Breaking Symmetry in Glass by Femtosecond Laser Irradiation

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Writing by femtosecond laser from left to right observed can be different from writing in the opposite direction. Structures with broken mirror symmetry in chalcogenide glass are discovered. Chiral patterns are demonstrated.

Recently direct writing in transparent materials by ultrashort light pulses has attracted considerable interest due to new applications and phenomena ranging from 3D optical waveguides [1] and micro-explosions [2] to photonic crystals [3] and 3D self-organized sub-wavelength structures [4]. It has been shown that self-assembled nanostructures are responsible for the negative birefringence of the regions processed by femtosecond laser pulses above a certain intensity threshold [5]. Self-organized nanostructures and related form birefringence with optical axes depending on the polarization of the writing laser have been observed in a limited number of materials including fused silica glass and sol-gel silica glass. In an attempt to clarify the reason of nonappearance of 3D self-assembled nano-structures in other optical materials we carried out experiments in chalcogenide glasses. Here we report the observation of a polarization rotation by imprinted structures, which is different from a polarization rotation caused by form or stress-induced birefringence. Moreover for the first time to the best of our knowledge it has been observed that the information on the direction of writing could be recorded inside transparent materials. The results give the first experimental evidence of structures with broken mirror symmetry by femtosecond laser irradiation.

In our experiments we used germanium sulfide glass, which is a promising material for nonlinear optical and optical amplifier applications [6]. The laser radiation from a regeneratively amplified mode-locked Ti: Sapphire laser (140 fs pulse duration, 250 kHz repetition rate) operating at a wavelength of 800 nm was focused via 50x (NA=0.55) microscope objective into the chalcogenide glass samples mounted on a 3D linear-motor translation stage. The speed of writing was 200 μm/s and the pulse energies were 0.6 - 0.8 μJ.

A series of lines was written in the glass sample about 200 μm below the surface by a focused beam. The sample was translated perpendicular to the beam in two opposite directions - from left to right and vise versa. After irradiation the sample was inspected under optical microscope using parallel and crossed polarizers.

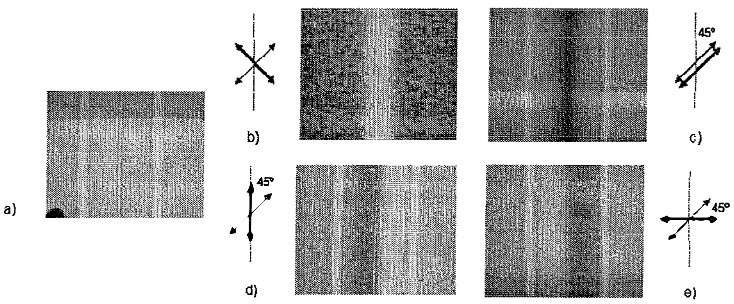

Fig.1 Microscope images of modified regions: without polarizers (a), polarizers crossed 90^0 (b), parallel (c), 45^0 (d) and -45^0 (e).

The width of the written structures was about 30 μm, which is significantly larger than the size of the focused beam of 1 μm (Fig. 1a). This indicates that thermal effects could be responsible for the material modification in the areas surrounding the irradiated regions. Imaging the structures in transmitted light using cross polarizers clearly shows the change in the state of polarization of the light passed through the modified regions (Fig. 1b). Imaging in parallel polarizers revealed the minimum in transmission, which was consistent with the image in cross polarizers (Fig. 1c). However we were surprised to observe the asymmetry between left and right parts of the image, when one of the polarizers was oriented 45^0 relative to the written stripe and another one was parallel to the stripe (Fig 1d, e). Moreover we observed a different left-right asymmetry for the structures written in two opposite directions and which were the mirror images of each other (Fig. 2 a, b).

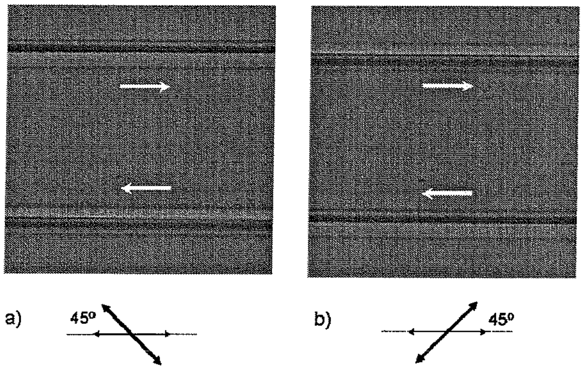

Fig. 2 Modified regions written in two opposite directions and imaged using polarizers crossed 45^0 (a) and -45^0 (b).

The observed left-right asymmetry indicates that the polarization of light, passing through the modified region, rotates in the opposite directions on two sides from the center of the region.

Imaging the cross-section of the structures in cross-polarizers did not reveal the asymmetry in the image and any significant polarization change in the imprinted structures apart from a small change in a broad area surrounding the modified regions, which is typical for a stress-induced birefringence. This indicates that the birefringence is not responsible for the polarization change in the modified region.

A possible explanation of the phenomenon is that polarization rotation occurs due to reflection from the boundaries of the modified region. Polarization rotation in different directions upon reflection could indicate the presence of chiral structures in the modified regions.

A series of circles were written by rotating the sample clockwise and anti-clockwise. Imaging in cross polarizers revealed the presence of chiral patterns (Fig.3).

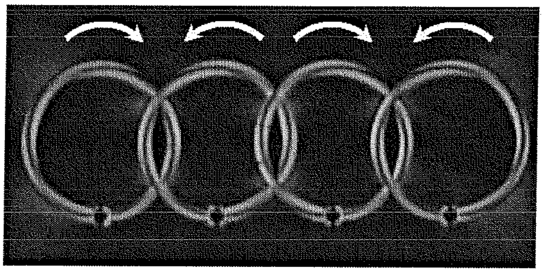

Fig. 3 Images of written circulars in crossed polarizers: evidence of chiral patterns.

References

[1] K.M. Davis, K. Miura, N. Sugimoto, and K. Hirao, "Writing waveguides in glass with a femtosecond laser," *Opt. Lett.*, **21**, 1729 (1996).

[2[E.N. Glezer and E. Mazur, "Ultrafast-laser driven micro-explosions in transparent materials," *Appl. Phys. Lett.* **71**, 882 (1997).

[3] H. Sun, S. Juodkazis, M. Watanabe, S. Matsuo, H. Misawa, and J. Nishii, "Arbitrary-lattice photonic crystals created by multiphoton microfabrication," *J. Phys. Chem.* **B** 104, 3450 (2000).

[4] Y. Shimotsuma, P.G. Kazansky, J. Qiu and K. Hirao, "Nanostructuring of transparent materials by ultrashort light pulses," Phys. Rev. Lett. **91**, 247405 (2003)

[5] E. Bricchi, B. Klappauf and P.G. Kazansky, "Form birefringence and negative index change created by fs direct writing in transparent materials", Opt.Lett **29**, 119 (2004).

[6] H. Takebe, R. Kitagawa and D. Hewak, "Non-Toxic Sulfide Glasses and Thin Films for Optical Applications", J. Ceram. Soc. Jpn. **113**, 37 (2005)