

FROM PS TO FS: DEPENDENCE OF THE MATERIAL REMOVAL RATE AND THE SURFACE QUALITY ON THE PULSE DURATION FOR METALS, SEMICONDUCTORS AND OXIDES

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Abstract

Ultra short laser pulses in the ps or fs regime are used, when high requirements concerning machining quality are demanded. However, beside the quality also the process efficiency denotes a key factor for the successful transfer of this technology into real industrial applications and the process efficiency is mainly influenced by the removal rate. In this work the results of a study about the influence of the pulse duration onto the removal rate for pulses from 10 ps down to 250 fs will be presented. The investigated materials were copper, stainless steel, silicon, germanium, zirconium oxide (ZrO₂), polycrystalline diamond (PCD) fused silica and soda lime glass. An increase of the removal rate with shorter pulse durations was observed for all materials except the two glasses. A very special behaviour with a sharp change between two regimes with different ablation processes was observed for soda lime glasses for both wavelengths.

Introduction

Ultra short pulsed systems show clear advantages concerning machining quality, heat affected zone, debris, etc [1-4]. But even if the excellent machining quality is one of the key advantages of ultra short pulsed systems, it may be more cost effective to use a Q-Switched system and to accept the reduced quality and additional costly post processing steps. One possibility to further increase the competitiveness of ultra short pulsed systems may be the change to fiber based amplifier technologies. Without CPA technology the pulse duration of these systems is expected to be in the range of several tens ps [5-7]. For metals the ablation efficiency significantly drops by about a factor of 5 when the pulse duration is raised from 10 ps up to 50 ps [8,9], for non metals this drop is less pronounced but still present [10]. The characteristics of the measured ablation efficiency as a function of the pulse duration implies, that the efficiency could increase

when the pulse duration is reduced from 10 ps into the sub ps regime. Results corroborating this belief have been reported in [11-13]. For dielectrics in [14] an increase of the efficiency is reported when the pulse duration is reduced 300 fs – 7 fs. This all encourages the development of fiber based ultra short pulsed systems in the fs-regime which can be driven to high average powers by succeeding amplifier stages; recently more than 1 kW average power with fs – laser pulses were demonstrated [15,16]. From this point of view sub – ps systems could to be very attractive for laser micro processing.

Ablation Model

Theory

For ultra short pulses the heat-transfer process in metals is described with the two temperature model [1,17-21] where the temperatures of the electrons and the lattice are treated separately. The results of the model and the experiments show, that the ablation depth z_{abl} can be written as a function of the fluence ϕ (pulse energy per unit area):

$$z_{abl} = \delta \cdot \ln\left(\frac{\phi}{\phi_{th}}\right) \quad (1)$$

with ϕ_{th} the threshold fluence and δ the energy penetration depth. Frequently two different ablation regimes are reported [19,22]: firstly the low fluence regime where the optical penetration depth dominates and secondly the high fluence regime where the energy transport is dominated by the heat diffusion of the hot electrons. In [23] it is shown, that for a top hat beam the efficiency of the ablation process depends on the ratio between the threshold fluence and the applied fluence ϕ_{th}/ϕ . The efficiency shows a maximum value of 1/e, i.e. about 37%. At this point of maximum efficiency the ablated volume per pulse reads:

$$\Delta V_{pulse} = \pi \cdot w_0^2 \cdot \delta \quad (2)$$

From this one can calculate the maximum removal rate per average power which reads for a top hat beam:

$$\frac{\dot{V}_{\max}}{P_{av}} = \eta_{\max} \cdot \frac{\delta}{\phi_{th}} = \frac{1}{e} \cdot \frac{\delta}{\phi_{th}} \quad (3)$$

The removal rate finally depends on the energy penetration depth δ and the threshold fluence ϕ_{th} .

Ablation for a Gaussian beam

Similar calculations have been done for a Gaussian shaped beam as emitted by most ultra short pulsed systems [24,25]. Again a maximum removal rate per average power (ablation efficiency) is observed:

$$\frac{\dot{V}_{\max}}{P_{av}} = \frac{2}{e^2} \cdot \frac{\delta}{\phi_{th}} \quad (4)$$

The ablated volume per pulse at this optimum points is again given by (2) i.e. this maximum efficiency is again obtained at a corresponding fluence. It has to be pointed out that the maximum value of the removal rate (4) is only obtained at this optimum point. A general expression for the removal rate of a Gaussian beam is also developed in [24,25] and reads:

$$\dot{V} = \frac{1}{4} \cdot \pi \cdot w_0^2 \cdot \delta \cdot f \cdot \ln^2 \left(\frac{\phi_0}{\phi_{th}} \right) \quad (5)$$

With ϕ_0 the peak fluence in the centre of the Gaussian beam:

$$\phi(r) = \phi_0 \cdot e^{-2 \cdot \frac{r^2}{w_0^2}} \quad \text{with:} \quad \phi_0 = \frac{2 \cdot E_p}{\pi \cdot w_0^2} \quad (6)$$

All these considerations clearly show that the ablation process can be optimized and that the maximum removal rate is finally given by the threshold fluence ϕ_{th} and the energy penetration depth δ . For metals and pulses longer than 10 ps the threshold fluence begins to increase [8-10,19]. But beside the threshold fluence also the penetration depth δ has an influence onto the maximum volume ablation rate. For pulse durations in the range from 10 ps to 50 ps the value of δ decreases with increasing pulse duration [8-10,19]. Therefore the maximum removal rate in general drops significantly down when the pulse duration is raised. The situation changes for shorter pulses. From the literature [3] one expects that the threshold fluence will rest constant when the pulse duration becomes shorter than about 10 ps. If these pulses would lead to higher ablation rates this can be caused only by a higher penetration depth δ for shorter pulses.

