Embedded Anisotropic Micro-reflectors by Femtosecond-Laser Nanomachining

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Abstract

Directly-written embedded structures created within fused silica by a femtosecond Ti:Sapphire laser are observed to strongly reflect blue light. Reflection emerges only in a direction parallel to the polarization axis of the writing laser. This anisotropic effect is caused by a periodic modulation of refractive index of amplitude $\Delta n \sim 10^{-2}$ with a characteristic period $\Lambda \sim 150$ nm over a spot size ~1.5 μm. We show that the origin of the anisotropic reflection is the primary cause of other anisotropic phenomena reported in recent experiments.

The use of a femtosecond laser source to directly write structures deep within transparent media has recently attracted much attention due to its simplicity compared to lithographic methods, and its capability for writing in three-dimensions [1-3]. Tight focusing of the laser into the bulk of material causes non-linear absorption only within the focal volume, depositing energy that induces a permanent material modification [4,5]. A variety of photonic devices have already been created by translating a sample through the focus of a femtosecond laser [6,7]. Although molecular defects caused by such intense irradiation have been identified in fluorescence, ESR and other studies [8], the mechanism of induced modifications in glass is still not fully understood. Moreover, such structures in Ge-doped silica [9] and other glass materials [10] show an unexpected anisotropic light scattering which is dependent on the plane of light polarization. This has been interpreted in terms of photoelectrons moving along the direction of light polarization inducing index inhomogenities. More recently, uniaxial birefringence imprinted in structures written within fused silica plates has been observed but the origin of this anisotropic phenomenon remains a mystery [11]. In this letter, we will describe a further anisotropic property observed in silica after being irradiated by a femtosecond laser – strong reflection from the modified region *occurring only along the direction of polarization of the writing laser*. Our analysis suggests that this effect is also the primary cause of all previously reported anisotropic phenomena.

A regeneratively-amplified mode-locked Ti:Sapphire laser (150 fs pulse duration, 250 kHz repetition rate) operating at λ=850 nm was used to directly write microstructures into a fused silica plate (40x40x1mm). In the experimental arrangement (Figure 1), the collimated laser passes through an electronic shutter, variable neutral density filter and half-wave plate before a dichroic mirror reflecting only in the 400-700 nm region. The infrared laser light travels through the mirror and is focused through a 50x (NA=0.55) objective into the bulk of the sample, down to a beam waist diameter estimated to be ~1.5 μm. The silica sample is mounted on a computer-controlled linear-motor 3D translation stage (of 20 nm resolution). To simultaneously observe the writing process, a CCD camera with suitable filters and white light source is used.

A range of embedded diffraction gratings with overall dimensions 700 μm x 700 μm, each consisting of 100 rulings with 7 μm period were directly written towards the edges of the plate at a depth of 0.5 mm below the front surface. In every case, the speed of writing was 200 μm/s and each grating ruling had only one pass of the laser. Pairs of embedded gratings were created with orthogonal writing polarizations directed parallel and perpendicular to the grating rulings respectively, with average fluence ranging from 270 mW (~1.1 μJ/pulse) down to 26 mW (~0.1 μJ/pulse). Additionally, pairs of embedded single lines of length 1mm were written by the same method. Finally, a regular array of 40x40 'dots' with a pitch of 10 μm was directly written into the corner of the plate. Each 'dot' was produced by holding the sample translation stage stationary at each writing point, and irradiating for 3 ms (~750 pulses) using the electronic shutter.

After writing, the samples were viewed through the silica plate's polished edges using a 200x microscope incorporating a color CCD camera. During inspection, the structures were illuminated with a randomly-polarized white light source in a direction along the viewing axis, either from below the structure (opposite side to microscope objective), or above the structure (through the microscope objective). The embedded structures were examined through the edge nearest to them, and the array of dots was examined in two orthogonal directions. A striking reflection was observed in the blue spectral region from a number of the structures when illuminated via the viewing objective. Closer analysis revealed that the reflection *only* occurred when the viewing axis was both parallel to the electric field vector of the writing beam and the structure was written with pulse energy greater than ~0.5 μJ. This indicates that the observed reflectivity is both fluence dependent and highly anisotropic. Fluorescence cannot account for the observation due to the directional dependence. Figure 2 shows a schematic of the reflection phenomenon. As can be seen, the macroscopic shape of the photonic structures does not determine the direction of the anisotropic reflection.

