Comparative study of UV absorption changes induced in germanosilicate glass by high-intensity femtosecond pulses at 267, 400 and 800 nm

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Abstract

We investigated UV absorption changes induced in 3.5 mol% Ge-doped fused silica at high-intensity (≈10^{11}–10^{13} W/cm^2) femtosecond (130 fs) irradiation at 267, 400 and 800 nm. We have shown that the induced spectra in the region 190–300 nm are similar in all three cases. At 800 nm irradiation, in addition to the UV absorption changes, we observed small-scale damage due to self-focusing. This damage appears when the incident pulse fluence value of about 1 J/cm^2 (pulse intensity of about 7.5 \times 10^{12} W/cm^2) is overcome, while the threshold for the induced absorption changes is twice lower.

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1. Introduction

High-intensity femtosecond pulses have been intensively used for photochemical fiber Bragg grating (FBG) inscription since 2003 [1,2]. Two types of femtosecond sources are most commonly used; in the infrared range a Ti:sapphire laser with the wavelength 800 nm [3,4], while in the UV range its third harmonic at 267 nm [5] or the fourth harmonic of a Nd:glass laser at 264 nm [6,7]. Standard telecommunication fibers (e.g., SMF-28 from Corning) consist of cladding made of pure fused silica, with an energy bandgap of 9.3 eV [8], and of core made of 3 mol% Ge-doped fused silica (energy bandgap 7.1–7.8 eV [4,9]). For FBG fabrication it is necessary to induce a periodic refractive index modulation in the germanosilicate fiber core. It was shown that under high-intensity 264 nm irradiation of standard telecom fibers, two-photon excitation of the core takes place [7], while at 800 nm five-photon absorption is essential [4], in both cases the bandgap energy value of germanosilicate glass is exceeded.

To explain the mechanisms of photochemical refractive index changes in the Ge-doped fused silica, different concepts were proposed, the first one is the colour-centre model [10]. It considers the release of electrons at photoexcitation, which become trapped in neighbouring sites, thus creating new colour centres. The following modification of the UV absorption spectrum of the germanosilicate glass leads to the refractive index modification at longer wavelengths.

Though the UV absorption spectra in germanosilicate glass induced by high-intensity 264 nm femtosecond radiation has been recently studied [11,12], no information is
available in literature on the absorption spectra induced by high-intensity 800 nm pulses. In this work, we report on a comparative study of induced absorption spectra in 3.5 mol% Ge-doped fused silica, caused by high-intensity ($\sim 10^{11} \text{ - } 10^{13}$ W/cm$^2$) femtosecond pulses with different wavelengths (267, 400 and 800 nm). The accompanying glass damage due to self-focusing was also studied under these experimental conditions.

2. Experimental set-up

Femtosecond pulses at 800 nm wavelength were produced by a Ti:sapphire chirped pulse amplification laser system [13]. This laser system delivers 1 mJ pulses with 130 fs duration at 800 nm with a repetition rate of 1 kHz. The pulse duration was measured using a multishot autocorrelator based on a BBO crystal, cut for type I noncollinear second harmonic generation ($\theta = 29^\circ$, $\phi = 90^\circ$). The IR beam diameter (3.5 mm at FWHM) was measured using a 14-bit CCD camera with a pixel number of $1200 \times 1024$ and a pixel size of $4.65 \times 4.65$ µm. The 800 nm beam profile was 90% fit by Gaussian distribution. The pulses at 400 nm were produced by frequency doubling of the fundamental wave in a 1 mm thick BBO crystal cut for type I collinear second harmonic generation ($\theta = 29.2^\circ$, $\phi = 90^\circ$). The second harmonic beam was separated from the fundamental one by two beamsplitters with a high reflection at 400 nm. The pulses at the third harmonic of the fundamental (267 nm) were produced by sum-frequency mixing. The pulses of the fundamental wave (800 nm) and its second harmonic (400 nm) were directed into a 1.5 mm thick BBO crystal cut for type I sum-frequency generation ($\theta = 44.3^\circ$, $\phi = 90^\circ$). The angle between the 800 nm and 400 nm beams was about 2° in the horizontal plane. Using mirrors, highly reflecting at 267 nm, and the spatial walk-off of the sum-frequency beam, we easily reached separation of the ultraviolet radiation.

To change the pulse intensity we slightly focused the fundamental laser beam (or its harmonics) using a fused silica lens with a focal distance of 50.2 cm (for 800 nm) or 48.5 cm (for 400 nm) and a CaF$_2$ lens with a focal distance of 47 cm (for 267 nm). For the exact calculation of intensity we used the expressions given in [14].

In the experiments, we used 3.5 mol% Ge-doped samples of germanosilicate glass, similar to those investigated earlier [11,12]. The thickness of the samples was 0.085 cm. For spectral measurements of the induced absorption changes, the samples were fixed on diaphragms, through which the femtosecond laser beam was directed. The diameters of the diaphragms were 0.6 mm (for 800 nm pulses) and 1 mm (for 267 and 400 nm pulses). Care was taken to install the diaphragm each time in exactly the same position on the optical bench or inside the UV spectrophotometer (UV-3101PC, Shimadzu, Japan).

For the observation of the sample damage after the irradiation we used an optical transmission microscope with a 10× magnification.