Two remarks have to be placed here: First, Due to incubation effects the threshold fluence may strongly depend on the number of pulses applied, which is described for metals in [8,9,23,26,27]. Additionally it was found in that also the energy penetration depth shows an incubation effect of the same kind. The maximum removal rate may therefore also strongly depend on the number of pulses applied; more details are given in [8-10,23].

Second, often in the literature two different materials, pulse durations, wavelengths etc. are compared. This is normally done with identical parameters such as repetition rate, focus radius and pulse energy. But one has to have in mind that the efficiency of the ablation process is strongly influenced by the threshold fluence and the penetration depth and that these two measures depend on many parameters. Figure 1 shows the removal rate for two different sets of ϕ_{th} and δ , an average power of 1 W and a spot radius of 20 μm . If the ablation rates are compared at equal repetition rates the blue curve will dominate up to about 200 kHz and afterwards the red curve will be the more efficient one i.e. depending on the chosen repetition rate one set of parameters can be more efficient than the other and vice versa. This might be a cause that sometimes contradictory results concerning the ablation rates are reported in the literature. It becomes clear, that a fair comparison between different situations should be done by deducing the maximum ablation rate. This can be done by experimentally deducing the threshold fluence and the penetration depth.

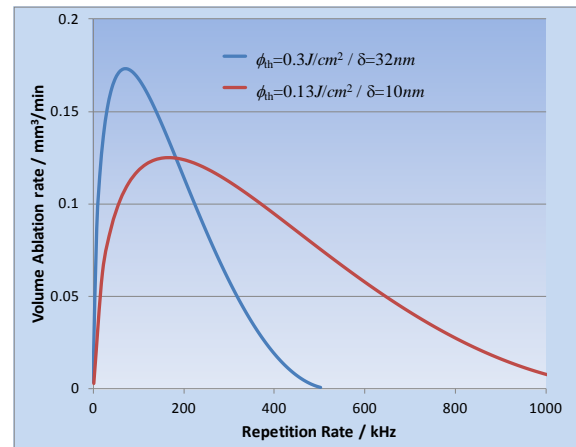


Fig 1: Removal rate for an average power of 1 W, a spot radius of 20 μm for two different sets of ϕ_{th} and δ .

Experimental Set-Up

The radiation of the used laser source was guided via a $\lambda/4$ -plate (to generate a circular polarized beam) and folding mirrors through a beam expander into a galvo

scanning head where it was focused by an f-theta objective onto the target. For the experiments three different laser systems were used:

The experiments for 10 *ps* to 50 *ps* were performed with a DUETTO™ (Time Bandwidth Products, Switzerland) ps-laser system working at a wavelength of 1064 *nm* with a pulse duration of about 10 *ps*. By introducing corresponding etalons into the master oscillator the pulse duration was raised to 20 *ps*, 30 *ps* and 50 *ps*. The pulse duration was controlled with an autocorrelator measurement whenever the etalon was changed.

A SATSUMA (Amplitude Systemes, France) fs-laser, working at a wavelength of 1030 *nm*, was used for the experiments with pulse durations between 500 *fs* and 10 *ps* for copper, stainless steel and silicon. The pulse duration was changed from 500 *fs* to higher values by detuning the pulse compressor.

For shorter pulses and second harmonic as well, a PHAROS laser system (Light Conversion, Lithuania) was used. Here the minimum pulse duration was specified to be 223 *fs* and it could again be changed up to 10 *ps* by detuning the pulse compressor. This system was used for the experiments at 1026 *nm* and 513 *nm* for copper, silicon, germanium, ZrO₂, PCD, fused silica and soda lime glass. The frequency conversion unit was optimized for the shortest pulse durations and therefore the pulse energy in second harmonic significantly dropped down for longer pulse durations and the experiments with 513 *nm* wavelength could often only be done up to a pulse duration of 1 or 2 *ps*.

Each system included an additional picker after the last amplifier stage as a pulse on demand (POD) option to be able to reduce the repetition rate down to single pulses. Also the control of the pulse energy going with the average power was done after the last amplifier stage. It was realized with polarizer and $\lambda/2$ -plates to guarantee constant beam parameters. For all different situations (laser system and wavelength) the beam quality and spot size was measured with a rotating slit beam profiler. All systems emitted a nearly Gaussian shaped beam.

