Research Article

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Residual heat during laser ablation of metals with bursts of ultra-short pulses

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Abstract: The usage of pulse bursts allows increasing the throughput, which still represents a key factor for machining with ultra-short pulsed lasers. The influence of the number of pulses within a burst on the specific removal rate is investigated for copper and stainless steel. Furthermore, calorimetric measurements were performed to estimate the residual energy coefficient as well as the absorptance of machined surfaces for copper to explain the reduced specific removal rate for a 2-pulse burst and the similar or even higher rate for a 3-pulse burst compared to single pulse ablation. Based on the measurements, a description of the process using single pulses and pulse bursts with up to three pulses is presented.

Keywords: laser ablation; pulse bursts; residual heat; specific removal rate; ultra-short pulses.

1 Introduction

1.1 Bursts in the context of laser material processing

Ultra-short pulsed (USP) laser systems are used if high quality and precision are demanded. Even if the systems have reached average powers in the 100-W regime [1] for industrial systems and already the kilowatt regime [2–5] for scientific systems, the demanded throughput for industrial applications still represents one of the key factors. Only for some special applications like cutting of carbon fiber reinforced plastics [6] can such high pulse energies offered by today's sources be used for processing. For treating metals, only moderate peak fluences are needed to work at the optimum point, where the ablation process is most efficient with respect to energy conversion and losses [7, 8]. Applying too high pulse energies leads to poor quality [9] and an inefficient process. Furthermore, the achievable scan speed of the mostly used galvanometer scanners limits the scale-up process, in which the average power is simultaneously increased with the repetition rate to preserve the optimum pulse energy and the scan speed to preserve the overlapping of two consecutive pulses, which principally allows to maintain the best quality [10–12]. Even if high scan speeds are reached in the laboratory [13], another limitation is given by the capability of the acousto-optic modulators to switch single pulses at high repetition rates and high pulse energies due to the damage threshold of the used crystals. This leads to pre- and post-pulses and therefore larger ablation spots on the sample surface. The state-of-the-art USP lasers offer the possibility to use so-called pulse bursts. The USP lasers are often built in master-oscillator-power-amplifier (MOPA) arrangement, where the seed oscillator with its frequency f_S, is followed by a pulse picker in order to reduce the internal frequency of the laser source to the desired output frequency f_L needed for processing. This pulse train is then amplified in the following amplifier stages. The pulse picker is capable of letting pass not only single pulses but also a sequence of N pulses, called pulse bursts, which are then amplified. The time spacing between two bursts is given by $\Delta t_L = 1/f_L$, while the temporal spacing between two individual pulses of the bursts is equal to $\Delta t_B = 1/f_S$. It is also possible to generate a burst in which each pulse has the same pulse energy. In addition, the temporal spacing, i.e. the time distance between two subsequent pulses in the burst, may be varied in multiples of the spacing of the pulses of the seed oscillator. In laboratory environment, pulse bursts can be generated using beam splitting and delay lines. With this technique, the time spacing between the pulses in the burst can be varied in smaller time steps than with the industrial laser sources. Pulse bursts are used in different applications,

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i.e. machining of glass [14], welding of glass [15], writing of gratings or waveguides [16], cutting of glass with different technologies [17, 18], or machining of metals [19, 20]. Using pulse bursts allows the possibility to use higher average powers in metal processing while still working at the optimum point.

1.2 Laser ablation of metals with ultra-short pulse bursts

It has been shown that the ablated volume per time and used average power, i.e. specific removal rate using ultra-short pulses in a single pulse mode and a Gaussian beam, can be calculated [7, 8]. This function shows a maximum specific removal rate for an optimum peak fluence. Both values depend on the treated material. The existence of this maximum specific removal rate has been proved many times and is the state of the art for micromachining, especially for engraving and texturing with USP lasers.

