Repetition rate dependence of absorption of fused silica irradiated at 193 nm

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Repetition rate-dependent absorbance measurements of synthetic fused silica at 193-nm irradiation are performed in the range of 50–1 000 Hz with an ArF laser calorimeter. The "apparent" single- and two-photon absorption coefficients are determined by measuring the laser fluence-dependent absorbance of fused silica samples with different thicknesses to separate the surface absorption and bulk absorption. The measurement results indicate a reversible nonlinear increase of both apparent single- and two-photon absorption coefficients with increasing repetition rate for the synthetic fused silica, whereas the surface absorption shows no dependence on the repetition rate.

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The excimer laser at 193-nm wavelength is widely used for ultra-large scale integrated circuit lithography manufacturing and laser micro-fabrication. For such applications, only a few wide band-gap optical materials with acceptable transmission are available. Among these optical materials, synthetic fused silica is currently the most widely used because of its high transmission, high chemical and physical stability, and small birefringence at 193 nm[1]. However, long-term excimer laser irradiation at 193 nm results in optical degradation of fused silica[2]. The absorption behavior of fused silica at 193 nm is characterized to predict the long-term stability of fused silica. In the past, fused silica absorption has been measured using laser-induced deflection (LID)[3−5], laser calorimetry (LCA)[6], and transmission measurement[7−9], which has a lower sensitivity compared with LID and LCA, as well as Hartmann-Shack wavefront sensor[10,11]. Among which, LCA was chosen as the international standard for absorbance measurements of optical laser components because of its high sensitivity and easy calibration[12].

Absorbance measurements are performed at 193 nm according to ISO 11551[12,13], using excimer laser calorimeter (Laser Zentrum Hannover, Germany). The experimental setup is shown in Fig. 1. An ArF excimer laser (IndyStar, Coherent), with a maximum repetition rate of 1 kHz, a pulse length of 10.5 ns (integral square value), and a beam diameter of 2 mm after the beam steering optics, served as the irradiation source. A variable attenuator inside the optical path is employed to allow laser density (laser fluence) adjustment. The whole optical arrangement is embedded into a sealed chamber and purged with high purity N_2.

The five fused silica samples (OH content<1 000 ppm) under investigation are ultraviolet (UV) optical materials used for 193-nm laser application. These samples are prepared from the same batch to have the same bulk absorption behavior. The samples are also prepared in an identical polishing process to have the same surface absorption. The root mean square (RMS) roughness of the polished surface is approximately 0.5 nm. Prior to the actual measurements, the samples were pre-irradiated at 193 nm with a total dose of approximately 2.2 kJ/cm^2 until the absorbance reached a stable value[14]. During an absorption measurement cycle, the samples are irradiated for 120 s.

For the fused silica samples with both surfaces optically polished, surface absorption, as well as linear and nonlinear bulk absorption properties are observed. Surface absorption originates from the surface and sub-surface defects, as well as the adsorbed contamination particles (such as polishing powders). In the bulk, energy levels within the energy gap between valence band (VB) and conduction band (CB) can be populated through the absorption of a single photon due to impurities and defects. This process is independent of energy density,

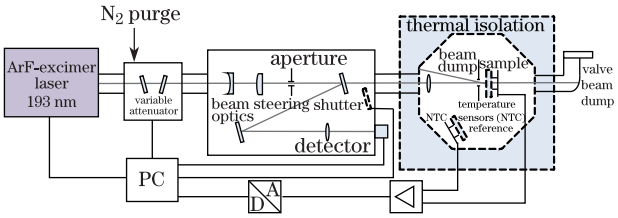

Fig. 1. Experimental setup.

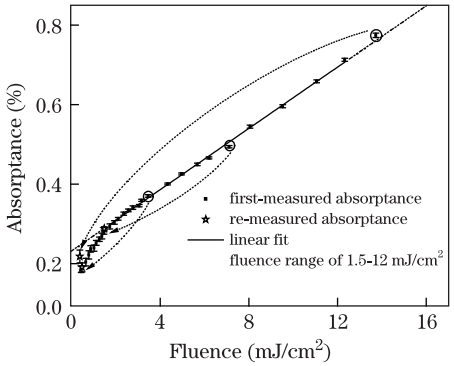

Fig. 2. Absorbance as a function of laser fluence for a fused silica sample at 193 nm for repetition rate of 1 000 Hz.

according to Beer's law. The absorption of two photons occurs if the sum energy of two photons exceeds the band gap. Hence, fused silica (approximately 8 eV) should exhibit two-photon absorption at 193 nm (6.4 eV). At a constant pulse length, this process is approximately linearly dependent on energy density. In addition, laser irradiation-induced defects and defect annealing have significant impacts on fused silica absorption[9].

