In-Situ Analysis of Ultrashort Pulsed Laser Ablation with Pulse Bursts

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Laser ablation using ultrashort pulsed (USP) laser sources enables contact free materials processing with very high precision at negligible thermal influence for the processed workpiece. However, the achieved productivity is still too low for industrial applications in many cases. To increase the throughput of USP laser processes, three approaches are followed: high repetition rates up to several MHz using fast beam deflection techniques, pulse bursts or parallel processing with multiple laser foci. In case of high repetition rates and pulse bursts the temporal and spatial distance of consecutive pulses becomes so small that heat accumulation and shielding effects of plasma and particles affect the processing quality and achieved efficiency significantly. In order to gain a better insight of these still barely investigated effects we apply in this work in situ imaging with an ICCD camera during ablation processes of copper and stainless steel with pulse bursts to evaluate the excitation of process luminescence as well as its relation to ablation efficiency and shielding effects.

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1. Introduction

Ultrashort pulse (USP) laser processing with pulse durations in the range of picoseconds to femtoseconds enables a better surface quality, higher precision and less thermal impact compared to materials processing using longer pulsed laser radiation [1]. However, despite the high quality, USP processes are still not established for broad industrial application due to the lack of achievable productivity [2-7]. In order to improve the processing speed and throughput, the utilization of high power USP laser systems in the range of hundreds of Watt average power is one major research topic in the field of USP processing. Since the well-known and often reported optimum fluence of a few J/cm² [6, 8, 9] for metal processing is limiting the usable pulse energy, the high average power must be put into effect by other approaches. The most promising techniques for this purpose are the usage of high repetition rates [2] up to several MHz using fast beam deflection techniques, pulse bursts or parallel processing with multiple laser foci [6, 10]. In this context, pulse bursts are trains of laser pulses with several rapidly following laser pulses, which are usually generated with the oscillator frequency of the laser source, which is typically in the range between 40 and 80 MHz. In the case of applying high repetition rates and pulse bursts the temporal and spatial distance of consecutive pulses becomes so small that the heat accumulation and shielding effects of plasma and particles affect the processing quality and achieved efficiency significantly [7]. The effect of heat accumulation refers to the rise of the cumulated residual thermal load of the individual pulses, which is not contributing to the ablation process and remains in the material. For high repetition rates, this residual heat is relevant for the processing, because of the insufficient time for the heat to spread into the workpiece. Heat accumulation can lead to a better surface finish by a small molten surface layer [10], but can also nullify the advantages

of the highly precise USP laser processing. Shielding effects on the other hand prevent the laser pulses from reaching the surface with their full pulse energy and therefore reduce the achieved ablation rate. This effects are attributed to the interaction of the incident laser beam with the ablation plumes of previous pluses in which the radiation is reflected, scattered or absorbed by particles, vapor or plasma originating from the ablated matter.

In order to gain a better insight into these still barely investigated shielding effects we present in-situ analysis during ablation using pulse bursts with varied number of pulses at high average laser powers of up to 200 Watts for different materials. Therefore, the intensity of the process radiation is compared to the observed ablation efficiency of the associated process parameters. Furthermore, the propagation speed of the luminescent ablation plume front is evaluated and the half-life period of the luminous matter is calculated. In addition, two spatial areas of luminous matter accumulations are identified. A surface-near, fast propagating accumulation of matter and a long-lasting accumulation which aggregate at a certain distance from the treated surface.

2. Experimental setup and procedure

For this study, the laser radiation of an Amphos 400 high power USP laser was focused with an f-theta objective with a focal length of 160 mm achieving a 50 μ m spot (1/e² of the intensity) on the samples to be investigated. The used USP laser system is capable of emitting laser radiation at a central wavelength of $\lambda = 1030$ nm and variable pulse durations between $\tau = 2$ and 20 ps. The repetition rate is adjustable between f_{rep} = 0.4 to 40 MHz at a constant maximum average power of P = 400 W. Furthermore, the Amphos 400 can be used in a burst mode which makes it possible to emit pulse bursts with 1 to 10 pulses each instead of single pulses. The time separation between the single pulses within one burst is - corresponding to the oscillator frequency of 40 MHz – 25 ns and the averaged energy distribution of the pulses in the bursts (measured with Thorlabs DET10A/M, a fast, amplified photo diode, Fig. 1) is shown in Fig. 2.