In order to deduce ϕ_h and δ as a function of the pulse duration and the number of pulses applied, series with single ablated craters were generated for each set of these parameters, i.e. 1,2,4,...,512 pulses and different pulse durations. For one series the pulse energy was raised from below up to multiples of the threshold. To avoid thermal accumulation effects the repetition rate was set to 1 *kHz* or less with the POD. The crater depths in the centre were deduced with a laser scanning microscope. These results were compared with the theoretical crater depth by introducing the

peak fluence (6) into (1). The two parameters ϕ_h and δ were then deduced by a least square fit to the experimentally obtained data. This analysis was done for copper, steel, doped and undoped silicon and germanium. Especially at low pulse energies i.e. low fluences or low number of pulses the deduction of the crater depth became extensive and sometimes almost impossible. On the other side, too high fluences and pulse numbers lead to deep craters where it was no longer possible to measure the depth in the center.

For the semiconductors a massive crater cone formation [28] constrained the exact measurement of the crater depths. Also for the porous materials ZrO₂ and PCD as well as for the glasses the upper kind of study could not be performed with the desired accuracy. Therefore by the experiments with the PHAROS system the removal rate was additionally determined by ablating squares. The squares were built up by machining 37 hatched slices with a pulse to pulse and line distance of 8 μm for 1026 *nm* and 3 μm for 513 *nm*, respectively. The removal rate can then be deduced by measuring the depth of the square with the laser scanning microscope. Together with laser and processing parameters the removal rate can exactly be calculated. Here only the real laser on time was considered and the jump time is neglected, i.e. the calculated removal rate is a physical one and the real rate, also containing the process strategy, will be lower.

Results

Copper

For Cu-DHP (in US: C12 200) the deduced values of ϕ_h and δ for 128 pulses and for all pulse durations and wavelengths are summarized in Fig. 2. For the IR radiation only the second ablation regime from the two reported in [19,22] was observed. The situation changes for the green radiation where the two regimes could clearly be separated. As expected for both wavelengths ϕ_h do not significantly change for pulses shorter than 10 *ps*. The situation changes for δ . For IR-radiation the energy penetrations depth continuously increases with shorter pulse durations. This increase is still present for green between 10 *ps* and 1 *ps*, but it is much less pronounced and δ rests almost constant for pulses shorter than 1 *ps*.

Based on these parameters the maximum removal rate can be calculated by (4). The corresponding results are summarized in Fig. 3: For IR the removal rate continuously increases from 50 *ps* pulse duration to 500 *fs*; between 10 *ps* and 500 *fs* this increase amounts about 75% (compared to the value at 10 *ps*). For the 2nd regime in green still an increase is observed, but it only

amounts about 30%. The maximum rates for the 1st regime are much lower. Concerning the maximum removal rate the obtained values for IR and green (2nd regime) are in the same range for the pulse durations between 10 ps and 223 fs i.e. for the ablation efficiency in copper there is no advantage to go to a shorter wavelength.

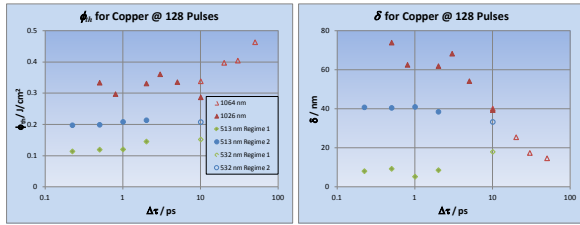


Fig. 2: Threshold fluence and penetration depth for copper as a function of the pulse duration and the wavelength.

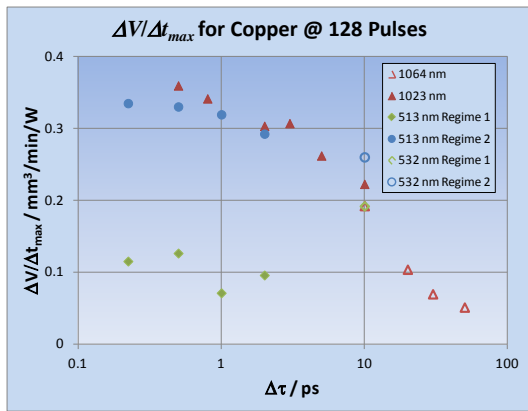


Fig. 3: Maximum removal rate for copper as a function of the pulse duration and the wavelength.

The surface quality is shown in Fig. 4; in the upper part for 1026 nm and in the lower part for 513 nm. The squares were generated with a laser repetition rate of 200 kHz and a peak fluence (ϕ) of about 0.85 J/cm² for 1026 nm and 513 nm as well. The spot radius amounted 19.5 μ m for 1026 nm and 7.5 μ m for 513 nm. Due to the decreasing conversion efficiency the pulse duration was limited to 2 ps for 513 nm. As it can be seen from the SEM-pictures the surface quality is not affected by the pulse duration, neither for 1026 nm nor for 513 nm. For the green wavelength the surface roughness seem to be better compared to IR but this is mainly explained by the smaller focus radius and hatch distance.