Around 2010, the first results of experiments using pulse bursts were reported [21], showing higher removal rates for different materials using pulse bursts compared to single pulses. Later, it was shown that this effect can be explained by the fact that the peak fluence of an individual pulse in the burst is closer to the optimum peak fluence compared to the fluence used in the single pulse experiments [22]. Furthermore, it was shown that for copper, silver, and gold, a 2-pulse burst leads to a drop of the specific removal rate >50% for a pulse duration of 10 ps and a temporal separation of 12 ns [23]. Similar observations were reported by other groups for copper [24], nickel [25], silver [26], and also stainless steel [27, 28] for a smaller temporal spacing of the pulses within the burst. Nevertheless, in Ref. [29], a 2-pulse burst with a total energy of 22 µJ showed a higher efficiency than twice a single pulse with a pulse energy of 11 µJ on steel for a pulse duration of 12 ps and a high temporal separation of the pulses of >100 ns. If the pulses are separated only 20 ns, the 2-pulse burst shows a lower efficiency than twice a single pulse. In contrast, for a lower pulse energy of 4 µJ, the influence of the time distance of the pulses is reduced and the 2-pulse burst no longer shows a higher efficiency. For a 3-pulse burst, the specific removal rate for copper, silver, and gold shows a maximum value that is similar or even a bit higher compared to the one observed with single pulses. In Ref. [20], it was shown that the temporal separation of the pulses in the burst, as well as the energy distribution in the burst, can have an influence. Furthermore, the spot size can also influence the specific removal rate [30]. The reported influence of the spot size on the threshold fluence [31] supports the finding of an influence of the spot size on the ablation process.

2 Volumetric investigations

First of all, the influence of pulse bursts on the specific removal rate was investigated. The specific removal rate was determined for the ablation with multiple burst pulses and multiple layers, which is the standard process for industrial applications. For the experiments, a 10 ps laser Fuego (Lumentum, Zürich, Switzerland, wavelength 1064 nm) was used. The laser beam was guided with some folding mirrors to the beam expander in front of the galvanometer scanner (Intelli$SCAN_{se}$14, Scanlab, Puchheim, Germany), which was used in a synchronized mode [10, 32]. The laser beam was focused with an $f = 160$ mm f-theta objective, resulting in a spot radius w_0 of 15.5 µm and a beam quality $M^2 < 1.36$, both measured with a slit scanning beam profiler (Thorlabs, Dachau, Germany). All individual pulses within a burst had the same energy, which was confirmed by using an internal photodiode of the laser system. The temporal spacing between the individual pulses within the burst amounted to 12 ns. All experiments were performed with an output repetition rate f_L of 200 kHz.

With this setup, square-shaped cavities were machined by ablating multiple layers using a spatial overlap of 75% in both lateral directions with respect to the focal diameter. The number of machined layers N_L was adapted to maintain an equal number of pulses per area for the different bursts. Afterwards, the depth of the machined squares z was measured using a white light interferometric microscope (smartWLI from GBS, Ilmenau, Germany). The specific removal rate could then be determined according to Ref. [22] by

$$\Delta V / (\Delta t \cdot P_{av}) = \Delta V / E_p = \frac{z \cdot p_x \cdot p_y}{N_L} \cdot \frac{f_L}{P_{av}} = \frac{z \cdot p_x \cdot p_y}{N_L} \cdot \frac{1}{E_p}, \quad (1)$$

where p_x is the pitch in x-direction (distance between two consecutive pulses) and p_y is the pitch in y-direction (distance between two lines). f_L denotes the used output frequency and P_{av} the used average power of the burst. E_p denotes the total energy of all pulses within the burst.

In the following, the specific removal rate is plotted as a function of the peak fluence of an individual pulse of the burst $\phi_{0,S}$. The total fluence of the burst is given by the sum of the fluences of the individual pulses [20]; hence,

$$\phi_0 = \sum_{i=1}^{N} \phi_{0,S} = \frac{2 \cdot P_{av}}{f_L \cdot \pi \cdot w_0^2}. \quad (2)$$

