

Measuring small absorption in highly transparent DUV materials by a pump and probe technique

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ABSTRACT

A compact experimental setup, based on the laser induced deflection technique (LID), is introduced as an alternative for measuring small absorption in highly transparent materials for UV/DUV laser applications with high sensitivity and accuracy. In order to achieve the absorption coefficient of a sample, a comfortable and precise electrical calibration procedure is applied. The investigation of two equivalent fused silica samples of different thickness confirmed that the setup allows to exclusively measure the bulk absorption of the material without contributions from the irradiated surfaces. Furthermore, influences of irradiation parameters like repetition rate and pulse width on the absorption coefficient of fused silica at a fixed applied energy density have been investigated. The results confirm the complexity of the absorption mechanism present in fused silica upon laser irradiation.

Keywords: fused silica, laser induced absorption, pump and probe technique, probe beam deflection

1. INTRODUCTION

High quality optical materials for UV applications exhibit very small initial absorption coefficients in the $\leq 10^{-3}/\text{cm}$ range. In addition, nonlinear optical phenomena occur upon pulsed UV laser irradiation. Usually, the absorption within the material is detected by transmission measurements. If, however, the attenuation is very weak ($< 1\%$), small changes in a large signal are to detect yielding a low sensitivity. Consequently, other methods using photo-thermal effects induced by the absorbed radiation energy have been developed. The main advantage of these techniques is a background-free measurement resulting in a high sensitivity¹.

At present, two photo-thermal techniques are applied to measure small bulk absorption in fused silica upon excimer laser irradiation, the calorimetric measurement²⁻⁴ and the laser induced deflection (LID) method⁵. The first measures directly the temperature of the investigated sample to obtain the absorption. Due to contributions of surface absorption, samples of different thickness have to be investigated and the bulk absorption is calculated from the slope of the measured absorption versus sample thickness.

In this paper a pump and probe technique, using the LID method, is introduced where pure bulk absorption values are obtained without surface absorption contributions. The measuring principle of the LID method⁵ is shown in figure 1.

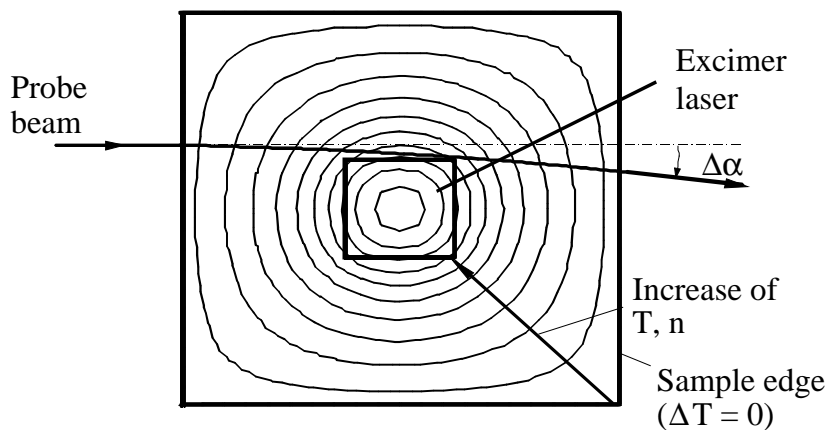


Figure 1: Sketch of the LID measurement principle including calculated isolines of temperature and refractive index as well as the probe beam propagation.

The central excimer laser irradiation induces absorption and subsequently a temperature gradient between irradiated and non-irradiated parts of the material. The resulting refractive index gradient deflects a probe beam that passes the sample perpendicularly to the excimer laser beam. The deflection is a measure for the absorbed pulse energy within the sample

and is detected by a position sensitive diode (PSD). Therefore, together with a calibration procedure, the bulk absorption of the material is obtained. In this paper a compact LID measuring device is introduced that combines an easy experimental handling with a high sensitivity.