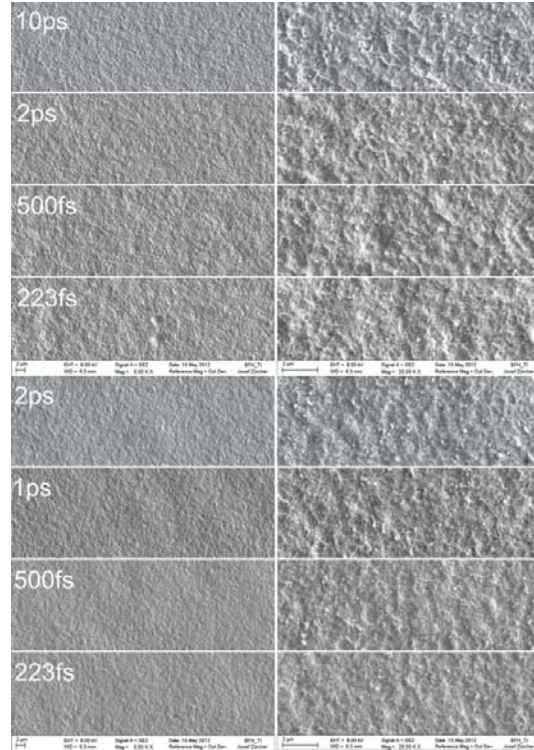


Fig. 4: Surface of ablated squares in copper for 1026 nm (up) and 513 nm (below).

Stainless Steel

Stainless steel 1.4301 (in US: AISI 304) was only investigated for a wavelength of 1064 nm between 10 ps and 50 ps and for 1030 nm between 10 ps and 500 fs. Fig. 5 shows the results of the study for 256 pulses. For long ps-pulses ϕ_{th} strongly depends on the pulse duration. For pulse durations equal or shorter than 10 ps ϕ_{th} rests again constant. In contrast δ significantly drops when the pulse duration is raised from 500 fs to 10 and 20 ps. For longer pulses this drop is stopped and δ even slightly raises for a pulse duration of 50 ps. This small increase could be an indication that for this pulse duration the conventional heat conduction begins to dominate the energy transport in the material. In this regime ϕ_{th} and δ are expected to depend on the square root of the pulse duration.

From these values again the maximum removal rate (4) is calculated and shown in Fig. 6. As for copper the removal rate increases with decreasing pulse duration. Compared to the value at 10 ps it amounts only about 20% at 50 ps, a strong drop. In contrast for the pulse duration of 500 fs an increase of about 65% is obtained.

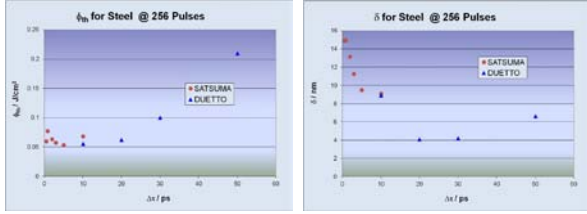


Fig. 5: Threshold fluence and energy penetration depth for stainless steel as a function of the pulse duration.

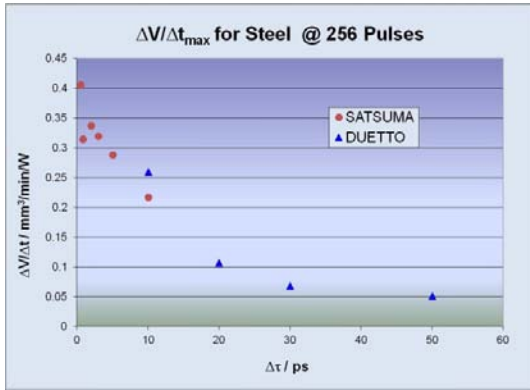


Fig. 6: Maximum removal rate for stainless steel as a function of the pulse duration.

Fig. 7 shows the SEM image with detail view of marked lines machined with an average power of 170 mW, a spot radius of 13.5 μm , a repetition rate of 100 kHz, a marking speed of 160 mm/s and 15 repeats. Please note that the scale for the upper images differs for 10 ps and 500 fs and the apparent smaller line width for 500 fs is due to this different scale. In the lower detail view small “worms of melted material” are observed between the ripples. These “worms” are much less pronounced for the pulse duration of 500 fs.

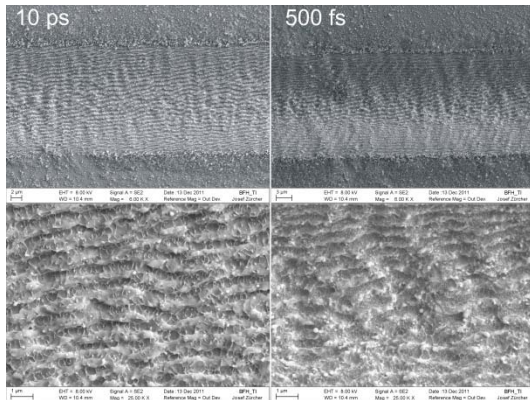


Fig. 7: SEM images with detail view of marked lines in steel.