N denotes the number of pulses in the burst and ranges from 1 to 3 in this study. The specific removal rate $\Delta V/(\Delta t \cdot P_{av})$ can be used as a measure of the energy efficiency of the ablation process. Comparing single pulses with pulse bursts should be done by considering the specific removal rate as a function of the peak fluence of the individual pulses. Even if the removal rate $\Delta V/\Delta t$ is increased when using pulse bursts at a fixed repetition rate due to the higher used average power (cf. Figure 1, left), the ablated volume per time and average power $\Delta V/(\Delta t \cdot P_{av})$ can be smaller when pulse bursts are applied on stainless steel (cf. Figure 1, right). From the industrial (or economic) point of view, in general, it is better to use single pulses for ablation at a doubled repetition rate (also resulting in doubled scan speed) than using burst pulses. Only if the scale-up process is limited due to the repetition rate of the laser or the scan speed, the use of pulse bursts can help to use the full average power given by the laser system. Still, it should be noted that for steel at 1064-nm wavelength, the specific removal rates are in the same order of magnitude for single pulses and bursts with multiple pulses.

In contrast, for copper, a large decrease of the specific removal rate for a 2-pulse burst is observed, while a 3-pulse burst leads to a higher specific removal rate than single pulses (cf. Figure 2, right). The ablated volume per time is significantly decreased (cf. Figure 2, left) when using a 2-pulse burst compared to a single pulse ablation at the same repetition rate. This tendency was also reported for silver and gold [23] and aluminum [33]. To gain more insight into the energy transfer during processing, calorimetric measurements have been performed. They are introduced in the following section.

3 Calorimetric investigations of copper

The used setup consisted of a thermally insulated copper substrate on which samples contacted with heat-conducting

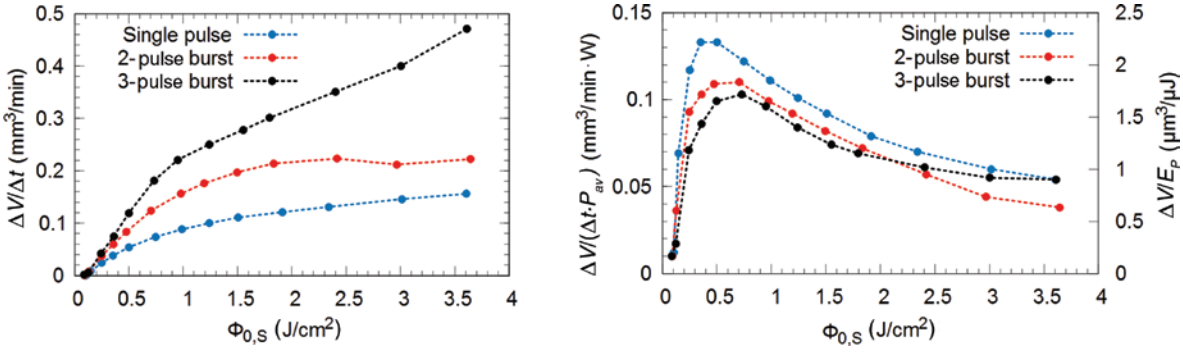

Figure 1: Removal rate of stainless-steel 1.4301 (in US: AISI 304) as a function of the peak fluence and the number of applied burst pulses at 200 kHz (left). Specific removal rate of stainless-steel 1.4301 (in US: AISI 304) as a function of the peak fluence and the number of applied burst pulses at 200 kHz (right).

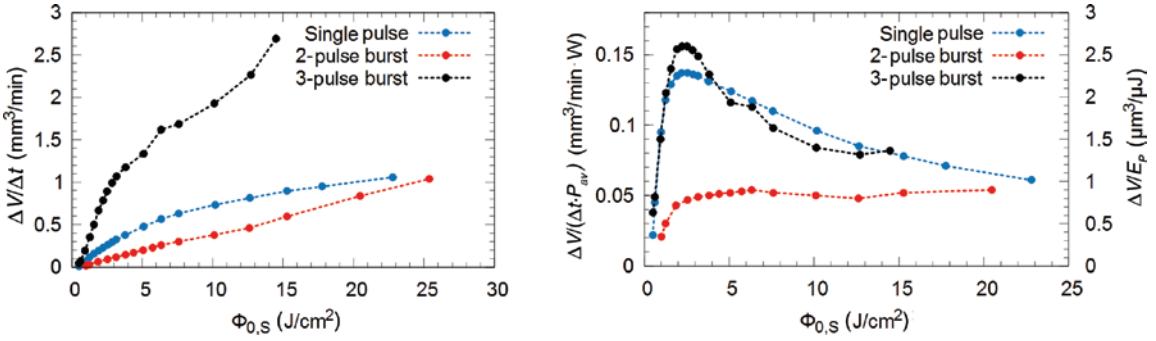

Figure 2: Removal rate of pure copper Cu-DHP (in US: Cu12-200) as a function of the peak fluence and the number of applied burst pulses at 200 kHz (left). Specific removal rate of copper Cu-DHP (in US: Cu12-200) as a function of the peak fluence and the number of applied burst pulses at 200 kHz (right).