2. EXPERIMENTAL SETUP

The conception of a compact design for carrying out LID experiments was derived from a former experimental setup⁵ where a long probe beam path, necessary to obtain a sufficient sensitivity of the setup, yielded some drawbacks concerning the stability and accuracy of the measurements. Therefore, for the new experimental setup, the path length of the deflected beam was shortened and capsuled to overcome the earlier problems. Since reducing the length of the probe beam path results in a smaller beam shift at the PSD and, therefore, a lower sensitivity, two ideas have been successfully implemented in the new setup to compensate for that. In contrast to the former setup, a second beam (the reference beam in the earlier setup⁵) is likewise guided through the sample in the same manner as the probe beam but on the opposite side of the excimer laser irradiated part (fig. 2a). This is realized by a beam splitter (BS) and a mirror (M1). The deflection is registered by a second PSD.

Each of the two PSDs has its own electronics providing a position dependent amplified voltage. The amplitudes of the two signals are added resulting in a doubled sensitivity (differential method). For a further enhancement of the overall sensitivity, the signal of each beam at its PSD is increased by multiple passing the beam through the investigated sample by two mirrors (M2 and M3) in an appropriate alignment (fig. 2b).

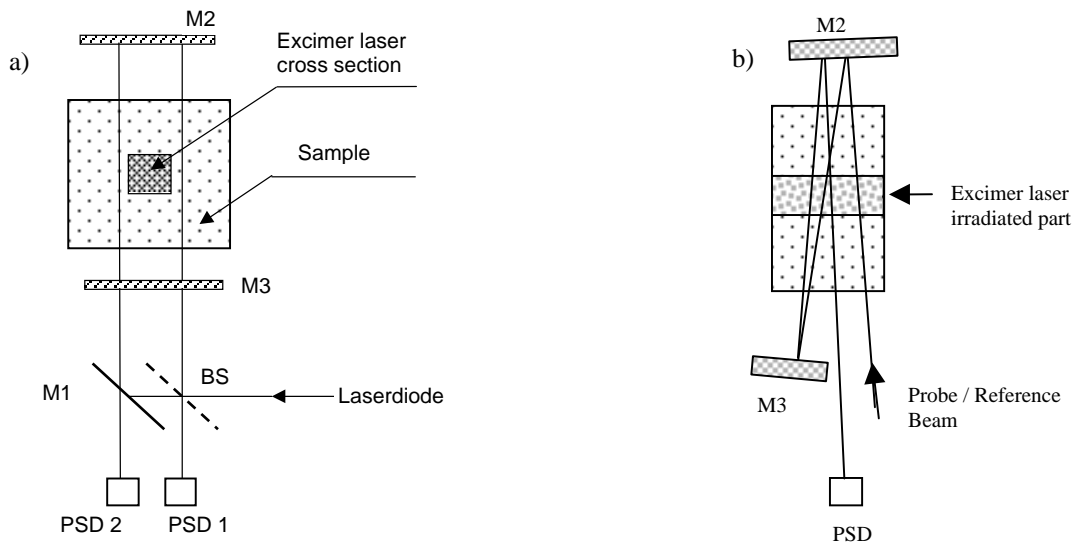


Figure 2: Sketch of the present experimental design showing the new double beam arrangement (a) and the multiple passes of the reference and probe beam, respectively (b).

The current setup allows to guide each beam four times along the irradiated part of the sample and enables the detection of 0.5 mW with an accuracy of 5 %. The number of possible passages is limited by the beam diameter and the thickness of the sample (10 mm for our samples) in a way that a certain distance from both irradiated surfaces is ensured to avoid contributions from surface absorption. Besides the electronics the described experimental setup is converted into a compact device (fig. 3), which is capsuled to reduce influences from air fluctuations.