3. Results and discussion

Fig. 1 demonstrates the experimental difference absorption spectra, taken at 800, 400 and 267 nm with different incident intensities and fluences values. It is clear that the obtained spectra are qualitatively similar. This means that the colour-centre model could explain the photosensitivity of germanosilicate glass not only when irradiating by femtosecond pulses at 267 nm, but also when using high-intensity 800 and 400 nm pulses. As it follows from Fig. 1, the change in the irradiation wavelength from 267 nm to 400 nm and further to 800 nm is accompanied by the substantial increase of irradiation intensity needed for a similar change in UV absorption, from ~200 to ~10 000 GW/cm$^2$. This is in line with the change in the order of the non-linear absorption process, from the two-photon in the case of 267 nm [7] to the five-photon in the case of 800 nm [4]. In the case of 400 nm excitation it should probably be a three-photon absorption process, similar to that recently demonstrated for the 352 nm femtosecond grating inscription in a telecom fiber [15].

It should be noted that at 800 nm irradiation, in addition to UV absorption changes, we observed small-scale damage (Fig. 2). The observed back surface morphology could be explained only as a result of beam small-scale self-focusing and whole beam breaking into intense filaments inside the material. Indeed, the damage structure was observed not only on the back surface, but had also already begun inside the volume. The number of damage points increased as the back surface was approaching.

The appearance of small-scale damage was accompanied by a strong fluorescence. We did not find any such damage either at 267 nm or at 400 nm. This is probably due to the much higher single pulse incident fluence value (up to 1.3 J/cm$^2$) in the 800 nm case.

The next goal was to determine the intensity thresholds for the induced linear absorption changes and for the optical damage at 800 nm. In this experiment, we exposed the different samples with the same number of exposing pulses ($5 \times 10^5$) but with varying values of incident energy from 600 to 150 µJ, so simultaneously the incident intensity and fluence were changed. For better accuracy of the intensity value, the 800 nm beam diameter was measured at the sample plane, the FWHM value was found to be of 160 µm. For the reason of reaching better sensitivity towards small absorption changes we used the smaller diaphragm of 0.4 mm diameter. This, however, brought more noise to the measured difference absorption spectra.

Our results for different incident irradiation intensities relative to the 800 nm case are presented in Fig. 3. We started at highest pulse energy (600 µJ), where the absorption change and the damage were clearly visible. Decreasing the energy resulted in decrease of the number of damage points on the output sample surface (Fig. 4). Visible damage disappeared fully when the energy dropped below the value of 300 µJ, corresponding to an intensity of $7.5 \times 10^{12}$ W/cm$^2$. However, the effect of the induced
absorption change remained until the second threshold (150 μJ or 3.7 × 10^{12} W/cm²) was reached. Interestingly, at the same time the absorbed fluence decreased from 65 kJ/cm² (600 μJ) to 12 kJ/cm² (300 μJ) and further to zero at 150 μJ.

The presence of two thresholds at high-intensity 800 nm irradiation corresponding to two types of damage, fully reversible by 900 °C heating (type I) and irreversible (type II), was found for pure fused silica in 2001 [16]. The threshold for the type II damage corresponded well to the critical power of self-focusing of 800 nm laser radiation in fused silica glass. Recently, the existence of two different intensity thresholds related to two different types of fiber photosensitivity was established in experiments on femtosecond 800 nm FBG inscription in 3 mol% germanosilicate glass.

Fig. 1. Difference absorption spectra induced by high-intensity femtosecond 800, 400 and 267 nm pulses in 3.5 mol% Ge-doped fused silica. Incident intensity and fluence values were: (1) 9.2 × 10^{12} W/cm², 289.2 kJ/cm²; (2) 9.1 × 10^{12} W/cm², 85.7 kJ/cm²; (3) 3.9 × 10^{12} W/cm², 30.1 kJ/cm²; (4) 2.8 × 10^{12} W/cm², 29.5 kJ/cm²; (5) 2.3 × 10^{11} W/cm², 0.361 kJ/cm²; (6) 2.3 × 10^{11} W/cm², 0.025 kJ/cm².

Fig. 2. Microphotograph of small-scale damage in 3.5 mol% Ge-doped fused silica taken at the back surface of the sample. Incident intensity, pulse fluence and number of laser pulses were 9.2 × 10^{12} W/cm², 1.2 J/cm² and 3 × 10^6.

Fig. 3. Difference absorption spectra induced by high-intensity femtosecond 800 nm pulses in 3.5 mol% Ge-doped fused silica. Incident intensity, incident fluence and absorbed fluence values were: (1) 14.9 × 10^{12} W/cm², 239 kJ/cm², 64.5 kJ/cm²; (2) 8.7 × 10^{12} W/cm², 139 kJ/cm², 19.5 kJ/cm²; (3) 6.2 × 10^{12} W/cm², 99.5 kJ/cm², 8.0 kJ/cm²; (4) 5.0 × 10^{12} W/cm², 79.6 kJ/cm², 4.0 kJ/cm²; (5) 3.7 × 10^{12} W/cm², 59.7 kJ/cm², 0 kJ/cm².
The direct comparison with our damage threshold is difficult due to the uncertainties in absorbed fluence in both studies. However, our measured absorbed fluence threshold value for single-quantum 248 nm type II FBG inscription in 15 mol% germanosilicate glass (0.22 J/cm²) [17]. Therefore, the obtained experimental data testify that the type II fiber photosensitivity at high-intensity 800 nm femtosecond irradiation is linked with the self-focusing in germanosilicate glass.

4. Conclusion

We observed a similarity between the induced absorption change spectra in the case of high-intensity femtosecond excitation of 3.5 mol% germanosilicate glass at 267, 400 and 800 nm. This means that the colour-centre model could explain the type I photosensitivity of germanosilicate glass in all these three cases. At 800 nm irradiation, in addition to UV absorption changes, we observed small-scale damage due to self-focusing (type II irreversible damage). This damage appeared when the characteristic incident pulse fluence value of about 1 J/cm² (pulse intensity of about $7.5 \times 10^{12}$ W/cm²) is overcome, while the threshold for the induced absorption changes at 800 nm was twice lower.

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