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Incubation of laser ablation in fused silica with 5-fs pulses

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Abstract. The threshold fluences for laser-induced damage of fused silica with single 5-fs pulses from a Ti:sapphire laser system were determined by extrapolating the ablated volume to zero. These thresholds are about 4 times as high as the values previously obtained from multi-shot experiments. This result is interpreted in terms of an irreversible modification of the original material below the single-shot threshold (incubation).

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For several decades, laser-induced breakdown resulting in damage to dielectrics has been the subject of extensive experimental and theoretical investigations [1–3]. Since femtosecond lasers became available as a standard tool in laboratory research, one of the major directions of this research is the machining of transparent substrates. Fused silica, being one of the most widely used optical materials, has been studied extensively with nanosecond, picosecond [4], and femtosecond [5–7] laser pulses.

One of the main disputes over the experiments with longer pulses (> ps), whether avalanche or multiphoton ionization is responsible for the generation of free carriers in transparent dielectrics, was recently extended to the femtosecond range of laser pulse durations. In the course of this discussion various experiments are compared with each other, often regardless of whether they have been achieved by single or by multiple pulses on the same sample spot. However, it was discovered earlier that a so-called N-on-one effect exists, which means that successive irradiation of the same sample spot alters the damage threshold of the material [8, 9]. In this paper, we compare the results of single-shot and multi-shot experiments performed with the same laser on the same sample in order to get insight into this phenomena in the sub-10-fs regime.

The Ti:sapphire laser system used in the experiments delivers femtosecond pulses with the following parameters [10]: pulse duration $\tau = 5$ fs, center wavelength $\lambda_{\text{center}} = 780$ nm, repetition rate $= 1$ kHz, maximum output energy $\text{E}_{\text{max}} = 500\,\mu\text{J}$, and beam quality parameter $M^2_{x,y} < 1.1$. For the single-shot experiments described in this paper, the sample (Corning 7940, 0.2 mm thick) was placed in a vacuum chamber (pressure $\approx 6 \times 10^{-4}$mbar) and irradiated through a 0.5-mm-thick glass window with a long-focal-length silver mirror (ROC $= 1000$ mm). The transmitted beam was steered out of the vacuum chamber and used for in-situ inspection of the irradiated spot. The sample positioned by a remotely controlled translation stage was exposed to a series of single pulses with varying above-threshold energies.

The post-experimental evaluation of the ablation results has been undertaken by using both a light microscope and, because of the low depth of the generated craters ($< 1\,\mu$m), an atomic force microscope (AFM, Digital Instruments, Dimension 3000 SPM) operated in tapping mode. A typical picture of an ablation crater is shown in Fig. 1. This hole was produced at a laser fluence of $F = 9.5\,\text{J/cm}^2$, which will be shown below to be about twice the damage threshold fluence of this material for single pulses. The 5-fs crater shows a very smooth surface compared to cavities generated with single pulses of several hundred femtosecond and picosecond duration.

The ablated volumes for a range of laser fluences above the damage threshold are represented by the dots shown in Fig. 2. Calculating a linear regression (solid line in Fig. 2) and extrapolating this line to an ablated volume of zero yields a threshold fluence of $F_{\text{th}} = 4.9\,\text{J/cm}^2$ for fused silica irradiated with 780 nm pulses of 5 fs duration.

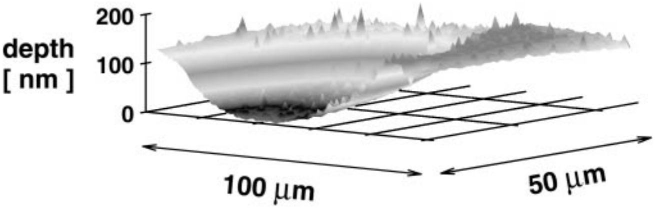

Fig. 1. AFM picture of a laser-generated hole in fused silica, obtained with a single pulse of $\lambda_{\text{center}} = 780$ nm, $\tau = 5$ fs, $F = 9.5\,\text{J/cm}^2$

