Optik 125 (2014) 6256–6259

Contents lists available at ScienceDirect

Optik

journal homepage: www.elsevier.de/ijleo

Absorption and luminescence spectroscopy of fused silica by multiple pulses irradiation at 355 nm

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ARTICLE INFO

Article history:
Received 14 November 2013
Accepted 16 June 2014

Keywords:
Transient absorption
Fused silica
Laser irradiation
355 nm

ABSTRACT

In this work, the effects of laser irradiation on fused silica at 355 nm are investigated by using transient absorption spectroscopy and luminescence spectroscopy. Our result shows that no transient absorption or luminescence in the spectra range from 400 nm to 600 nm is observed when laser energy density is below the damage threshold. When the laser energy density reaches the threshold, an initial damage site will be created. After subsequent laser pulses irradiation, the damage size grows. At the same time, the intensity of the transient absorption and luminescence spectra at the damage site also raises remarkably with the laser pulse number increasing. The absorption band from 420 nm to 520 nm is probably related to the absorption of impurity such as metal ion of iron, cerium and copper. Laser modified fused silica exhibits intense broad luminescence bands due to oxygen-deficiency centers at 444 nm and 580 nm.

1. Introduction

Due to its simple composition, vast availability in pure form and ease of processing, vitreous silica is often used as a model to study the physicals of amorphous solids. Research of amorphous silica is also motivated by its ubiquity in modern technology, a prominent example being as bulk material in transmissive and diffractive optics for high-power laser applications [1,2]. Laser induced damage of optical components has been one of the limiting factors in developing high power laser systems [3]. While the defect, which is introduced by manufacturing process and polishing process, could commonly decrease the damage threshold of optical materials. New finishing method [4], etching combined with laser conditioning [5] and CO_2 laser mitigation have been introduced to minimize these defects.

Single pulse damage is triggered by localized surface defects while multiple pulse damage is the result of accumulation of plastic deformation resulting from laser induced thermal stress [6]. If the bulk material contains absorbing inclusions, the damage threshold is often determined by the thermal stress induced by the heating of the inclusion [7]. The majority of energy deposit through a laser-drive absorption front during laser induced damage on the exit surface of fused silica [8]. Smith et al. [9] found that the dissolved molecular hydrogen exerts influence on laser induced absorption in fused silica. The energy via linear absorption mechanism and the absorption coefficient [10] of the modified material are estimated. Miyamoto et al. [11] evaluated the nonlinear absorptivity in internal modification of bulk glass. Deng et al. [12] developed a non-perturbative quantum theory for phonon-assist photon absorption of conduction band electron in intense laser.

On the other hand, fluorescence spectroscopy can also been used as a diagnostic and development tool in the interaction between laser pulse and optical materials. Laser modified fused silica exhibits intense broad fluorescence bands due to non-bridging oxygen hole centers (at 650 nm) and oxygen-deficiency centers (at 281 nm and 478 nm) [13,14]. Considerable attention has been paid to the fluorescence spectroscopy at subsurface damage [15] and damage [16–18] sites of silica. In this work, the effects of multiple pulses laser irradiation on fused silica at 355 nm are investigated by using transient absorption spectroscopy and luminescence spectroscopy. The investigation of transient absorption

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http://dx.doi.org/10.1016/j.ijleo.2014.08.023
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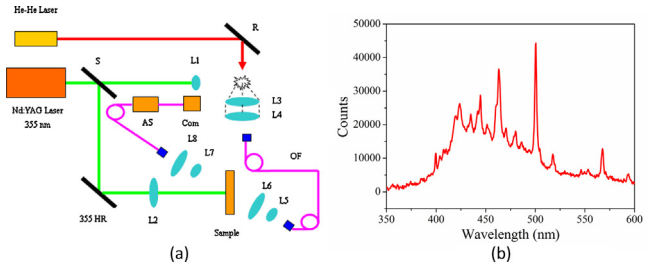

Fig. 1. (a) Experimental setup for laser induced transient absorption spectroscopy. R: reflector; S: spectroscope (1/3 split ratio for transmission/reflection); L1, L5, L7: lenses, $f = 50$ mm; L2: lenses, $f = 40$ mm; L3, L4, L6, L8: lenses, $f = 150$ mm; HR: high reflector, OF: optical fiber; AS: Avaspec spectrometer, Com: compute. (b) Typical spectra generated through air ionization. The energy density is about 5.28 J/cm^2.

and luminescence spectroscopy in fused silica is meaningful for understanding damage mechanisms in optical materials by UV laser irradiation.

2. Experiment

The substrates used for the experiments were UV-grade fused silica which was manufactured by flame hydrolysis. The impurities in fused silica are introduced by the polishing process. The fused silica samples with a dimension of 50 mm × 50 mm × 5 mm were optically polished on both surfaces using best-known continuous pitch polishing techniques. Prior to laser irradiation, samples have been washed thoroughly with distilled de-ionized water to rinse off any particles induced by storage and handing on the optic surface, which was followed by an alcohol rinse with absolute ethanol to remove residual water.