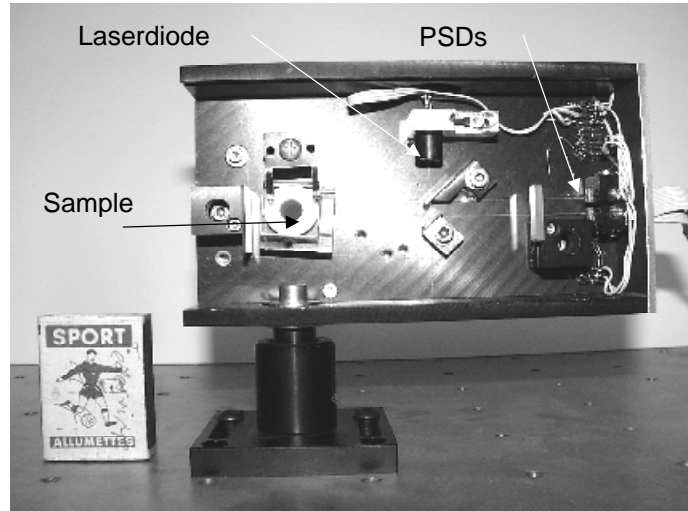


Figure 3: Photograph of the open compact experimental setup.

The irradiation was carried out using an excimer laser LPX 240i (Lambda Physik AG) at a wavelength of 193 nm. The beam was shaped by lenses and an aperture was used to obtain a top-hat like beam size of $5 \times 5 \text{ mm}^2$ at the sample (OH-rich fused silica, type III; cross section $20 \times 20 \text{ mm}^2$, thickness 10 mm). The deflection of the probe beams was registered by an oscilloscope (LeCroy LC334A). A typical record of the LID signal is shown in figure 4 where the voltage difference between the signals upon and without laser irradiation indicates the power absorbed within the sample. The ratio of the absorbed power to the incident mean power yields the absorption of the sample. The latter is equal to the absorption coefficient (cm^{-1}) for the chosen sample thickness of 10 mm.

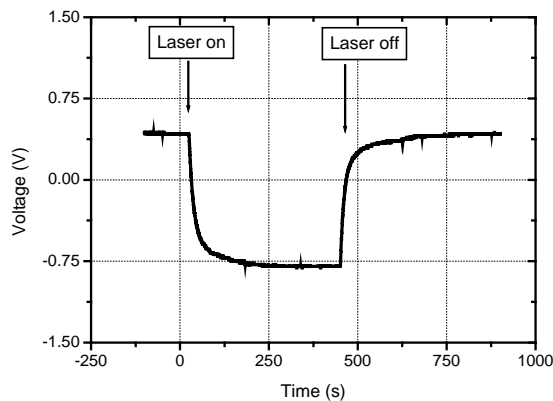


Figure 4: Temporal development of the deflection signal. The applied energy density is 30 mJ/cm^2 at a repetition rate of 150 Hz.

3. EXPERIMENTAL RESULTS

3.1 CALIBRATION PROCEDURE

For calibrating the present compact device, a comfortable procedure was applied⁶. A fused silica sample, having the size of the investigated ones ($20 \times 20 \times 10 \text{ mm}^3$) was drilled centrally and a resistor was fixed therein. By applying a defined voltage and current, the absorption of a certain power within fused silica is simulated and the sensitivity of the setup is obtained.

In order to achieve a higher accuracy and to rule out differences in the sensitivity over a wide range of absorbed power, values between 1 mW and 100 mW have been adjusted by the electrical power at the resistor. Figure 5 shows the voltage, proportional to the deflection, for each applied electrical power. The results are fitted very well by a linear function through zero and its slope gives the sensitivity in mV/mW. In addition, the results confirm the application of the compact setup for a wide range of absorbed power within fused silica samples.

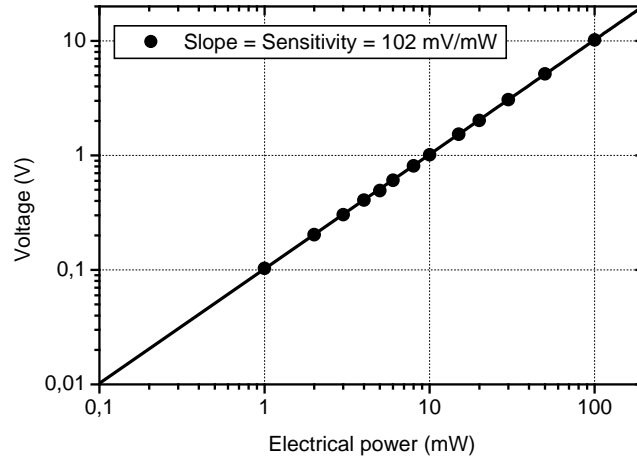


Figure 5: Voltage due to the deflection induced beam shift at the PSDs versus electrical power for the fused silica calibration sample