Undoped Silicon

For undoped silicon again an almost constant ϕ_{th} is observed for 1030 nm. For 513 nm the threshold seems to have a tendency to lower values at shorter pulse durations but this is not confirmed by the other numbers of applied pulses. For both wavelengths the penetration depth δ increases with decreasing pulse duration. This behaviour will again lead to an increased removal rate for shorter pulse durations which is confirmed, see Fig. 9, by the obtained removal rates from the ablated squares. Here the wavelength of 513 nm is more efficient. Introducing the values of the threshold and the penetration depth into (4) will lead to a contrary result were the IR radiation should show higher removal rates. The reason for this difference is not clear yet and further investigations have to be done.

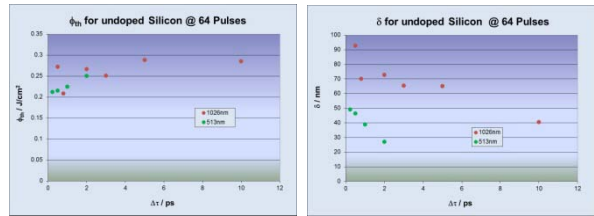


Fig. 8: Threshold fluence and energy penetration depth for undoped silicon.

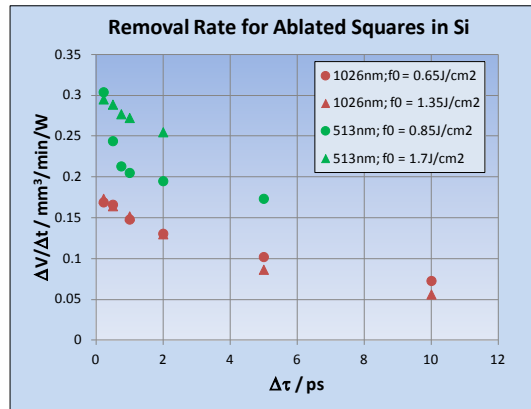


Fig. 9: Removal rate obtained from the ablated squares for the different wavelengths and two different fluences.

The pulse durations has an influence onto the crater and cone formation at the bottom of the ablated squares, as shown in Fig.10. These squares were generated with a repetition rate of 100 kHz and a peak fluence of about 2.7 J/cm² (left) and 5.3 J/cm² (right). For a pulse duration of 10 ps the surface is flat for both fluences. For the higher fluence the crater and cone formations starts at a pulse duration of about 5 ps

whereas it is already present for the lower fluence. For lower pulse durations the crater and cone formation seems to be unaffected. The fluence has an influence onto the dimensions of the crater and cones in the way that higher fluences lead do bigger cones.

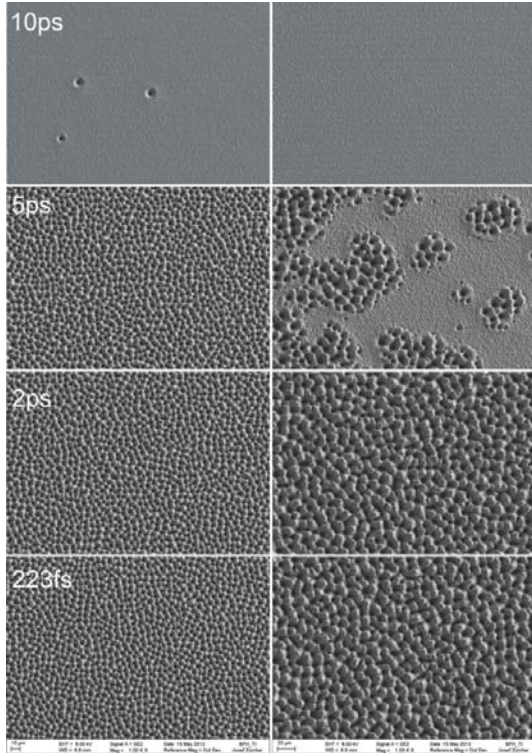


Fig. 10: Surface of the ablated squares in undoped silicon for different pulse durations and peak fluencies; left: $2.7 J/cm^2$ right: $5.3 J/cm^2$

Germanium

The threshold fluence ϕ_{th} and the penetration depth δ were only investigated for $513 nm$ and pulse durations between $223 fs$ and $2 ps$. The results show the same tendency as already obtained for silicon e.g. for 64 pulses the threshold fluence is in the range of $0.2 J/cm^2$ and the penetration depth changes from $80 nm$ to $120 nm$ when the pulse duration is reduced from $2 ps$ to $223 fs$. The corresponding maximum removal rates are between 0.75 and $0.9 mm^3/min/W$ which is about twice the value from silicon. This higher value is also confirmed by the removal rate measured from the ablated squares shown in Fig. 11. The increase in the ablation rate for shorter pulse durations is still present but is much less pronounced than for silicon and again for $513 nm$ the removal rate is up to 2 times higher than for $1026 nm$.

