the number of laser pulses. The results indicate that the maximum change of birefringence increased rapidly at the beginning and then began to level off as the number of laser pulses increased. This phenomenon is similar to that observed in an experiment using a linearly polarized excimer laser source.⁶ However, under excimer laser irradiation, the laser energy is linearly absorbed only near the surface of the glass, while under femtosecond laser irradiation, the photon energy can only be absorbed through a nonlinear process where a threshold intensity is required for multiphoton absorption.

Although the fundamental mechanisms that create the birefringence in the bulk glass by femtosecond laser irradiation are not yet clear, several observations in this investiga-

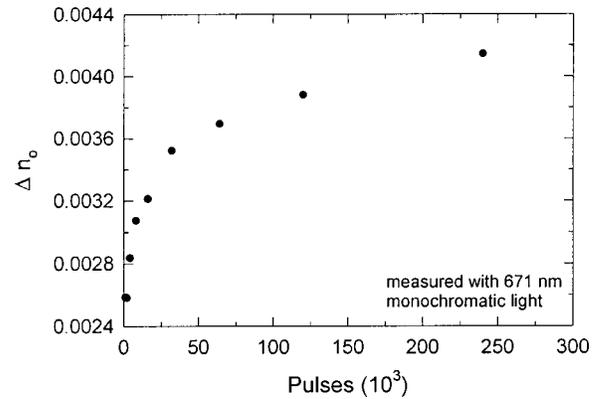


FIG. 3. Change of laser-induced birefringence in bulk silica as a function of the number of accumulated laser pulses.

tion have helped to shed light on this issue. Our results show two interesting phenomena: (1) the high-energy linearly polarized laser pulse can induce birefringence in optically isotropic glasses and (2) the optical axes of the birefringence depend on the polarization direction of the laser beam. The dependence of birefringence on polarization suggests that the optical birefringence cannot be solely attributed to a thermal effect generated by multiphoton absorption or plasma formation. If a thermal-heating effect is a major contributor, the laser-modified region should be optically isotropic. Some investigators¹⁻⁵ have attributed this increase in refractive index to the local densification of glass by the femtosecond pulses. It is well known that glasses can be poled under ultraviolet light^{6,9} or a high-dc-bias condition at high temperature¹⁰ to create nonlinear optical behavior. Furthermore, a recent investigation¹¹ on the local change in atomic structure resulting from *in situ* laser irradiation has demonstrated a polarization-dependent structure change in amorphous chalcogenide glasses. These field- or polarization-dependent mechanisms are especially applicable to femtosecond laser processing since the estimated electric field generated by the femtosecond pulse is on the order of 10^{11} V/m. Details of the mechanisms involved in the creation of birefringence in isotropic glass media and the fundamental optical properties of the laser-modified region will be investigated in future work.

IV. SUMMARY

In conclusion, we have created optically birefringent regions within transparent, isotropic glass materials by using a high-energy, linearly polarized femtosecond laser. The orientation of optical birefringence is controlled by the polarization direction of the femtosecond laser beam. The maximum laser-induced optical birefringence in amorphous silica is 0.004. Experimental observations indicate that the laser-modified regions possess a biaxial optical birefringence. The laser-induced optical birefringence depends on the power level, accumulated pulses, and polarization direction of the femtosecond laser.

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