Experimental setup for laser induced transient absorption spectroscopy is shown in Fig. 1. A Nd:YAG laser (Innolas, Germany) with repetition rate of 1 Hz, pulse duration of 6.8 ns, central wavelength of 355 nm was used for the experiment. Laser pulse was split into two components with a spectroscope (1/2 split ratio for transmission/reflection). The reflected component was used for the pump beam and the transmitted component for generating white light continuum as the probe beam. The intense reflected component was focused on the exit surface of fused silica by lens ($f = 385$ mm). The transmitted laser was focused ($f = 50$ mm) to ionize air to produce white light continuum ranged from 400 nm to 600 nm. Typical spectra of the white light were shown in Fig. 1(b). The pulse duration of continuum was record by a oscillograph (Lecroy) with the resolution of ~200 ps. The pulse duration of the white light is about 10 ns with the best coupling angle between the probe light and the optical fiber A low-power, He–Ne laser beam with approximately 2-mm diameter was used as a collimated beam. After air ionization, white light continuum is collected by an optical system composed of two lenses. The narrow-band pump and the broadband probe beams are focused onto the same site of the sample. The profile of the pump beam is near-Gaussian with $1/e^2$ diameter of about 1.6 mm at the rear surface of fused silica. The diameter of the probe beam is about 1.5 mm, which is completely covered by the pump beam. Different time delays between the pump beam of 355 nm and the probe beam are determined by changing the length of optical fiber.

In the absorption mode, the absorbance is calculated using the current sample, reference and dark data sets in the following equation:

$$A_n = -\log\left(\frac{sample_n - dark_n - sca_n}{ref_n - dark_n}\right) \quad (1)$$

Here, ref_n represents the transient white light spectrum without pump pulse laser irradiation, $dark_n$ stands for the optical spectrum without probe and pump pulse irradiation with the sample, sca_n refer to the scatter light after the pump pulse irradiated the front surface of fused silica and $sample_n$ means the transient white light spectrum when the probe light transmit the front surface of the sample. All the spectra were normalized by the spectral peak which centered at 564 nm and smoothed by the method of Fast Fourier Transform (FFT) to reduce the noising signal. It should be noted that the fluorescence information of fused silica by laser irradiation at 355 nm can also be obtained by the scatter light spectra if the probe beam was removed.

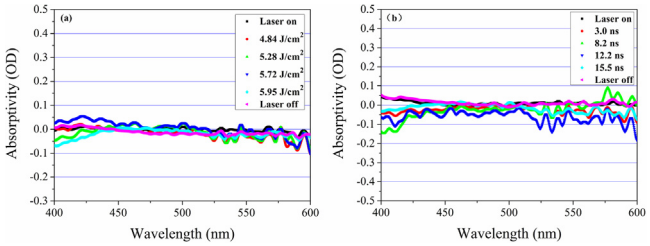

Fig. 2. (a) Transient absorption spectroscopy of fused silica by laser irradiation at different energy densities. The pump beam and probe beam are nearly synchronized. The energy densities are 4.84 J/cm^2, 5.28 J/cm^2, 5.72 J/cm^2 and 5.95 J/cm^2, respectively. (b) Transient absorption spectroscopy with different time delays between pump beam and probe beam with energy density 4.84 J/cm^2.

3. Results and discussion

The damage threshold of fused silica is near 5.95 J/cm^2 when laser irradiated on the exit surface of fused silica. Fig. 2(a) shown transient absorption spectroscopy of fused silica irradiated by laser with different energy densities. The energy densities are 4.84 J/cm^2, 5.28 J/cm^2, 5.72 J/cm^2 and 5.95 J/cm^2, respectively. There is no transient absorption before laser irradiation. No transient absorption can be observed when the laser energy density is lower than the damage threshold. Fig. 2(b) shows time-resolved transient absorption spectroscopy of fused silica under laser irradiation. It reveals that there is no transient absorption with the time delay increasing. This finding results from the fact that the transient absorption was probed only in the vis region, rather than in the UV where the interesting physics triggered by laser irradiation should be observable (as most point defects in SiO_2 absorb in the UV).

In Fig. 3, transient absorption spectroscopy of fused silica irradiate by different number of pulses is shown. The pump beam and probe beam are nearly synchronized. The energy density is about 4.84 J/cm^2. With the increase of number of pulses from one to seven, (a) and (b) show the transient absorption spectroscopy and luminescence spectroscopy separately at un-damage site. When the number of pulses is one, there is no absorption and scatter whether the laser is on or off. Number of pulses cannot affect the intensity of transient absorption spectroscopy at the un-damage site. This

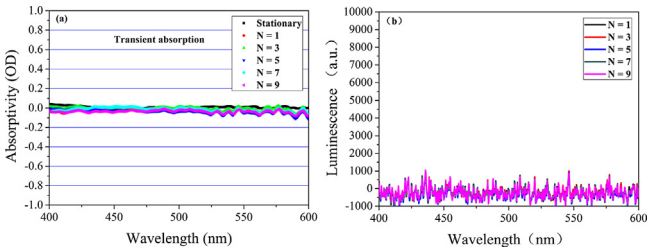

Fig. 3. Transient absorption spectroscopy of fused silica irradiate by different number of pulses. The pulses were not selected. The delay is about 3 ns. The energy density is about 4.84 J/cm^2. With the increase of number of pulses from one to seven, (a) and (b) show the transient absorption spectroscopy and luminescence spectroscopy separately at the un-damage site.