3.2 ABSORPTION VERSUS REPETITION RATE

The absorption mechanism of fused silica can in a simplified way be expressed as a combination of defect formation (E'- and NBOH-centers) upon laser irradiation and a partial relaxation of defects between the laser pulses⁶⁻⁸. Since the strength of the relaxation depends among others on the time between two consecutive laser pulses, a dependence of the absorption on the repetition rate has to be taken into account in the experiments^{2,7}. To estimate this effect a fused silica sample has been irradiated with a constant energy density of 10 mJ/cm² and repetition rates between 30 and 180 Hz. The results (figure 6) exhibit a dependence of the absorption coefficient which flattens towards higher repetition rates. Thus, it can be assumed that applying repetition rates of about 150 Hz or higher at a constant energy density does not yield significant differences in the measured absorption coefficient.

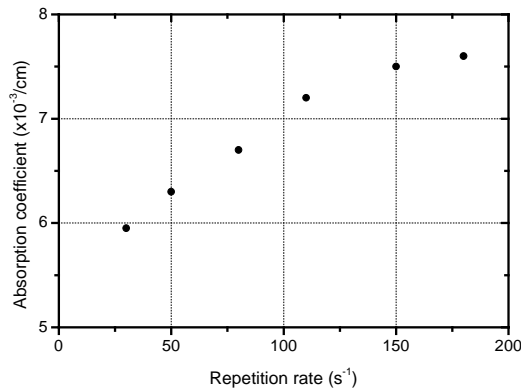


Figure 6: Absorption coefficients of a fused silica sample versus repetition rate. The applied energy density is fixed at 10 mJ/cm².

3.3 ABSORPTION VERSUS LASER PULSE WIDTH

The absorption coefficient of fused silica upon pulsed laser irradiation is composed of contributions from both one- and two-photon absorption processes^{2,4, 6-8}. As a result, the shape of the laser pulse influences the measured absorption due to the dependence of the two-photon process from the temporal behavior of the pulse intensity. Therefore, to evaluate excimer laser pulses a pulse width τ_{IS} (IS = integrated square) is commonly introduced comparing the applied laser pulses to a rectangular laser pulse of equal duration and peak intensity⁹.

Considering our experiments the pulse shape is changed by varying the discharge high voltage of the excimer laser. As an example two different adjustments of the high voltage (18 and 23 kV) have been investigated. The energy density