In contrast to silicon, crater and cone formation is also observed for the pulse duration of $10 ps$. This is

illustrated in Fig.12 for the peak fluences of $4 J/cm^2$, $2.7 J/cm^2$, $1.35 J/cm^2$ and $0.67 J/cm^2$ and the pulse durations of $223 fs$ (left) and $10 ps$ (right). The same dependence of the cone dimension on the fluence as for silicon is observed. For the two high fluences some kind of small deep holes in the cones are observed for $223 fs$ pulse duration.

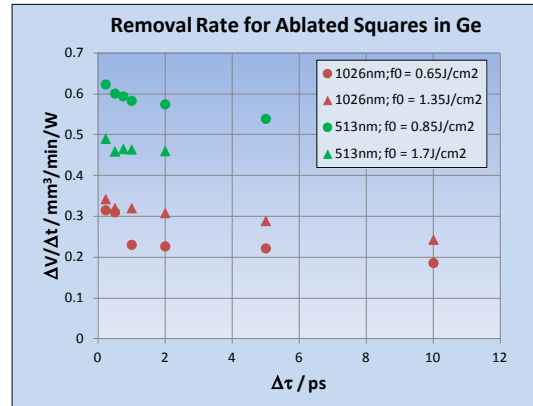


Fig. 11: Removal rate obtained from the ablated squares for the different wavelengths and two different fluences.

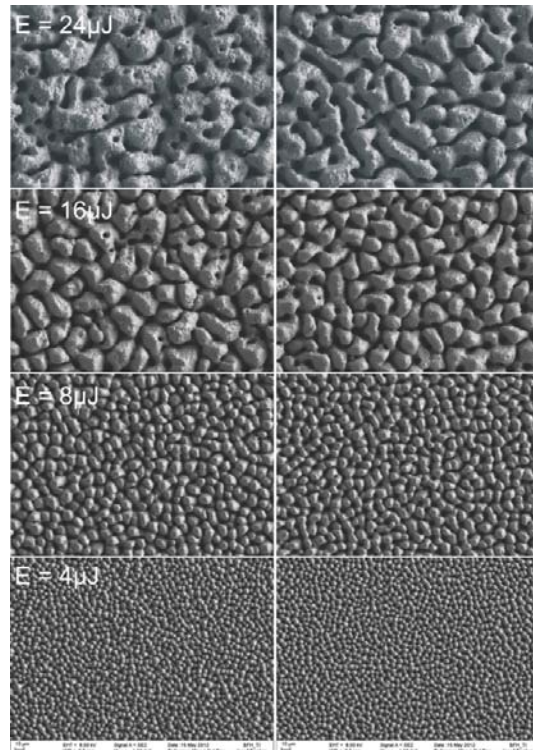


Fig. 12: Surface of the ablated squares in germanium for $223 fs$ (left) and $10 ps$ (right) pulses and the four peak fluencies (top down) $4 J/cm^2$, $2.7 J/cm^2$, $1.35 J/cm^2$ and $0.67 J/cm^2$.

PCD

For pulse durations shorter than 10 ps the removal rate was only deduced via the ablated squares and the values are compared with the ones obtained from earlier work [10] (see Fig. 13). For IR radiation a continuous increase is observed when the pulse duration is reduced from 50 ps to 223 fs. This increase is higher for ps pulses and becomes low in fs regime. For 513 nm the removal rate rests almost constant and equals the one obtained for 1023 nm. In general the obtained removal rate is very low compared to other materials. Almost no difference in the surface quality was observed in the full range of the investigated pulse durations.

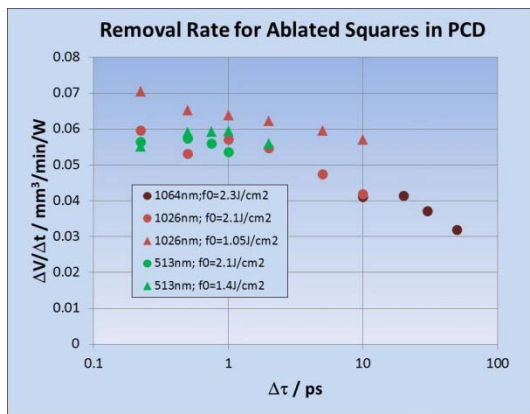


Fig. 13: Removal rates from ablated squares for PCD.

ZrO₂

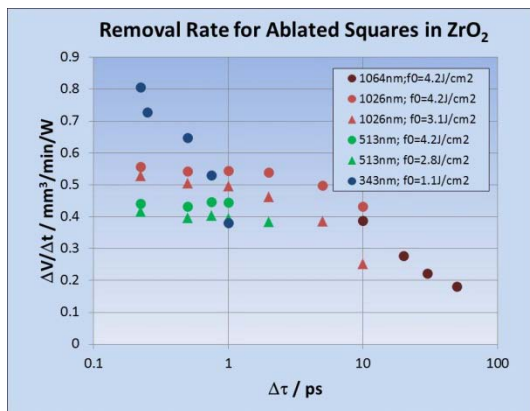


Fig. 14: Removal rates from ablated squares for ZrO₂

Also for ZrO₂ the removal rates were deduced via the ablated squares and compared with former results (see Fig.14). For IR radiation a strong increase of the removal rate between 10 ps and 50 ps was observed, followed from the region between 10 ps and 1 ps with a moderate increase. For shorter pulses no increase is

observed any more. For 513 nm almost no change of the removal rate can be seen between 2 ps and 223 fs. In contrast for UV radiation an about two times higher value of the removal rate at 223 fs compared to 1 ps is obtained, but at a fluence value which is low compared to the other wavelengths.

