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Laser-induced damage in SiO_2 and CaF_2 with picosecond and femtosecond laser pulses

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Abstract. Single- and multiple-shot damage thresholds and plasma-emission thresholds for fused silica and CaF_2 are reported for 790 nm photons as a function of laser pulse width (190 fs – 4.5 ps). The results are compared with single-shot plasma-emission measurements [1] and with multiple-shot damage measurements [2]. Both the damage threshold and the plasma-emission threshold are shown to decrease with decreasing pulse width over the entire pulse-width range investigated.

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The interaction of ultrashort (< ps) laser pulses with optically transparent materials is a subject which is becoming increasingly important due to the rapid advances in laser technology. Apart from the fundamental questions which have to be answered concerning the interaction mechanisms and energy transport phenomena occuring in the materials there are also a number of practical considerations which arise. For example, damage to optical components is often the limiting factor in the performance of high peak power/short pulse laser systems. A more positive aspect of the same behaviour is that short pulse lasers may prove to be flexible tools for micro-structuring a wide range of materials which cannot be adequately treated with nanosecond laser pulses or etching methods.

Two recent publications have investigated laser-induced breakdown or damage in optically transparent materials as a function of laser pulse length τ [1,2]. Du et al. [1] studied the threshold fluence (energy per area) for plasma emission from fused silica for pulse lengths in the range from 150 fs to 7 ns. They found the expected $\tau^{1/2}$ dependence of the threshold fluence for long pulses (> 10 ps) where the process is controlled by the rate of thermal conduction through the lattice [3]. Similar dependencies have been found in a number of experiments with a variety of dielectric materials using laser pulses in the range from 100 ps to 10 ns [4]. However, for pulse lengths < 10 ps they reported an increase in the threshold fluence with decreasing pulse length, scaling as τ^{-1}. Stuart et al. [2] determined the damage threshold for fused silica and CaF_2 with pulses in the range 270 fs $\leq \tau \leq$ 1 ns. Their definition of the threshold fluence was the value at which visible permanent modifications to the surface could be observed with a Nomarski microscope. In contrast to Du et al., they did not observe an increase in the damage threshold for pulses < 10 ps. However, they did observe a deviation from the $\tau^{1/2}$ behaviour for $\tau <$ 20 ps with the rate of decrease of the damage threshold decreasing as the pulse length was reduced. Both groups interpreted their results in terms of theoretical models in which electrons, initially produced by multiphoton ionization by the short pulses, are further heated resulting in avalanche ionization and rapid plasma formation. The increase in the plasma threshold observed in the experiments of Du et al. was attributed to the saturation of the ionization rate per unit length for electric fields on the order of a few MV/cm [1].

The discrepancy in the results of the two groups may be due to a number of reasons. The most obvious differences in the experiments (apart from the diagnostic methods used to determine the occurence of breakdown or damage) are, firstly, that Du et al. used single-shot measurements, whereas Stuart et al. used multiple pulses of a given fluence on each site and, secondly, that different wavelengths were used (780 nm and 1053 nm, respectively). The use of multiple pulses to determine threshold values for damage is particularly questionable since incubation effects may serve to reduce this value.

We have determined the damage thresholds and plasma-emission thresholds for fused silica and CaF_2 as a function of pulse length at a wavelength of 790 nm. The measurements were carried out for single-shot experiments and, to determine the possible role of incubation effects, for experiments in which five laser shots per site were used. The experiments were carried out both in air and under vacuum conditions ($\leq 10^{-5}$ mbar). The short-pulse laser used in our experiment was a Ti:Sapphire oscillator-amplifier (50 Hz) system based on the Chirped-Pulse-Amplification technique (CPA) [5]. As in the other experiments [1, 2], CPA allowed us to vary the pulse length without changing other important parameters. The laser pulse was focused by an f = 58 mm lens onto the sample surface. The laser spot size (1/e^2) was 500 μm^2 (spot diameter 25 μm) as determined with a beam profiler. The fused-silica samples were 1 mm thick and pol-

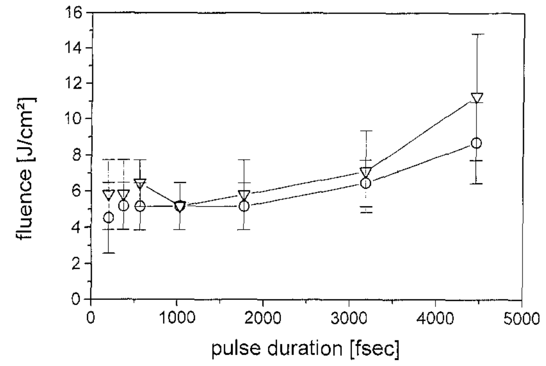

Fig. 1. Experimental damage thresholds obtained with a Nomarski microscope for fused silica under vacuum conditions for a wavelength of 790 nm. *Triangles*: single-shot measurements; *circles*: 5-shot measurements. Thresholds obtained from AFM and REM analysis as well as from in situ reflection measurements gave identical results within the experimental error bars

