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Rapid Communication

Laser-induced defects in fused silica by UV laser irradiation

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Abstract

The structure of fused silica with irradiation of the third harmonic of the Nd:YAG laser was investigated by Fourier transform infrared, Raman and Photoluminescence spectroscopy. Some variation in the Si/O stoichiometry of silica in the ablated spot was induced. The primary defect species are oxygen deficient centers and oxygen interstitials. The frequency shift of the Si–O–Si vibration proves that the central force constant between oxygen and silicon atoms, and the band angle of Si–O–Si increases in the UV-laser ablation spot. Small silicon clusters within SiO_x appear to be a possible explanation for the 564 nm Fluorescence peak, and the 181 cm^{-1} Raman peak.

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Keywords: Laser-matter interactions; Optical properties

1. Introduction

With the advent of the high power laser systems such as National Ignition Facility (NIF) and Laser MegaJoule (LMJ) [1], and recent industrial interest in far UV lithography for microelectronics and telecommunication [2], laser-induced damage in transparent optics under high power UV irradiation has aroused considerable basic and technological interest in recent years [3–6]. Fused silica is an important material, particularly for high quality optical elements to sustain large pressures and temperatures accompanying energy absorption. During intense irradiation of fused silica with 3ω (355 nm) laser, significant damage in the form of craters can be produced in the optics. The lateral size of this damage grows exponentially with the number of pulses and limits the lifetime of the optics [6]. In the damage crater irradiated with pulse energy of 45 J/cm^2, a compaction layer of about 10 μm thick and 20% higher in density has been identified at the bottom of the crater with X-ray tomography [7,8]. Both of thermal diffusion and shock wave generation could lead to rapid cooling or quenching of the modified region. While the exact process is uncertain, the end results are localized physical, chemical, and structural changes of the material exposed to the laser beam. These alterations can be involved with densification, refractive index changes, and/or defects formation [7–9].

Radiation-induced defects in fused silica have been studied extensively, such as β irradiation [10], energetic ions [11], protons [12], γ rays and neutron beam radiation [13], UV [7] and IR laser radiation [9]. These defects may be electronic in nature including a re-distribution of the local electron density to give rise to paramagnetic as well as diamagnetic centers, or structural in nature involving local atomic displacements from the normal random network structure of fused silica. The latter defects may be oxygen-excess or oxygen-deficient. These radiation-induced defects, such as NBOHC (non-bridging oxygen hole center), and ODCs (oxygen deficient center), a STE (self-trapped exciton), and E′ center, and O_{int} (interstitial oxygen, peroxy radicals or O_2 molecules) have been

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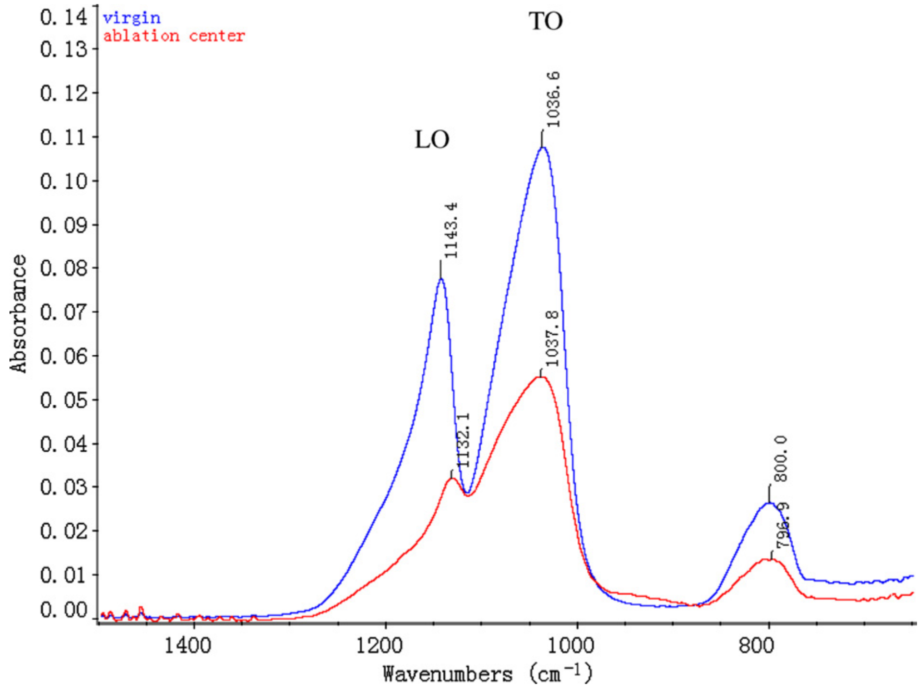

Fig. 1. Dependence of the IR absorption spectrum on the virgin and ablated spot. The peaks were found and labeled with 'Find Peaks' toolbar of Thermo OMNIC software. The spectral resolution was 1.9285 cm^{-1}.

extensively studied and characterized over the last few decades using a variety of spectroscopic probes [7,9,14].

In this work, a series of structural analysis of fused silica before and after 3ω (355 nm) Nd:YAG laser irradiation were carried out by using of infrared, Raman, and photoluminescence spectroscopy. We then discussed how the structure was modified.

2. Experimental

FT-IR (Fourier transform infrared) spectra of the fused silica samples were recorded with a Nicolet 5700 spectrometer.[1] A reflection mode was employed to measure the FTIR absorption spectra in the frequency region associated with the Si–O–Si stretching and bending vibrations, ranging 400–1600 cm^{-1}. Raman spectra measurement were performed with a Fourier-transform Raman module, using a 1064 nm laser as excitation source with a power of 3 mW. All Raman spectra were obtained in the 90° scattering configuration. The beam was consistently focused on the interested area. The Raman spectra were measured over the range from 100 to 1500 cm^{-1}.

The nature of defects was confirmed by the photoluminescence (PL). The instrument was F900,[2] equipped with a 450 W xenon lamp, and double monochromator in excitation and detection arms. PL spectra with detection scope of 300–600 nm were gained by the single photon photomultiplier detection system S900, working in single photon counting mode. PL spectra with detection scope of 300–1400 nm were gained by the C9940, which is a liquid nitrogen cooler specifically designed for the Hamamatsu R5509 series near-infrared photomultiplier tube (PMT).

The sample was dry fused silica with a dimension of $40 \times 40 \times 5\ mm^3$. The investigated area was ablated by five laser pulses with specs of 355-nm, 6.4-ns, and 24 J/cm^2. The results of the virgin area were also gained.

3. Results

3.1. IR absorption spectroscopy

Fig. 1 shows the dependence of the IR absorption spectrum on the virgin and ablated spot. The main features of the infrared reflection spectrum of fused silica are a peak centerd around 800 cm^{-1}. It is attributed to the bending motion of the oxygen atoms, and a strong absorption peak at 1036 cm^{-1} with a high-frequency shoulder at 1143 cm^{-1} due to the splitting of transverse optical (TO) and longitudinal optical (LO) mode components of the asymmetric stretching (AS) of tetrahedral Si–O units [15,16]. Due to the IR absorption spectrum of ablation center with UV laser, first, the AS mode (LO) shifts to lower frequencies, from 1143 to 1132 cm^{-1}. Secondly, the shoulder of the AS mode (LO) gradually disappears and the full width at half maximum (FWHM) of the AS mode (TO) increases from 81 to 92 cm^{-1}. Thirdly, the oxygen bending mode becomes weaker and also shifts to lower frequencies.

[1] Thermo Electron Ltd.
[2] Edinburgh Instruments Ltd.