Glasses

Soda lime glass

Soda lime glass shows a completely different behaviour as illustrated in Fig. 15. Here the removal rate was deduced from the ablated squares. For short pulses the removal rate is small, in the range of 0.1-0.2 mm³/min/W. When the pulse duration is raised, a rapid changeover into a second regime with a much higher removal rate is observed. The pulse duration when this changeover happens depends on the wavelength and the peak fluence. For 513 nm it amounts about 310 fs for $\phi_0 = 4.2 \text{ J/cm}^2$ and 400 fs for $\phi_0 = 5.7 \text{ J/cm}^2$. For 1026 nm the corresponding values are 660 fs for $\phi_0 = 3.1 \text{ J/cm}^2$ and 940 fs for $\phi_0 = 4.2 \text{ J/cm}^2$. After reaching the maximum value the removal rate is continuously dropping down to zero when the pulse duration is further raised. For 1026 nm no ablation was observed for 10 ps and $\phi_0 = 4.2 \text{ J/cm}^2$ or for 5 ps and $\phi_0 = 3.1 \text{ J/cm}^2$. For 513 nm the pulse duration was limited to values equal or lower than 1 ps because the desired pulse energy could not be reached for higher pulse durations. Therefore it was not possible to deduce how the rate drops after reaching its maximum value.

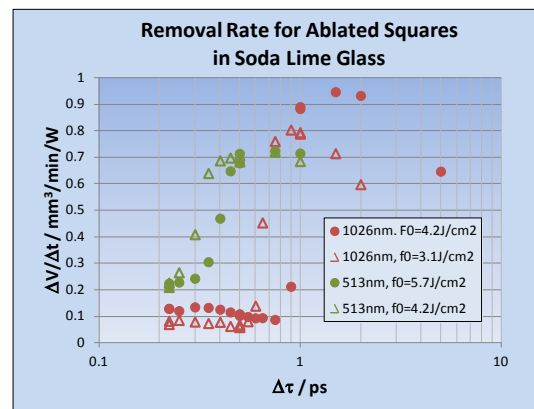


Fig. 15: Removal rates for 1026 nm and 513 nm from ablated squares for soda lime glass.

Fig. 16 shows SEM images for 1026 nm and $\phi_0 = 4.2 \text{ J/cm}^2$ for the shortest pulse durations of 223 fs, at 750 fs where the removal rate begins to increase, at the edge of the increase (900 fs) and at 1 ps shortly before the removal rate reaches its maximum

value. Huge differences of the surface are observed. For 223 fs and 750 fs the bottom surface of the ablated squares is flat with a low roughness, shown on the right side of Fig. 16. For 223 fs the bottom is covered by melted particles with sizes of less than 500 nm and by melting splashes. For 750 fs the number of particles and splashes is much reduced. The overview on the left side shows small cracks in the surface. For 900 fs two different ablation regimes are observed at the same time. In the first one the surface equals the ones from 223 and 750 fs whereas in the second one the surface becomes significantly rougher and also porous. It looks like particles with dimensions higher than 1 μm were somehow “sintered”. This type of surface equals the ones obtained for pulse durations of 1 ps and more where the removal rate is now enlarged by about a factor of 7 compared to the short pulse durations. The cracks in the surface observed for 750 fs and 223 fs may still be present but can’t be detected as easy as before. Similar results have been obtained for the wavelength of 513 nm. Further investigations have to be done to clarify the corresponding ablation processes.

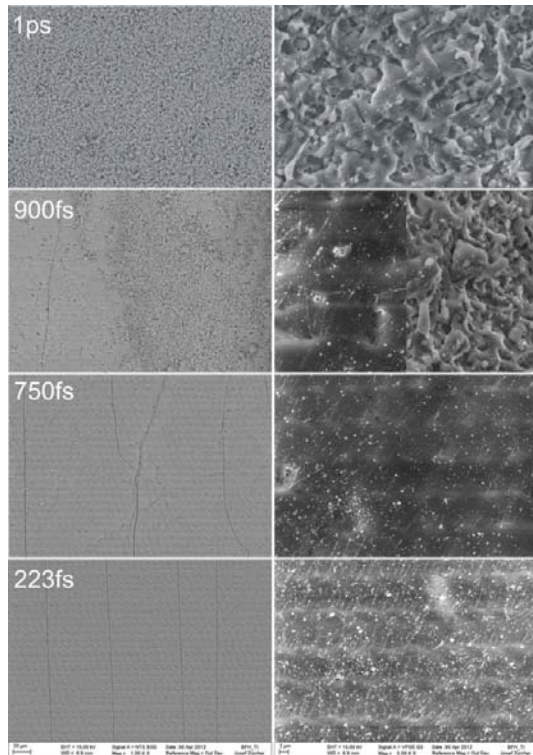


Fig. 16: SEM images of surfaces obtained at the wavelength 1026 nm for soda lime glass with a peak fluence of 4.2 J/cm². Left: overview; Right: Detail