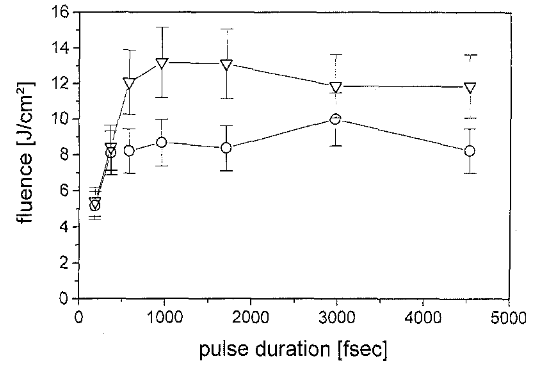

Fig. 2. Thresholds for plasma emission from fused silica under vacuum conditions. *Triangles*: single-shot measurements; *circles*: 5-shot measurements

ished on both sides. The CaF_2 samples (Korth, Germany) were polished on the front surface. The front-surface damage threshold was determined ex situ by using a Nomarski optical microscope, an atomic force microscope and an electron microscope and in situ by monitoring the reflection from the surface. Rear-surface damage was not observed. The results obtained from all four methods were identical within the experimental error bars. The plasma emission from the focal region was collected by a lens and focused onto a photodiode with appropriate filters. To minimise statistical uncertainty, the measurements were carried out for 5–10 individual spots at each value of the laser pulse length and fluence.

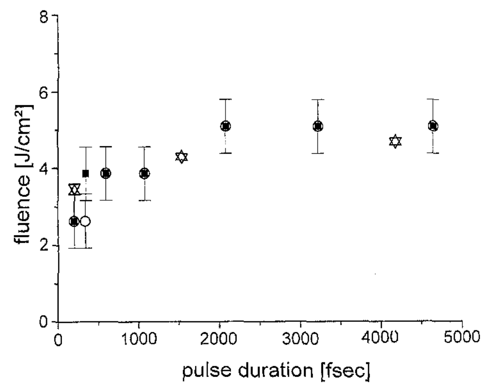

Fig. 3. Damage thresholds for CaF_2 obtained under vacuum conditions *Squares*: physical damage for single-shot measurements as determined by REM analysis; *circles*: physical damage for 5-shot measurements; *up-triangles*: plasma emission for single-shot measurements; *down-triangles*: plasma emission for 5-shot measurements

The results we obtained for fused silica are shown in Figs. 1 and 2. The trend observed for the optical damage threshold (Fig. 1) is very similar to that observed by Stuart et al. [2][1]. There is a gradual decrease of the damage threshold as the pulse width is reduced from 5 to 1 ps. For pulse widths below 500 fs there is an indication that the rate of decrease of the threshold increases again. This would be in agreement with the model proposed by Stuart et al. [2]. There is very little difference between the single-shot and multiple-shot measurements although the multiple-shot results lie consistently below those of the single-shot experiments. The thresholds for plasma emission are shown in Fig. 2. Here, we see a very similar trend with the rapid fall in threshold for the shortest laser pulse widths being more noticeable than in the physical damage measurements. The significantly higher threshold values which we give for the single-shot measurements reflect the sensitivity of our detector (the plasma signal for the single-shot measurements under vacuum conditions was very weak). In contrast to Du et al. [1], we do not see an increase in the plasma-emission threshold for the very short laser pulses. The plasma-emission threshold is seen to correlate very closely with the physical damage thresholds.

In Fig. 3, we compare the results for CaF_2. Again, our results follow the same trend as those of Stuart et al. and do not show any evidence of an increase in the damage threshold for pulse lengths below 10 ps. There are no plasma-emission results available in the literature to compare with.

In conclusion, we have combined a variety of both in situ and ex situ techniques to determine the single-shot damage thresholds for fused silica and CaF_2 as a function of laser pulse width. All methods confirm the trend shown by Stuart et al. [2] for multiple-shot optical damage on the same materials (but at a different wavelength). In addition, our results show evidence of a more rapid decrease in the threshold fluence for pulse widths below ca. 500 fs, in agreement with the model suggested in [2]. We find no evidence of a rise in threshold fluence for plasma emission at very short pulse widths as reported by Du et al. for fused silica [1].

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[1] The absolute values of the threshold fluences in our experiments are a factor of two higher than those given by Stuart et al. [2] for both fused silica and CaF_2