Point-by-point inscription of 250 nm period structure in bulk fused silica by tightly focused femtosecond UV pulses

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Abstract

By conducting point-by-point inscription in a continuously moving slab of pure fused silica at the optimal depth (170 μm depth below the surface), we have fabricated a 250 nm period nanostructure with 30 nJ, 300 fs, 1 kHz pulses from a frequency-tripled Ti:sapphire laser. This is the smallest value for the inscribed period yet reported, and has been achieved with radical improvement in the quality of the inscribed nanostructures in comparison with previous reports.

Keywords: femtosecond UV laser, microfabrication, fused silica, nanostructure, differential interference contrast microscopy, atomic-force microscopy

1. Introduction

In 2004, two groups independently reported the application of a point-by-point (PbP) technique to fibre Bragg grating (FBG) inscription in standard non-photosensitive fibres [1–3]. Both groups used tightly focused femtosecond (fs) IR laser (λ = 800 nm) radiation and a sub-micron precision positioning system. The FBGs produced exhibited either fourth-order (Λ = 2.14 μm) [1] or from first- to fourth-order (with the strongest second-order, Λ = 1.07 μm) [2, 3] periods for the reflection peak at the telecommunications wavelength 1.55 μm. Since that time, the PbP technique employing the output of a femtosecond Ti:sapphire laser has evolved to become a relatively routine method for the fabrication of gratings in fibres with typical values of the inscribed period of 1.07 μm [3–8] or 1.12 μm (third-order grating for the 1080 nm reflection) [9].

Recently, with tightly focused 800 nm femtosecond light pulses, injected into a slab of pure fused silica using a special, reflective micro-objective, the fabrication of first-order gratings for 1550 nm wavelength (Λ = 0.535 μm) was reported [10, 11]. Very recently, by introducing index-matching fluid between the planar microscope cover slip and fibre, the same 535 nm period was achieved with FBG recording in standard fibre [12]. It should be noted that all of the foregoing investigations on point-by-point microfabrication have employed 800 nm femtosecond light pulses, which excite the samples of fused silica (or germanosilicate glass) via five-photon absorption [13]. Such a multi-photon approach can employ the different wavelengths and different numbers of photons in one elementary absorbing act [14], so facilitating inscription inside various non-photosensitive optical materials. It is also known that the propagation of a femtosecond 800 nm pulse inside a bulk dielectric (e.g. fused silica glass) with a peak power exceeding the threshold (critical power) results in self-focusing. Remarkably, this regime is characterized by reduction of the spatial dimensions of the photoinduced material modifications below the diffraction limit [15]. Since the achieved size of the pitch (modification) is about 270 nm and much smaller than the inscribing wavelength of 800 nm, further feature size reduction would appear to be highly unlikely using this method. However, shifting the wavelength of the inscribing Ti:sapphire laser radiation into the UV range (e.g. to 267 nm with simultaneous decrease in the order of absorption process from five-photon to two-photon [14])
immediately makes it possible to record structures with even smaller periods. Such a development is very important, for example, for the point-by-point fabrication of first-order Bragg gratings possessing a peak reflectance wavelength of \(\sim 1 \mu m\).

In our previous work, the use of 267 nm femtosecond pulses with 82 nJ energies led us to the inscription of 300 nm period structures [16]. In the current work, the optimizing of the inscription depth has allowed us to decrease the inscription energy down to 30 nJ and to inscribe the 250 nm period structure. In addition, the quality of the nanostructures was significantly improved in comparison with our previous report.

2. Experimental set-up

Femtosecond pulses at 800 nm were produced by a Ti:sapphire chirped pulse amplification laser system consisting of a ‘Tsunami’ oscillator and a ‘Spitfire’ amplifier (both from Spectra-Physics). The laser system delivered 0.8 mJ pulses with 150 fs duration and 1 kHz repetition rate. The IR beam diameter after the amplifier was 2.5 mm at FWHM. The set-up for third-harmonic generation (THG) was similar to that described earlier [17]. The pulses at 267 nm were produced by non-collinear sum-frequency mixing between fundamental radiation and that at the second harmonic (figure 1(a)). A half-wave plate was used to distribute the energy of 800 nm pulses between two channels, which allowed us to manipulate the energy at the entrance of second-harmonic generator and, thus, the energy of the output radiation at 267 nm. The pulses at 400 nm were produced in a 1 mm thick BBO crystal cut for type I collinear second-harmonic generation (\(\theta = 29.2^\circ\), \(\phi = 90^\circ\)) [18]. A second half-wave plate was used for 90\(^\circ\) polarization rotation of the 400 nm beam. Using three mirrors with high reflectance at 400 nm, the second-harmonic beam was separated from the fundamental. The 400 and 800 nm pulses were directed into a 1.0 mm thick BBO crystal cut for type I sum-frequency generation (\(\theta = 44.3^\circ\), \(\phi = 90^\circ\)) [18]. The angle between the 800 and 400 nm beams was less than 2\(^\circ\) in the horizontal plane. Using highly reflecting UV mirrors and the non-collinear geometry of THG, we easily separated the 267 nm radiation. The UV pulse energy was monitored by a PD10 photodiode (Ophir Optronics). The energy of the third-harmonic pulses was about 80 \(\mu\)J with pump energy at 800 nm of 800 \(\mu\)J. We estimated the width of 267 nm pulse to be about 300 fs.