paste can be applied for calorimetric investigations [34]. A Pt100 thermocouple was attached to the substrate on the opposite side of the sample to be examined. The voltage signal from the Pt100 thermocouple was evaluated by using a data module (USB-2401, Adlink Technology, Mannheim, Germany) and a computer. The result is a time-resolved temperature signal with a temporal resolution of 300 ms. In order to measure the residual energy that is deposited into the workpiece during an ablation process, the workpiece was processed for several 10 s periods with a focused laser beam, which was moved by a galvanometer scanner. The measured temperature increase ΔT of the sample can be used to calculate the residual energy ΔE [34, 35]. As the totally incident energy E_{in} is known from the process parameters laser power P_{av} and the total time the laser was turned on t_{laser}, the so-called residual energy coefficient η_{res} can be calculated by

$$\eta_{res} = \frac{\Delta E}{E_{in}}. \tag{3}$$

It represents the relative amount of energy that is deposited in the workpiece during the ablation process, and therefore lies between 0% and 100%. If its value is close to zero, the ablation process is very efficient with respect to energy as most of the incident energy is used for ablation. When it is close to 100%, most of the energy is used to heat the material instead of removing it.

To examine the residual energy coefficient properly, each scanning process when using pulse bursts was tuned in a way that the measured temperature increase amounted to a minimum of 4°C. This resulted in a maximum measurement error for η_{res} of ±1.6%. Taking into account the deviations between several measurements under the same experimental conditions, the error for the residual energy coefficient during processing was estimated to be ±2%. In order to be able to realize the mentioned minimum temperature increase, the number of passes across the sample, i.e. number of machined layers, was adjusted and varied from 12 to 48 passes.

To investigate the influence of the number of pulses within a burst on the residual energy, single pulses as well as 2-pulse and 3-pulse bursts were applied during the scanning ablation process. The used fluence of each pulse amounted to 2.66 ± 0.05 J/cm², which corresponds to the optimum fluence for a single pulse ablation process. Hence, the total fluence of a 2-pulse burst and a 3-pulse burst was approximately 5.3 and 8.0 J/cm², respectively. The spatial overlap of consequent pulses amounted to 75% with respect to the focal diameter.

The results of this study are given in Figure 3. It can be seen that there is no significant difference in the measured residual energy coefficient when applying different burst configurations. Instead, all measured values lie between 17.0% and 18.6%. This very surprising fact will be discussed in more detail in the next section.

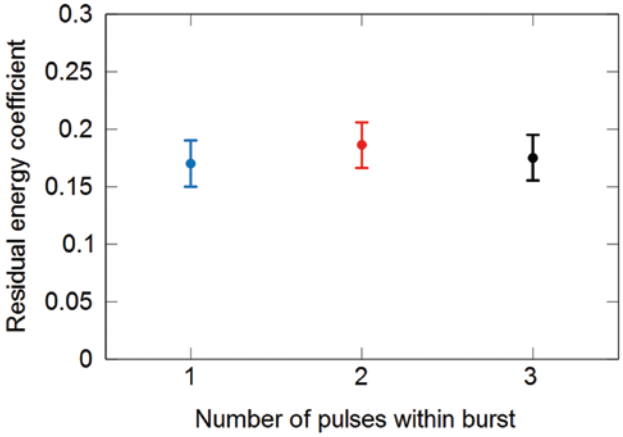

Figure 3: Residual energy coefficient η_{res}, i.e. the ratio of residual energy and incident energy, as a function of the number of pulses within a burst for a fluence of 2.66 ± 0.05 J/cm² and a spatial overlap of pulses of 75%.