Figure 3 shows microscope images of the reflection from several directly-written embedded structures. The illuminating light in all cases was incident above the samples through the viewing objective and set to a level that ensured that the weak-contrast microstructure itself was not imaged. The spatial position of the embedded objects relative to the focus of the microscope objective was checked beforehand by illuminating from below, when the embedded structure in all cases could be clearly observed. The displayed images were chosen from regions of modification created with a pulse energy of ~0.9 μJ. In each example the orientation of the direct-write laser's electric-field is indicated, while the $\boldsymbol{k}_w$-vector marks the incident direction of the writing laser beam. Figure 3(a) displays the reflection from a single line which has dimensions ~1.5 μm into the page due to the focal width of the beam, and ~30 μm down the page due to the beam's confocal parameter, enhanced by self-focusing effects [5]. Figure 3(b) shows the reflection from the side of a 100-line grating, with its rulings going into the depth of the page. Not all of the 100 lines of the grating contribute to the recorded reflection because of the ~2 μm focal depth of the imaging objective. Nevertheless, the reflection is considerably enhanced compared to the single line in Figure 3(a). Figure 3(c) shows the result of imaging an identical structure to Figure 3(b), except written with orthogonal polarization. From this orientation, no reflecting structure at all can be observed. Figure 3(d) shows a section of the 40x40 array of 'dots' described above, once again producing strong reflection along the writing beam polarization axis. These particular structures are interesting because they are approximately circular with a diameter of ~1.5 μm when viewed from the direction of the writing laser, and therefore have a uniform cross section. However when viewed from a direction orthogonal to the axis of the writing beam's polarization, there is no reflecting component as Figure 3(e) clearly demonstrates. The reflected light observed from the structure shown in Figure 3(b) is analyzed yielding the spectrum displayed in Figure 4. This data shows a strong peak at 460 nm, which accounts for the blue color when observed under a microscope.

We suggest that the anisotropic reflectivity can only be explained as a consequence of Bragg reflection from a self-organized periodic structure. Indeed, a modulation in refractive index of period Λ~150nm, produced only along the direction of the incident laser's electric field can account for the observed anisotropic reflection at λ~460nm ($\Lambda = \lambda/2n$). Such a grating does not reflect when viewed edge on. The magnified section at the bottom right-hand corner of Figure 2 demonstrates how the periodic nanostructuring is arranged in the case of a single 1.5μm diameter 'dot'. The orientation and the period of the nanostructure are almost identical to the periodic nanostructure implicated in the phenomenon of anisotropic light scattering [9]. Closer inspection of Figure 4 shows an additional smaller peak at 835nm. This suggests that an extra grating component may be formed which has double the periodicity of the laser-induced structures. Alternatively, this long-wavelength reflection could be a second-order diffraction peak of a grating component with period equal to the wavelength of the incident light [12]. Surface ripples with a period equal to the wavelength of incident laser radiation and that are likewise aligned in a direction orthogonal to the electric field have been observed in experiments involving laser deposition [12]. Nevertheless, we believe that our data is the first reported evidence of periodic nanostructures (much smaller than the wavelength of incident light) being generated within the bulk of a material. We speculate they arise from a mechanism associated with the creation of a hot electron plasma by multiphoton absorption of incident light. Anisotropic index inhomogeneities are then induced by electrons moving along the direction of light polarization [9]. Self-organized nanostructures are in turn produced by a pattern of interference between the incident laser radiation and a plasmon-polariton wave generated within the sample. Positive feedback leads to exponential growth of the periodic nanostructures in the plane of light polarization, which become frozen within the material.

Further microscope characterization was carried out on the same samples by positioning the fused silica plate between crossed polarizers and illuminating from below along the direction of the original writing beam. With the polarizers oriented at angles of $\pm\pi/4$ to the writing-beam electric-field vector, this enables regions of birefringence to be identified. The onset of birefringence occurs at a writing-fluence level ~0.5 μJ/pulse, equal to that found in the case of the anisotropic reflection. This strongly suggests that the mechanism responsible for inducing reflection along the writing beam polarization axis is the same mechanism that causes birefringence in the orthogonal direction. The femtosecond-laser-induced birefringence, which up to now has not been well understood, is therefore likely to be caused by the laterally-oriented small-period grating structures. Birefringence of this nature is well known as 'form' birefringence [13].