Fused silica samples of 50 mm \(\times\) 20 mm size and 1 mm thickness (Schott Glas) were used in the experiments. They were moved in the horizontal plane in two perpendicular directions by an air-bearing translation stage ABL-1000 (Aerotech). The translation speed was varied in the range of 0.25–1.0 mm s\(^{-1}\). The absolute and relative micropositioning accuracies were both better than 50 \(\mu\)m.

The UV laser beam was directed in a strictly perpendicular direction on to the surface of the fused silica sample from the top (figure 1(b)). It could be focused to any selected depth between 0 and 600 \(\mu\)m below the surface with accuracy of 1 \(\mu\)m. For focusing, we used a reflective microscope objective with numerical aperture of 0.65 (Ealing), manipulated by a 3D-micropositioning manual translation stage 17 MAX 303 (Melles Griot). The inscription energy values were varied between 20 and 400 nJ, whilst the length of inscribed tracks was usually between 0.5 and 4 cm.

3. Characterization of inscribed structures

To establish the optimal conditions for inscription, one should carefully adjust at least four parameters including, particularly, the laser pulse energy, the speed of translation stage movement, the focus depth inside the sample, and the polarization of the inscribing light with respect to the direction of sample movement. Other parameters, including the numerical aperture of the microscope objective, the repetition rate, the wavelength and the duration of the inscribing pulses, also could be varied. The resulting number of experimental tracks could easily reach several thousands: thus, an express visualization method is needed for such optimization.

We perform the characterization of the irradiated samples using an optical microscope, Axioscope-2 MOT plus (Zeiss), which was equipped for both transmitted light and differential interference contrast (DIC) measurements. The resolution of a conventional optical microscope is considered to be of the order of the illumination wavelength: in our experiment, even with the use of a blue filter it was rather difficult to distinguish the 600 nm period perturbations induced in the bulk of the material. However, the use of DIC technique enabled us to monitor structures with periods down to 250 nm. In the experiments, we used the combination of a plan-apochromat oil immersed objective \((\times100/1.40/DIC)\) and an achromat–aplanatic condenser (1.4H/PH/DIC) with numerical aperture of 0.6 (or higher). A DIC prism (III/1.4) and DIC slider \((\times100/1.40III)\) were also used; such a combination seems to be the best one commercially available from Zeiss. The refractive
index variation is expected to be of the order of $10^{-4}$, which corresponds to a few times more than the sensitivity level of this DIC microscope, based on comparable experiments conducted with known samples.

It is important to note that DIC microscopy does not present the real image; rather, the resulting picture contains the information of both the intensity distribution and the derivative of the optical phase between two orthogonally polarized beams, spatially separated by a distance smaller than the resolution of the $\times 100$, $NA = 1.4$ microscope objective. Thus, periods of about few hundred nanometres can be detected.

For the independent resolution check of DIC microscopy, some tracks exhibiting topographic changes of the sample surface were characterized with atomic force microscopy (AFM) [19]. We used a commercial instrument, the Dimension Nanoscope III (VEECO), working in tapping mode.

4. Results and discussion

We allied the optimization procedure to the search for the optimal focus depth whereby inscription with smaller energy (leading to smaller beam diameter and, hence, to smaller nanostructure period) will be possible. In contrast with [16], in this work we used only the $\pi$ polarization of the 267 nm inscribing beam (parallel to the translation direction).

We started our inscription experiments by focusing our microscope objective on the surface of the fused silica sample. With pulse energy as small as 38 nJ, we were able to record nanostructures with a translation speed of 1.0 mm s$^{-1}$. Taking into account the repetition rate of 1 kHz, it is easy to deduce that the recorded grating nanostructure possesses a 1000 nm period (figure 2). The DIC microphotography taken at the surface of our fused silica sample shows that the laser beam cross-section in the focal plane is not ideal and varies in size from pulse to pulse. A similar picture was revealed by topographic changes of the sample surface while using the AFM technique (figure 3(a)). The asymmetry seen in individual voxels is probably related to the light diffraction on wire holders of a small mirror inside the micro-objective. From further consideration it will be evident that such diffraction becomes unimportant while focusing inside the slab of fused silica. We have also applied the AFM method for the independent calibration of our DIC microscopy approach.