4 Discussion

In the previous sections, it was shown for copper that, on the one hand, the ablated volume per pulse decreases significantly when using a 2-pulse burst compared to a single pulse, while for a 3-pulse burst the ablated volume per pulse even surpasses the value of a single pulse. In the literature, the behavior of a 2-pulse burst has been investigated experimentally and numerically. The first report on the decrease of volume when applying double pulses was given by Semerok and Dutouquet for time delays between 1 and 100 ps [36]. As, additionally, the plasma emission of the process was measured and showed an increase for longer time delays, it was concluded by the authors that the material ablated by the first pulse shielded the second pulse [36]. This hypothesis was supported by the numerical investigations of Povarnitsyn et al. for copper [24, 37]. Furthermore, these authors explicitly showed for aluminum that not only a shielding of radiation takes place but also a re-deposition of material that already left the surface after the first pulse [38]. Recent experimental findings using shadow-graphic imaging showed that a suppression of ablation indeed takes place for copper when using a 2-pulse burst with a temporal delay of 12 ns [39]. The authors also found that the ablation cloud generated after applying a third pulse 24 ns after the first pulse contains more mass compared to the cloud visible after the first pulse [39]. This coincides well

with the findings in Section 2 and a significant increase of ablated volume when using a 3-pulse burst.

To gain more insight and find reasons for the measured constant residual energy, which is independent of the applied number of burst pulses, the absorptance was investigated. It was measured as laser-irradiated surfaces in general show an increased absorption due to surface structuring in the scale of microns and nanometers [40–42]. The absorptance is measured by using the calorimetric setup in Section 3 with an enlarged beam diameter of 2 mm, which is realized by using a defocused and motionless beam (=no scanner active). When the laser beam is highly defocused, the measured residual energy coefficient given by Eq. (3) is equal to the absorptance, as the fluence is below the ablation threshold. The absorptance was measured after the ablation process, when the sample again has reached room temperature. The overview of these measurements is given in Figure 4. The measured value for the untreated surface, which represents the absorptivity of the material, is in good agreement with the value given in Ref. [43] and amounts to 1.2 ± 0.6%. For a single pulse, the measured value of 13.2 ± 2.8% is in good agreement with the values presented in Ref. [44]. For a 2-pulse burst and also a 3-pulse burst, the absorptance of the surface after the machining is significantly increased to 17.9 ± 2.2% and 26.8 ± 2.3%, respectively. The absorptance is independent of the number of machined layers and increases with the number of pulses in the burst. The given values are calculated by taking into account the values given in Figure 4. They are averaged and their standard deviation combined with the measurement error amount to the stated errors.

For a fair comparison, the absorptance immediately before a subsequent burst pulse irradiates the surface has to be known. Unfortunately, this value cannot be measured by using this setup. Instead, a measurement system

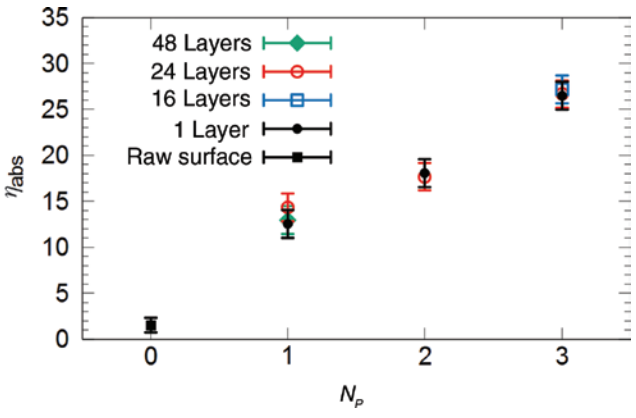

Figure 4: Absorptance of laser-machined areas as a function of the number of pulses within a burst for copper.