The absorbance of a fused silica sample (thickness 2.1 mm) is measured at a repetition rate of 1 000 Hz for a laser fluence range from 0.4 to 14.0 mJ/cm^2, the results. are presented in Fig. 2. After measurements at 3.5, 7.1, and 14 mJ/cm^2, the absorbance was re-measured at a lower laser fluence to check the possible occurrence of irradiation-induced optical degradation. Negligible optical degradation is observed when laser fluence is less than 8 mJ/cm^2. A nonlinear increase of the absorption with laser fluence is observed from Fig. 2. However, at a laser fluence range from 1.5 to 2 mJ/cm^2, an approximate linear dependence of the absorbance on the laser fluence is evident, from which "apparent" single- and two-photon absorption coefficients can be defined and extracted.

In the laser fluence range of 1.5 to 12 mJ/cm^2, the total absorbance A can be written as

$$\begin{aligned} A(d,H,f) &= A_{\text{surf}} + \left[\alpha(f) + \beta(f)\frac{H}{\tau}\right]\cdot d \\ &= [A_{\text{surf}} + \alpha(f)\cdot d] + \beta(f)\frac{d}{\tau}\cdot H \\ &= A_0(d,f) + B(d,f)\cdot H, \end{aligned} \tag{1}$$

where A_{surf} denotes the surface absorption, α and β are defined as the apparent single- and two-photon absorption coefficients respectively, d is the sample thickness, f is the laser repetition rate, H is the laser fluence, and τ is the pulse length. Both apparent single- and two-photon absorption coefficients are attributed to the bulk absorption of fused silica material as well as absorption of defects generated by laser irradiation.

According to Eq. (1), the separation of surface and bulk absorptions can be established through measuring the thickness dependence of the total absorption A, provided that the samples have the same α, β, and A_{surf}. The apparent single- and two-photon absorption coefficients can be determined via measuring the laser fluence dependence of the bulk absorption.

The absorbances of four fused silica samples (diameter 25.4 mm, thickness 2.1, 4.0, 5.7, and 8.1 mm, respectively) are measured with different laser fluences and repetition rates. To minimize irradiation-induced optical degradation during the whole round of measurements, absorption measurements are performed within the fluence range from 1.5 to 5.0 mJ/cm^2. The laser fluence dependencies of the total absorbance of the four samples are measured at a repetition rate of 400 Hz, and the results are presented in Fig. 3. A linear dependence of the absorbance A on the laser fluence H is evident. According to Eq. (1), the laser fluence-independent absorbance $A_0(d,\ 400\ \text{Hz})$ is given by the intercept with the y-axis, and the apparent two-photon absorbance $B(d,\ 400\ \text{Hz})$ is extracted from the slope of the linear fit.

Measuring the laser fluence dependencies of the total absorbance of the four samples at different repetition rates, the laser fluence-independent absorbance $A_0(d,f)$ and the product of apparent two-photon absorbance and pulse length $(B\cdot\tau)(d,f)$ are plotted against the sample thickness, as shown in Fig. 4. Clearly, both are a linear function of the thickness, indicating that the four samples have approximately the same apparent single- and two-photon absorption coefficients because they are prepared from the same batch.

The surface absorbance A_{surf} can be determined from the intercept, which almost remains constant and independent of the repetition rate, whereas the apparent single-photon absorption coefficient $\alpha(f)$ determined from the slope of the linear fit increases with increasing repetition rate, as both shown in Fig. 5. An average surface absorbance of approximately 0.07% (including absorption from both surfaces) and an apparent single-photon absorption coefficient

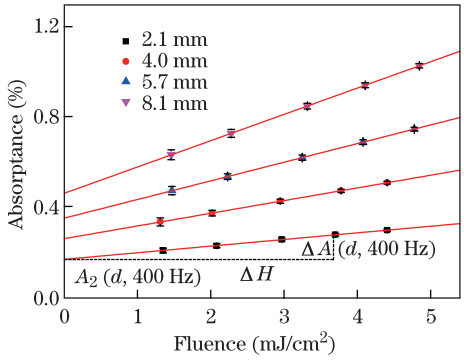

Fig. 3. (Color online) Absorbance versus laser fluence H for fused silica samples with different thicknesses at 193 nm with repetition rate of 400 Hz.