Figure 3(b) (cross-section of the image of figure 3(a) along the grating) shows that 10 periods of our nanostructure inscribed on the surface of fused silica sample correspond exactly to 10 $\mu$m length, confirming our calibration shown at figure 2 (the scale bar is 10 $\mu$m). The cross-section in the perpendicular direction (presented in figure 3(c)), which is an ablation profile, gives an upper estimate of 400 nm for the diameter of the laser beam cross-section at FWHM at the surface of our sample, which agrees with the spot sizes deduced from figure 2 (210–310 nm).

To estimate the beam-waist diameter in the focal plane, one can use the well-known expression for diffraction-limited focusing, $w_0 = C \lambda/NA$, where $\lambda$ is the inscription
Figure 4. (a) DIC microphotographs of structures with 1000, 500 and 400 nm periods inscribed at the optimal focusing depth of 170 µm with sample translation speeds of 1.0, 0.5 and 0.4 mm s⁻¹, respectively. The energy of the inscribing pulses was 30 nJ. The size of the bar corresponds to 10 µm. (b) DIC microphotographs of similar structures inscribed earlier at a focus depth of 300 µm [16]. The energy of the inscribing pulses was 82 nJ. The size of the bar corresponds to 10 µm.

Figure 5. DIC microphotographs of structures with 300 and 250 nm periods inscribed at the optimal focus depth of 170 µm with sample translation speeds of 0.3 and 0.25 mm s⁻¹, respectively. The energy of the inscribing pulses was 30 nJ. The size of the bar corresponds to 10 µm.

wavelength, \( \lambda \) is a constant \( \sim 1.2–1.6 \) defined by the exact profile of the laser beam, and \( NA \) is the numerical aperture of the microscope objective. Substituting \( \lambda = 267 \) nm and \( NA = 0.65 \) gives the beam-waist diameter \( w_0 \) of 500–600 nm, which agrees with the experimental values given above.

Much better results—inscription at smaller translation speed values down to 0.25 mm s⁻¹—were obtained with the inscribing light tightly focused to a depth of 170 µm below the surface of a fused silica sample. Figure 4(a) demonstrates the periodic structures obtained using 30 nJ pulses with sample translation speed values of 1.0, 0.5, and 0.4 mm s⁻¹, respectively, i.e. with periods 1000, 500 and 400 nm, respectively. The excellent quality of the gratings obtained should be emphasized (cf structures with similar periods obtained in our previous work and depicted in figure 4(b)). Furthermore, the irregularity in spot size was strongly reduced.

The measurements of the visible diameter of the spots presented in figure 4(a) show that the decrease of the translation speed value from 1.0 to 0.5 mm s⁻¹ and further to 0.4 mm s⁻¹ leads to the simultaneous decrease of the visible diameter from 630 nm to 300 nm and then to 230 nm (with accuracy ±10%). Interestingly, a similar feature could be seen in the tracks with similar periods presented in our previous report (figure 4(b)). This could be related to the change of glass properties (e.g., induced UV absorption and/or refractive index change) performed by the previous neighbour inscription pulse (pulses), which increases the absorption and hence following self-focusing for the next inscription pulse (pulses). The decrease of the translation speed value should increase the probability of such an effect.

Figure 5 shows the nanostructures obtained at the same focus depth (170 µm) using sample translation speed values 0.3 and 0.25 mm s⁻¹, i.e. with periods of 300 and 250 nm, respectively. The deterioration of the quality of these gratings could be connected with overlapping between neighbouring spots (if the size of a spot exceeds a half of the nanostructure period, i.e., 150 nm) and/or with the nanostructure period
smaller than the resolution limit of DIC microscopy. Future investigation of recorded structures by scanning electron microscopy would help to elucidate this point. In any case, in this work at optimal focus depth, we have obtained evidence for nanostructure inscription with a period as small as 250 nm. We recall that, in our previous report [16], for this particular polarization of the inscribing laser beam only the 400 nm period structure was recorded.

The importance of using the optimal focus depth can be illustrated by experiments conducted at 330 μm below the sample surface. At this focus depth, using 89 nJ pulses, we managed to record structures with periods down to 500 nm (figure 6); at the optimal inscription depth (170 μm), with 92 nJ pulses, we recorded the structures of 400 and 300 nm periods (figure 7).

It should be emphasized that the value of the optimal inscription depth obtained in this work (170 μm), differs from the one obtained in our previous report (300 μm). The reason for this could be the inaccurate positioning of the correction ring of the microscope objective. Due to this, in our previous work, the microscope objective was not aligned of the fused silica layer thickness led us also to the inscription of the nanostructures with better quality then before.

In conclusion, using 30 nJ, 267 nm, 300 fs laser pulses, tightly focused at a depth of 170 μm below the surface of a fused silica sample, we have succeeded in recording nanostructures with periods from 1000 to 250 nm. To the best of our knowledge, the latter period has never been achieved before from this type of inscription process. The optimization of the fused silica layer thickness led us also to the inscription of the nanostructures with better quality then before.

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