with a minimum temporal accuracy of nanoseconds or shorter is needed. Indeed, it has been reported before by using pump-probe microscopy or pump-probe ellipsometry that the absorptivity for time delays of up to several 100 ps can increase dramatically for metals [45–47] and especially copper [48]. Typically, the increase of absorptivity for fluences that are multiples of the ablation threshold ranges between approximately 20% (molybdenum, rise from ≈41% to ≈59% [45]) to 75% (aluminum, rise from ≈15% to ≈90% [46, 47]) for ultra-short pulses. For copper, Winter et al. [48] found for a wavelength of 528 nm a rise of 8% from the cold material (39.0%) to the excited material (46.7%) after 20 ps and for a fluence of 2.5 J/cm². Measurements in the infrared conducted by the authors of this paper in steady state show a rise from 3.4% absorptivity of an untreated solid surface to 6.2% absorptivity of a liquid surface. The experiments have been performed in vacuum and were described previously in Ref. [49]. In conclusion, the change of absorptance during the ablation process may rise from just a few percent to up to a factor of 2 for copper. Even a further increase seems possible, as the fluences used for investigations by other authors do not reach 10 times the ablation threshold.

Based on these findings and the assumption that the surface is cold before the next burst or single pulse impinges on the surface, the ablation process for a single pulse, a 2-pulse, and a 3-pulse burst can be described as follows:

1. A single pulse ablation process of copper results in a cloud expanding from the surface, which contains a certain amount of mass. The ablated volume per time approximately equals to 0.26 mm³/min when the fluence is approximately 10 times the ablation threshold (≈2.66 ± 0.05 J/cm²; cf. Figure 2, left).
2. The first pulse of a 2-pulse burst irradiates a surface showing an increased absorptance of about 5% compared to single pulses. After processing the first layer, the absorptance is increased to this value and stays constant for follow-up ablation processes (cf. Figure 4). In the end, a higher fluence contributes to the ablation process compared to a single pulse and more material is ablated. Taking into account Figure 2 (left), the first pulse within a 2-pulse burst ablates 32% more material compared to a single pulse (0.35 mm³/min compared to 0.26 mm³/min). The second pulse of the 2-pulse burst, which is applied 12 ns after the first pulse, is shielded by the ablation cloud that was induced by the first pulse. This results in a re-deposition of material and hence leads to less material removal of a 2-pulse burst compared to single pulse ablation. The ablated volume per time therefore is significantly decreased and amounts to

approximately 0.10 mm³/min. Due to the much smaller ablated volume per pulse for single pulse processing in the case of stainless steel, which leads to a less dense particle plume, the shielding of the second pulse in the burst is less pronounced. Also, the second pulse in the burst ablated material, as shown in Figure 1.

3. For a 3-pulse burst, the first pulse irradiates a surface with an absorptance that is approximately doubled compared to single pulses. The fluence contributing to the ablation is therefore also doubled, leading to an 86% higher removal rate of the first pulse of the burst compared to a single pulse (0.49 mm³/min compared to 0.26 mm³/min). The second pulse of the burst pushes back a certain amount of the ablated material. When a third pulse with the same fluence is applied 12 ns after the second pulse, it hits a surface that is still hot and therefore significantly more material is ablated compared to a single pulse due to the higher absorptance of the hot surface.

Depending on the amount of material that was pushed back by the second pulse, the possible range of the absorptance of the hot material of point 3 can be estimated in the following way. As the dynamics of the second pulse for a 2-pulse and a 3-pulse event can be different, there are two extremes, (a) and (b), and a highly probable case, (c):

(a) All material that was ablated by the first pulse is pushed back to the surface. In this case, the third pulse needs to ablate a volume per time of approximately 1 mm³/min. If this takes place, the used fluence to ablate this amount equals to approximately 17.8 J/cm². The consequence would be that the third pulse has to be fully absorbed, and hence $\eta_{abs} \approx 100\%$. This means the cold absorptance is increased by a factor of 7.

(b) The second pulse is only shielded but does not push material back to the surface. In this case, the ablated volume per time after a 2-pulse burst amounts to 0.26 mm³/min. Hence, the third pulse only would ablate a value of 0.74 mm³/min of volume per time, which means that a fluence of approximately 10 J/cm² needs to be applied. This results in an absorptance of $\eta_{abs} \approx 52.7\%$ or an increase by a factor of 4, respectively.