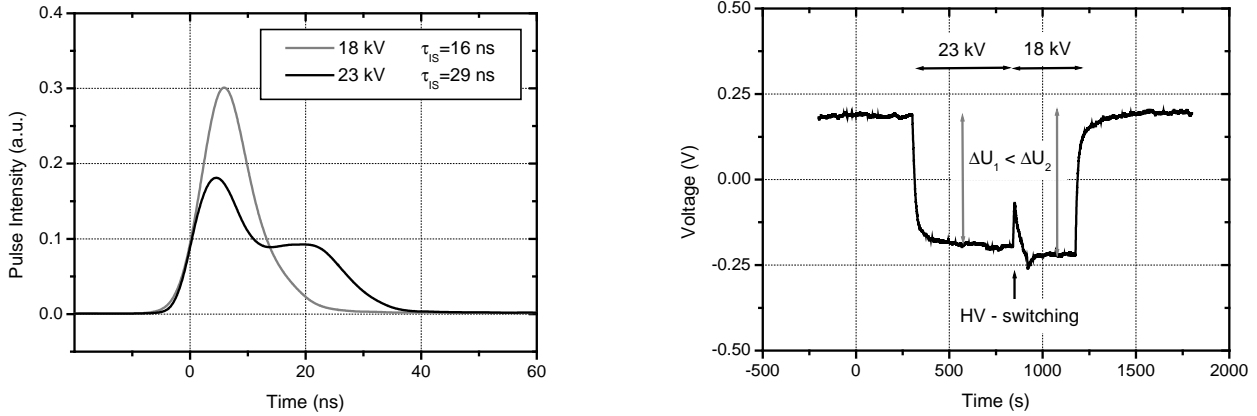


Figure 7: Laser pulse shapes of same energy generated by different discharge high voltages of the excimer laser (a) and the different deflections measured upon laser irradiation of the same sample (b).

was kept constant by using a variable beam attenuator. Figure 7a shows the shape of the different laser pulses and the corresponding integrated square times τ_{IS} . The two laser pulses show a remarkable difference in their pulse widths (16 ns for 18 kV, 29 ns for 23 kV) and subsequently peak intensities. Accordingly, the contribution of nonlinear effects to the overall absorption for both pulse shapes is expected to be in disagreement. The corresponding deflections of the probe beams (figure 7b) verify this and show that the overall absorption for the higher peak intensity pulse exceeds the other by about 15 %. Considering the nonlinear, intensity dependent absorption, the relative change between the two applied pulse widths is even higher.

3.4 ABSORPTION VERSUS SAMPLE THICKNESS

In order to give evidence that the measured absorption is only caused by bulk absorption two fused silica samples with different thickness (10 mm and 20 mm), made from equivalent material, have been investigated at energy densities between 2 and 10 mJ/cm² and a repetition rate of 150 Hz. In contrast to the 10 mm thick sample the distances of the two probe beams from the irradiated sample surface (see fig. 2b) are substantially greater for the 20 mm thick sample. Therefore, if any contribution from the surfaces influences the measured absorption, the latter should be somewhat smaller for the thicker sample. Figure 8, however, shows virtually no difference between the values for the 10 mm and 20 mm thick sample. Therefore we can conclude that no absorption contributions from the irradiated surfaces are contained in the measurement.

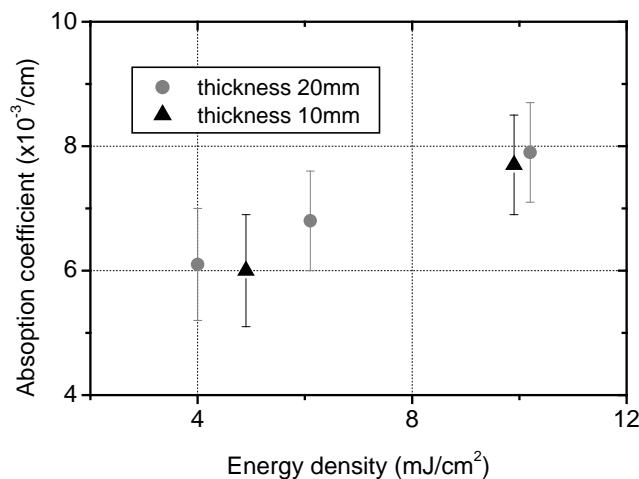


Figure 8: Absorption coefficient versus energy density for 2 equivalent samples of different thickness.

4. SUMMARY

Measuring small absorption by laser induced deflection is shown to be an alternative to the commonly applied calorimetric measurement technique. By applying a compact experimental setup and an electrical calibration it could be demonstrated that only bulk absorption of the samples is measured. Therefore, time consuming experiments to separate bulk from surface absorption effects are avoided.

The results of absorption measurements of OH-rich fused silica upon excimer laser irradiation at 193 nm depend on several excitation parameters like repetition rate and temporal laser pulse shape. Therefore, the understanding of the corresponding processes is necessary to evaluate experimental results with respect to a separation of linear and nonlinear absorption contributions. Furthermore, a detailed consideration of the irradiation parameters is required for a comparison of results between different laboratories.

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