Reflectivity measurements made from the side of a macroscopic grating as shown in Figure 3(b) give a value of ~1% per 1.5μm-long micro-grating. Given that each micro-grating contains 10 periods of refractive-index modulation across its 1.5μm width, the maximum reflectivity,

$$R_{\max} = \tanh^2\left(\frac{\pi\delta n L}{\lambda_{reflection}}\right)$$

where L is the length of the reflecting region and $\lambda_{reflection}$ is 460nm, implies that the amplitude of index variation, δn ~0.01. This also suggests that it is possible to achieve much higher reflectivities by increasing the depth of the writing laser irradiated spot.

In conclusion, we have directly written sub-micron photonic structures into the internal bulk of fused silica with a femtosecond laser system. The nanostructures are observed to strongly reflect in the blue spectral region but only along the polarization axis of the original writing beam. We show this can arise from a self-organized periodic refractive index modulation. The effect can explain systematically the origin of other observed anisotropic behavior reported with such pulsed laser patterning. The anisotropic micro-reflectors described here should be useful in many monolithic photonic devices and can be harnessed for information storage, MEMS applications or quasi-phase matching where nanoscale periodic structuring is required.

Figure Captions

Figure 1. Experimental setup for short-pulse direct-writing of embedded structures.

Figure 2. Schematic showing the anisotropic reflection from embedded photonic structures. Reflection only occurs for incident light parallel to the electric-field vector of the incident writing laser. The magnified region (bottom) illustrates the laser-induced self-organized periodic nanostructuring responsible.

Figure 3. CCD camera images of the reflection from different embedded structures directly-written with the laser (incident direction $\boldsymbol{k}_w$, polarization orientation $\boldsymbol{E}$). (a) Reflection from a single line, (b) reflection from 100 lines one behind another into the page, (c) identical structure to (b) showing no reflection. (d, e) Array of 'dots' which show reflectivity dependent only on the writing polarization orientation.

Figure 4. Spectrum of the reflected light from the structure shown in Figure 3(b).

References

[1] K.M. Davis, K. Miura, N. Sugimoto, and K. Hirao, Opt. Lett. **21**, 1729 (1996).

[2] K. Miura, J.Qiu, H. Inouye, T. Mitsuyu, and K. Hirao, Appl. Phys. Lett. **71**, 3329 (1997).

[3] E.N. Glezer and E. Mazur, Appl. Phys. Lett. **71**, 882 (1997).

[4] M.D. Perry, B.C. Stuart, P.S Banks, D. Feit, V. Yanovsky, and A.M. Rubenchick, J. Appl. Phys. **85**, 6803 (1999).

[5] K. Yamada, T. Toma, W. Watanabe, K. Itoh, and J. Nishii, Opt. Lett. **26**, 19 (2001).

[6] Y. Kondo, K. Nouchi, T. Mitsuyu, M. Watanabe, P.G. Kazansky, and K. Hirao, Opt. Lett. **24**, 646 (1999).

[7] D. Homoelle, S. Wielandy, A. L. Gaeta, N.F. Borrelli, and C. Smith, Opt. Lett. **24**, 1311 (1999).

[8] H. Sun, S. Juodkazis, M. Watanabe, S. Matsuo, H. Misawa, J. Nishii, J. Phys. Chem. B 104, 3450 (2000).

[9] P.G. Kazansky, H. Inouye, T. Mitsuyu, K. Miura, J. Qiu, K. Hirao, and F. Starrost, Phys. Rev. Lett. **82**, 2199 (1999).

[10] J. Qiu, P. G. Kazansky, J. Si, K. Miura, T. Mitsuyu, K. Hirao, and A. Gaeta, Appl. Phys. Lett. **77**, 1940 (2000).

[11] L. Sudrie, M. Franko, B. Prade, A. Mysyrowicz, Opt. Commun. **171**, 279 (1999).

[12] S.R.J. Brueck and D.J. Ehrlich, Phys. Rev. Lett. **48**, 1678 (1982).

[13] M. Born and E. Wolf, *Principles of Optics* (Cambridge University Press, UK, 1999), p. 837.