(c) There is no change in dynamics between the second pulse of a 2-pulse burst and a 3-pulse burst. Then, the ablated volume per time after the second pulse of a 3-pulse burst equals to 0.1 mm³/min. The third pulse then needs to ablate a volume per time of 0.9 mm³/min, which is equal to an applied fluence of 15 J/cm². This results in an absorptance of $\eta_{abs} \approx 77\%$ or an increase by a factor of 5.8, respectively.

It is assumed that each ablation event (single pulse, 2-pulse, or 3-pulse burst) is independent of the one that happened before. In particular, heat accumulation effects can be neglected for the used scanning parameters ($f_L = 200$ kHz, overlap 75%) [44]. In general, possibilities (a–c) allow explaining the findings of the volumetric measurements (cf. Figure 2). However, the exact values of the absorptance of the first pulse during irradiation, the absorptance of the second pulse, as well as the amount that is pushed back by it and the absorptance of the third pulse remain unclear. The used setups are not capable of identifying these values due to a lack of temporal and spatial resolution. Instead, the authors of this paper can only give possible ranges for some of these values as described before.

The results of the calorimetric measurements with respect to the residual energy coefficient even give rise to more questions. A constant η_{res} cannot be attributed to the amount of ablated and re-deposited mass. Instead, secondary effects not directly connected to the ablated volumes seem to dominate the energy transfer to the material. A highly probable explanation is that either thermal radiation or radiation emitted by a plasma (or a mixture of both) dominates the residual energy that remains within the material. The effect of energy transfer by plasma heating, which increases the residual energy coefficient, has been investigated for a single pulse and platinum by Vorobyev and Gou [50]. It was shown that with a decrease of the pressure of the ambient atmosphere, the residual energy coefficient also decreases significantly. As photographs of the occurring plasmas were additionally taken, the occurrence of the weakly pronounced plasma in vacuum and a low residual energy coefficient could be directly correlated for platinum [50]. Another explanation could be given if the exact values of the absorptance of the first pulse during irradiation, the absorptance of the second pulse, as well as the amount that is pushed back by it and the absorptance of the third pulse were known. There is a chance that other effects balance out the residual energy coefficients. As this is subject to speculation, a proper description of the ablation dynamics by both numerical and experimental studies with much better temporal and spatial resolution is needed.

5 Conclusions and outlook

In order to gain deeper insight into the physical processes during laser ablation with single pulse, 2-pulse,

and 3-pulse bursts, volumetric and calorimetric investigations have been performed. In contrast to stainless steel, copper shows a significant decrease of the ablated volume and hence the specific removal rate when using a 2-pulse burst compared to a single pulse ablation process. The reason for this behavior is the shielding of the incident second pulse, leading to a re-deposition of material. When applying a 3-pulse burst, an increased absorptance of the surface leads to a removal rate that is higher than if three single pulses would have caused ablation.

This study revealed that the residual energy coefficient is practically constant and independent of the number of applied pulses in the burst. A $17.0 \pm 2.0\%$ to $18.6 \pm 2.0\%$ proportion of the applied energy is deposited within the material after processing. This surprising fact is attributed to side effects such as thermal radiation or radiation emitted by a plasma, which dominate the energy transfer processes.

Furthermore, the absorptance of the blank and the machined surfaces was determined. For a raw surface, it amounts to $1.2 \pm 0.6\%$; for single pulses, it amounts to $13.2 \pm 2.8\%$; for 2-pulse bursts, it amounts to $17.9 \pm 2.2\%$; and for 3-pulse bursts, it amounts to $26.8 \pm 2.3\%$. As these values partially are smaller than the residual energy coefficient, the authors discussed possible explanations with respect to the real values of absorptance on the ultra-short time scale. A possible range of the increased absorptance during processing finally was given, which can fully explain the behavior of the specific removal rate. Unfortunately, the used setup had a too small sensitivity and resolution, which did not allow measuring a difference between the different pulse bursts.

For a complete understanding of the ablation process using pulse bursts, additional experiments with higher temporal and spatial resolution should be performed to clearly distinguish the influence of each pulse within the burst. Other materials like metals and semiconductors should be investigated as well in order to allow a detailed explanation of the processes involved during multi-pulse ablation.

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