

Nanostructuring of Transparent Materials by Ultrashort Light Pulses

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The study of the smallest (20 nm width) and the strongest (-0.2 index change) embedded periodic structures ever created by light in transparent materials are reported. These are the first gratings created by light-matter (electron plasma) interference.

Introduction

The ability to control light and properties of matter in small space regions and over short time intervals becomes more important as performances of electronic and optical devices are pushed to the limits. In this respect the use of femtosecond lasers to directly write photonic structures deep within transparent media has recently attracted much attention due to its capability for precise microfabrication in three-dimensions¹.

Recent observations of anisotropic light scattering and reflection from the regions modified by intense femtosecond light in the direction parallel to the polarization of writing laser, have given the evidence of sub-wavelength index gratings imprinted in irradiated materials²⁻³. Form birefringence induced by self-organized sub-wavelength index gratings has been also proposed⁴ to explain a puzzling phenomenon of uniaxial birefringence of structures written within fused silica plates⁵. Recently we reported direct proof of the existence of such gratings⁶. Surface gratings with a period equal to the wavelength of incident light have been observed in many experiments involving laser deposition and laser ablation⁷. The results of our work give the first evidence self-organized structures within the bulk of

material. Here we report recent results of the study of the self-organized nanostructures in transparent materials.

Results and conclusion

In our experiments we used commercially available silica glass. The laser radiation in Gaussian mode produced by regenerative amplified mode-locked Ti:Sapphire laser (150 fs pulse duration, 200 kHz repetition rate) operating at a wavelength of 800 nm was focused via 100x (NA=0.95) microscope objective into the silica glass samples placed on the piezo-translation stage.

After laser irradiation the sample was polished to the depth of the beam waist location. The surface of the polished sample was analyzed by a scanning electron microscope and Auger electron spectroscopy. The backscattering electron images reveal a periodic structure of stripe-like dark regions with low density of material these are ~ 20 nm width which are aligned perpendicular to the writing laser polarization direction. The Auger spectra mapping indicates that the oxygen defects (SiO_{2-x}) are periodically distributed in the focal spot of the irradiated region ($x \sim 0.4$) and the intensity of the silicon signal is the same for the whole imaged region (Fig.1).

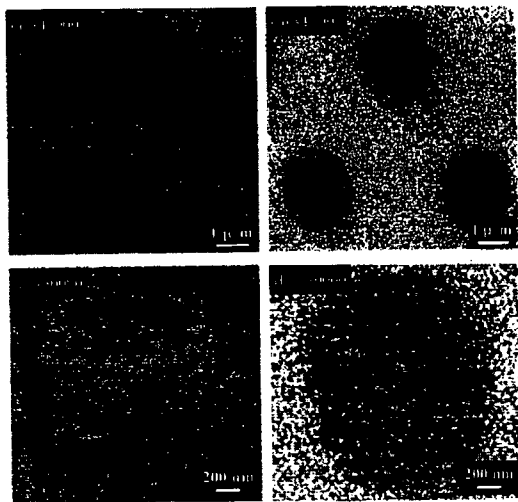


Fig.1 (a) Secondary electron images of silica glass surface polished close to the depth of focal spot. (b) Light "fingerprints": Backscattering electron images of the same surface. The magnification of the upper and lower images is $\times 10000$ and $\times 30000$ respectively.

The gratings periods range from 140 nm to 320 nm depending on pulse energies and number of light pulses.

The phenomenon is explained by the interference between the light wave and the bulk electron plasma wave (Langmuir wave), launched as a result of Cherenkov type of generation, which leads to modulation of the electron plasma concentration and subsequent material modifications. The theoretical estimates give high electron concentrations of $1.7 \times 10^{21} \text{ cm}^{-3}$ and high electron temperatures of $2 \times 10^6 \text{ K}$ for the interpretation of the experimental results. The plasma electrons are created in the process of breaking of Si-O-Si bonds via multi-photon absorption of light which is accompanied by the generation of non-bridging oxygen-hole centers (NBOHC, $\equiv \text{Si-O}^\cdot$) and interstitial oxygen atoms (O_i). Negatively charged oxygen ions can be repelled from the regions of high electron concentration.

The theory of form birefringency which takes into account the measured

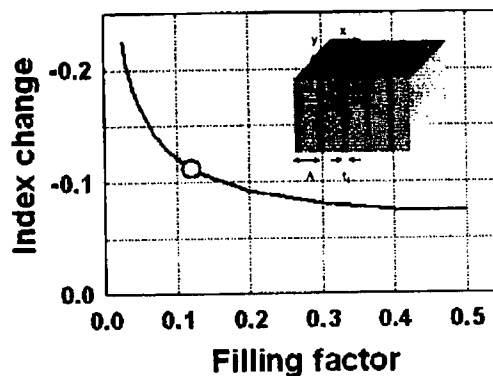


Fig.2 Index change in oxygen deficient layer versus filling factor t_1/Λ . Average index changes of birefringent structure are $n_{xx} = 0.005$ and $n_{xy} = -0.002$. Experimental conditions are indicated by circle.

values of the refractive index change and the dimensions of the periodic structures gives the negative index change in the

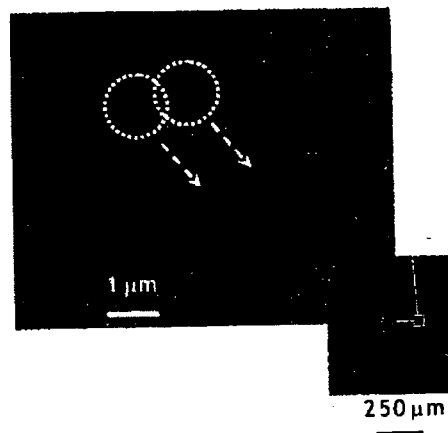


Fig. 3 Image from scanning electron microscope in back scattering configuration of a portion of a Fresnel zone plates (experimental details in reference 8). The processed zone was written translating the sample along concentric circular paths of $1.5 \mu\text{m}$ widths. The picture clearly shows that the nano-gratings written during two adjacent scans are spatially coherent.

oxygen depleted regions as high as 0.2 (Fig. 2). Furthermore, successful imaging of nano-gratings produced in previously reported structures⁸ (Fig. 3), shows the high spatial coherence of the nanograting demonstrating its self-organization. The observed nano-structures are the smallest and strongest embedded periodic structures ever created by light, which is a universal phenomenon in transparent materials and could be used for optical recording and photonic crystal fabrication.

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