Fused Silica

Squares were machined into 1 mm thick fused silica for different pulse durations and wavelengths. The removal rates were in the range between 0.25 and 0.45 mm³/min/W and did not show such a high dependence on the pulse duration compared to soda lime glass as shown in Fig. 17. Again it was not possible to get pulses longer than 1 ps with the desired fluence for 513 nm. At the wavelength of 1026 nm and for 2 ps and $\phi_0 = 3.1 \text{ J/cm}^2$ or 5 ps and $\phi_0 = 4.2 \text{ J/cm}^2$ the location of the ablation process moved from the front side to the back side due to self-focusing.

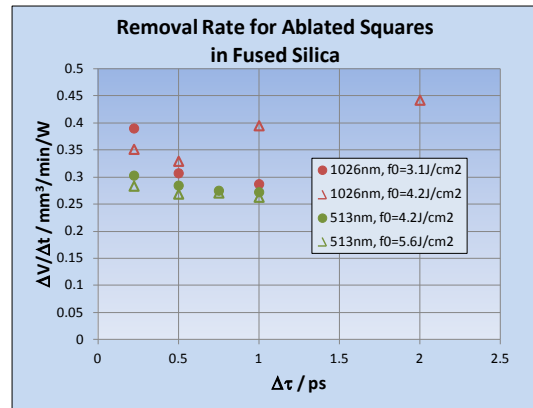


Fig. 17: Removal rates for 1026 nm and 513 nm from ablated squares for fused silica.

The surface of the ablated squares for 513 nm and $\phi_0 = 4.2 \text{ J/cm}^2$ are shown in Fig. 18. For all pulse durations the surface looks porous and again like particles were sintered. In contrast to soda lime glass the particle dimension here is only a few 100 nm. Similar surfaces were also observed for 1026 nm.

Conclusion

For several materials the influence of the pulse duration onto the removal rate was investigated. All materials, except the glasses, show a tendency to higher removal rates when the pulse duration is decreased from 10 ps to 500 fs or 223 fs. For the investigated metals copper and steel as well as for silicon and germanium, this growth can be explained by an increased energy penetration depth for shorter pulses. Compared to 1 μm radiation the increase of the removal rate may be less pronounced as observed for copper, ZrO₂ and PCD. For copper the surface quality is almost independent from the pulse duration whereas for steel better surface qualities are observed for fs pulses. For semiconductors a strong crater and cone formation was observed. For silicon the appearance of the cones can be influenced by the pulse duration, but its dimensions are affected from the applied fluence

and not from the pulse duration. For ZrO_2 and PCD no dependence of the surface quality on the pulse duration was observed, also for pulse durations from 10 ps to 50 ps.

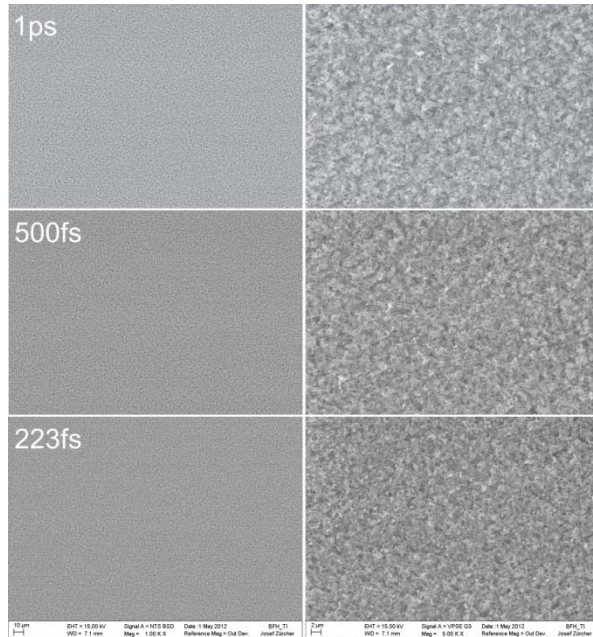


Fig. 18: SEM images of surfaces obtained at the wavelength 513 nm for fused silica with a peak fluence of $4.2 J/cm^2$. Left: overview; Right: Detail

A very interesting dependence on the pulse duration was observed for soda lime glass. Here the removal rate shows a sharp edge to significantly higher values when the pulse duration is raised from fs towards ps. The two regimes separated by this sharp edge shows completely different surface qualities. For short pulses going with low removal rates the surface is flat and covered by malted particles and splashes. For higher pulse durations, going high removal rates, the surface looks like to be covered by sintered particles. This is also the type of surface quality which was observed for fused silica and all investigated pulse durations.

For all materials a pulse duration around 1 ps seems to be a good choice concerning the removal rate and the surface quality.

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