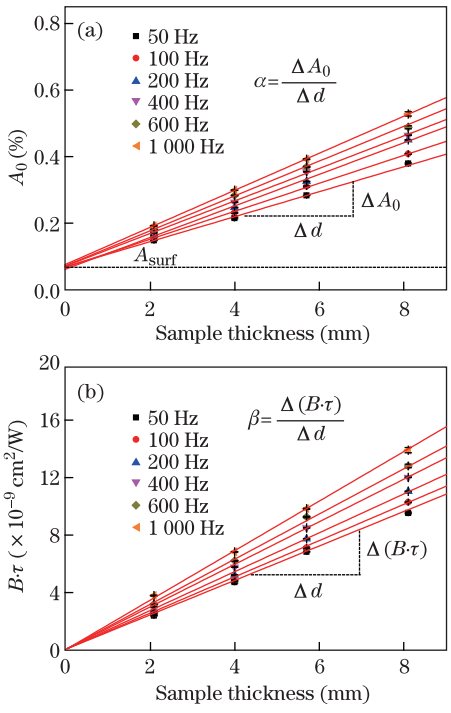

Fig. 4. (Color online) (a) Laser fluence-independent absorbance A_0 and (b) product of apparent two-photon absorbance and pulse length $(B\cdot\tau)$ versus sample thickness at different repetition rates.

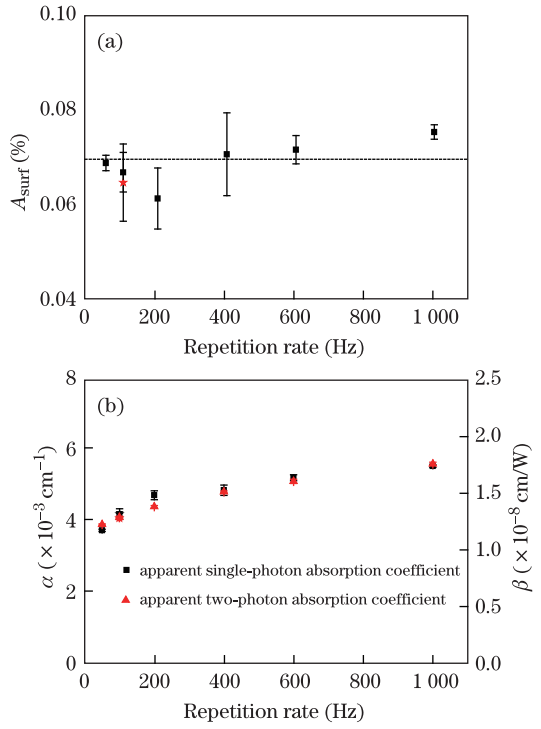

Fig. 5. (a) Surface absorbance A_{surf}, (b) apparent single-photon absorption coefficient α and apparent two-photon absorption coefficient β versus repetition rates. The symbol $\star$ indicates the re-measured data at 100 Hz.

of 5.6×10^{-3} cm^{-1} at 1 000 Hz are obtained.

The results presented in Fig. 4 (b) show that the product $(B\cdot\tau)(d,f)$ is proportional to the sample thickness, with zero offset at the y-axis, indicating that two-photon absorption has a pure bulk effect. The slope represents the apparent two-photon absorption coefficient β, which also increases with the increasing repetition rate. Moreover, repetition rate dependencies of both apparent single- and two-photon absorption coefficients have the same trend, as seen from Fig. 5(b), strongly implying that both single- and two-photon absorption may be related to the same origin(s) in synthetic fused silica. The apparent two-photon absorption coefficient at 1 000 Hz is determined to be 1.76×10^{-8} cm/W. After finishing the whole round of measurements on the laser fluence and repetition rate dependencies of the total absorbance, the samples were re-measured at 100 Hz with different laser fluences and surface absorbance, and the apparent single- and two-photon absorption coefficients are re-determined; the results are plotted in Fig. 5 with symbol $\star$. The good agreement between the first-measured and re-measured results at 100 Hz indicates negligible irradiation-induced optical degradation (absorption rise) during the whole round of measurements.

In conclusion, in previous research, the dependence of the absorption behavior of fused silica on the repetition rate is explained by the generation and annealing of E' centers[5,14]. The exposure of fused silica to high intensity UV light leads to the formation of absorbing defect centers such as E' and Non Bridging Oxygen Hole (NBOH) centers[15−17]. Meanwhile, E' center annealing between laser pulses (also called dark periods) during laser irradiation occurs due to the interaction with hydrogen molecules in fused silica. Thus, at a higher repetition rate, i.e., a shorter dark period, the amount of annealed defects is reduced, thereby yielding a higher defect center concentration and consequently a higher absorbance.However, only the total absorbance is taken into account[5,18] and the two-photon absorption is believed to be independent of repetition rate in synthetic fused silica in the existing theoretical and experimental investigations on the repetition rate dependence of the fused silica absorption. In this study, the separate measurements of the dependences of the single- and two-photon absorptions on the repetition rate suggest that both apparent single- and two-photon absorption coefficients have a reversible nonlinear increase with the repetition rate from 50 to 1 000 Hz. The similar increasing trend strongly implies is that both single- and two-photon absorptions may be related to the same origin(s). These results provide more information to the insight into the absorption mechanism of synthetic fused silica.

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