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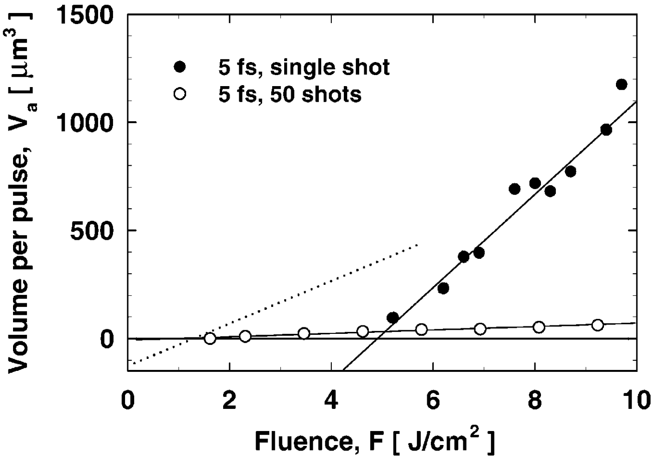

Fig. 2. Ablated volume per pulse V_a versus laser fluence F for craters generated by a single laser pulse and by 50 laser pulses on the same sample spot. The *straight lines* are obtained from linear regression. The intersection of the regression line with the horizontal axis ($V_a = 0$) yields the threshold fluence F_{th} for optical damage. In the case of 50 pulses per spot only data obtained at $F \leq 6\,J/cm^2$ were considered for regression to avoid the influence of plasma formation in air. The *dotted line* is the tenfold enhanced regression line for the multi-shot measurements to allow comparison with the single-shot data

We compared results from earlier experiments with similar experimental parameters, but with each site irradiated with 50 consecutive pulses [7]; those results are shown as open circles in Fig. 2. To determine the threshold we restricted the linear regression to fluences below $6\,J/cm^2$ because these experiments were conducted at atmospheric pressure. By comparing the results with experiments in vacuum and by checking the interferometric autocorrelation of the focused pulses it was established that there is no significant distortion of the 5-fs pulses by plasma formation in air up to this level. On the other hand, the measured values above this level deviate only slightly from the regression line. The single-shot experiments had to be carried out in vacuum because of the higher damage threshold fluence.

The data depicted in Fig. 2 reveal that both the threshold fluence for ablation and the (average) ablated volume per pulse are significantly different for multi-shot and single-shot excitation. That means, for a certain laser fluence above both thresholds a single pulse ablates much more material than a pulse in a series of 50 pulses does on an average. This suggests, that the ablated volume per pulse decreases dramatically during a pulse train on the same sample spot.

The significant difference between single-shot and multi-shot ablation thresholds implies a pre-ablation modification of the irradiated material when laser fluences between the two threshold fluences are used. Here, the first laser pulse encounters the original fused silica and already alters the material. This alteration might be, for instance, the formation of color centers as summarized in [11]. All the possibilities of material modification addressed by Bäuerle [11] result in the generation of electronic states within the forbidden gap, which enhance light absorption of the sample compared to the intrinsic material exhibiting only multiphoton absorption. This increased absorption eventually leads to

1. a lower damage threshold in the case of multi-pulse irradiation of the same spot and
2. a lower penetration depth of the laser radiation resulting in a smaller ablation depth and ablation volume.

These simple qualitative considerations are in accordance with our experimental observations. This suggests that incubation effects take place in fused silica even at pulse durations as short as 5 fs. This finding is also supported by the observation that single-pulse irradiation of fused silica with a fluence between these two thresholds leads to coloring of the sample.

We have compared the damage threshold fluences for single-shot and multi-shot experiments with 5-fs pulses on fused silica and found them to be about four times enhanced for the single-shot case. Furthermore, we noticed a significantly decreased ablation rate for pre-irradiated fused silica. These observations lead us to the conclusion that incubation effects alter the optical properties of the material.

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