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Nanostructuring of Transparent Materials by Light

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The smallest embedded structures ever created by light are observed in the experiments on femtosecond direct writing. The phenomenon is interpreted in terms of interference between the incident light field and the electric field of *bulk* electron acoustic wave, resulting in periodic structural changes in glass.

Progress in high power ultra-short pulse lasers has opened new frontiers in physics and technology of light-matter interactions from coherent x-ray generation to 3D direct writing of photonic structures [1-4]. A critical advantage of using femtosecond pulses relative to longer pulses for optical writing and data storage is that such pulses can rapidly and precisely deposit energy in solids [5, 6]. However, fabrication of sub-wavelength structures by light still is a challenging problem. Surface gratings with a period equal to the wavelength of incident light have been observed in many experiments involving laser deposition and laser ablation [7]. Here we report the first observation of periodic structures *within the bulk of a transparent material* written by a single femtosecond infrared laser beam. The structures in silica glass consist of oxygen depleted regions of 20 nm size with periods as small as 140 nm. *These are the smallest embedded structures ever created by light.*

Although molecular defects caused by intense femtosecond irradiation have been identified in fluorescence, ESR and other studies, the mechanism of induced material modifications is still not fully understood [8, 9]. Recent observations of anisotropic light scattering and reflection from the regions modified by intense femtosecond light pulses have given the evidence of sub-wavelength index gratings imprinted in irradiated materials [10, 11]. Form birefringence induced by self-organized sub-wavelength index gratings has been also proposed to explain a puzzling phenomenon of uniaxial birefringence of structures written within fused silica plates [11, 12]. However, until now there has been no direct proof of existence of such gratings.

In our experiments we used commercially available synthetic silica glass samples. The laser radiation 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.

After laser irradiation the sample was polished to the depth of the beam waist location. The surface of the polished sample was analyzed by scanning electron microscope and Auger electron spectroscopy. Secondary electron (SE) images and backscattering electron (BE) images of the same surface were compared (Fig. 1). The SE images of the polished silica sample indicate that the morphology of an irradiated sample in the examined cross-section does not change, namely, a void does not exist. On the other hand, the BE images reveal a periodic structure of stripe-like dark regions with low density of material and of ~ 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}, x ~ 0.4) are periodically distributed in the focal spot of the irradiated region.

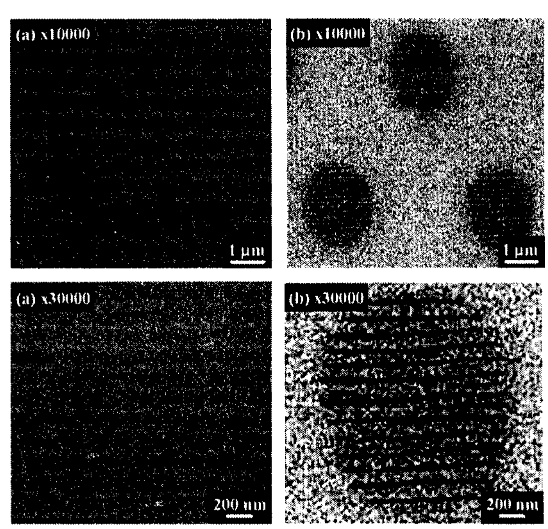

Fig.1 (a) Secondary electron images of silica glass surface polished close to the depth of focal spot. (b) Light "fingerprints": Backscattering electron images of the same surface. The magnification of the upper and lower images is x10000 and x30000 respectively.

The grating periods were about 240 nm, 180 nm and 140 nm for the number of light pulses of 5 x 10^4, 20 x 10^4 and 80 x 10^4 respectively and for the pulse energy of 1 μJ. Grating periods of 180 nm, 240 nm and 320 nm were measured at pulse energies of 1 μJ, 2 μJ and 2.8 μJ respectively and for the number of light pulses of 20 x 10^4 (Fig. 2a).

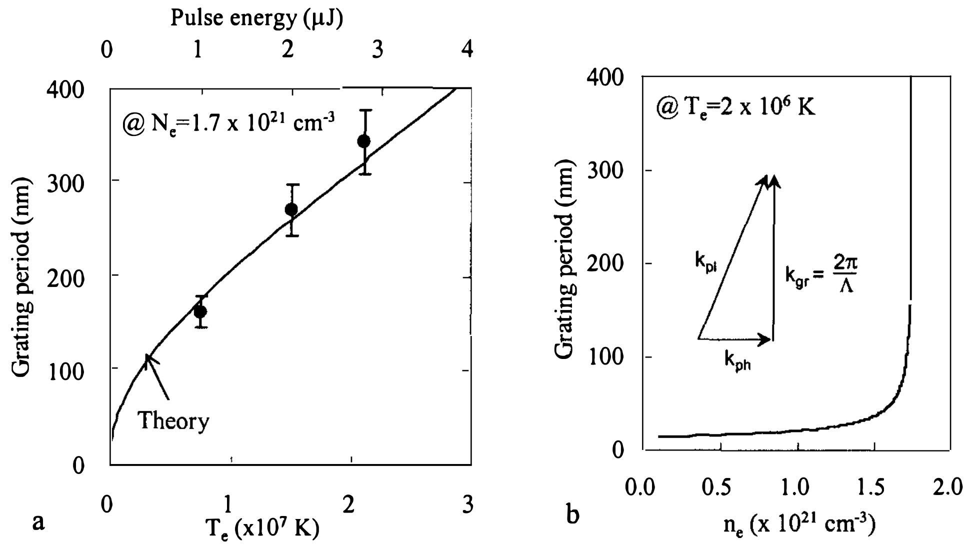

Fig.2 Theoretical dependence of self-organized grating period on electron temperature for n_e = 1.7 x 10^{21} cm^{-3} (a) and electron concentration for T_e = 2 x 10^6 K (b). Experimental grating periods versus pulse energy are also shown (a). Insert is a wave vector phase-matching diagram (b).

The following explanation of the observed phenomenon is proposed. The light absorption in the electron plasma will excite bulk electron acoustic waves. These are longitudinal waves with the electric field component parallel to the direction of propagation. The coupling is increased by a periodic structure created via a pattern of interference between the incident light field and the electric field of the bulk electron acoustic wave, resulting in the periodic modulation of the electron plasma concentration and the structural changes in glass. The electron acoustic wave is generated only in the plane of light polarization and only in the direction defined by conservation of longitudinal component of the momentum (insert Fig. 2b). The latter condition is similar to the condition in Cherenkov's mechanism of nonlinear wave generation. Taking into account the energy conservation condition, the momentum conservation relation and the dispersion relation for Langmuir waves it is possible to calculate the dependences of the grating period on the electron temperature and the electron plasma density (Fig. 2a, b). Assuming that the electron temperature is proportional to the pulse energy due to one-photon absorption of light by the electron plasma, the experimental dependence of the grating period on the pulse energy is in very good agreement with the theoretical prediction (Fig. 2a). The interference between the light wave and the electron acoustic wave leads to modulation of the electron plasma concentration and the oxygen concentration.

Apart from the fundamental importance of the observed phenomenon as the first evidence of interference between light and electron acoustic waves, the observed light "fingerprints" are the smallest embedded structures ever created by light. The reported light-induced nanostructuring is a universal phenomenon in transparent materials and could be useful for optical recording and photonic crystal fabrication.

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