With this setup, parameter studies were carried out on copper (CW024A) and stainless steel (1.4301) samples with varied number of pulses per burst and fluences to find suitable process parameters that enable good surface quality within surface ablation processes. Therefore, rectangular cavities were fabricated and the ablation efficiency was calculated based on the obtained ablation depth. Optimum parameters that give a good compromise between ablation efficiency and surface quality for all applied number of bursts were used for the in-situ experiments (see Table 1).

For the in-situ experiments a shadowgraphy setup (Fig. 3) with a 4 Picos ICCD camera from Stanford Computer Optics was used and synchronized to the lasers system with a Quantum Composers 9530 pulse generator. A high power LED (2.1 W) at 450 nm was used as an optional backlight illumination used to align the samples in front of the objective lens. The ICCD sensor with an intern upstream multichannel plate is capable of exposure times down to 200 ps which was set to 1 ns in the presented experimental investigations. With this setup images of the process luminescence were taken in which the background illumination was only used to align the sample and is outshined by the process luminescence due to its high intensity but due to the shadowgraphical setup a bandpass filter is integrated in the objective lens so only 450 ± 8 nm of the process luminescence spectrum reached the ICCD detector.



Fig. 1 Example of the energy distribution measurement of the bursts with a fast, amplified photo diode for a burst with four pulses.



Fig. 2 Averaged energy distribution of the pulses in the bursts of the used Amphos 400.



Fig. 3 Setup for in-situ investigation of the ablation process with optional background illumination for shadowgraphical images.

Table 1 Summary of processing parameters.

laser parameter	
wavelength λ	1030 nm
focus diameter 2w ₀	50 µm
pulse duration τ	2 ps
pulses per burst PpB	1 - 8
repetition rate f _{rep} (bursts)	400 kHz
seed frequency f_{seed} (burst pulses)	40 MHz
pulse overlap (parameter study)	80 %
single pulse fluence F ₀ (parameter study)	0.1 - 15 J/cm ²
single pulse fluence F ₀ (in situ examinations for copper)	5 J/cm ²
single pulse fluence F ₀ (in situ examinations for steel)	1.5 J/cm ²
camera parameter	
exposure time	1 ns
observed wavelength	$450\pm8\ nm$
observed period of time	0-500 ns*
	*after the first pulse

3. Experimental results

For the examinations in this work the used fluences for copper and steel were determined by previous studies in which the number of pulses per burst and the single pulse fluence was varied while fabricating 3 mm x 3 mm cavities into with a multi-pass process (parameters in Table 1). The number of layers has been set so that round about 100 µm depth of the cavities were reached. The real depth was subsequently measured by laser scanning microscopy and used to calculate the ablation efficiency η with $\eta = \frac{d_{ablation} \cdot v_{scan} \cdot \delta y}{n_{layers} \cdot \overline{P}}$ where $d_{ablation}$ is the ablation depth, v_{scan} is the scan speed, δy is the line pitch, n_{layers} is the number of the passes and \overline{P} is the average output power. The determined optimum single pulse fluences are 5 J/cm² for copper and 1.5 J/cm² for stainless steel and were selected accordingly to

optimize the optical surface quality, average surface roughness S_a and the ablation efficiency for all number of pulses per burst investigated (see Fig. 4 and Fig. 5). Further, it should be mentioned that the term fluence in this work refers always to the irradiated single pulse peak fluence $F_0 = \frac{2 \cdot E_P}{\pi \cdot w_0^2}$, which was determined based the average power measured

by a thermal power head. For copper the used fluence (5.0 J/cm²) is approximately

15 times and for steel (1.5 J/cm²) 21 times the by Neuenschwander et al. [4] reported single pulse threshold fluence of the respective material for a pulse duration of 2 ps. The referred values of Neuenschwander et al. [4] are in the range of $F_{th} = 0.05 - 0.07 \text{ J/cm}^2$ for steel and $F_{th} = 0.30 - 0.07 \text{ J/cm}^2$ 0.35 J/cm for copper. The used fluence is therefore more than double the reported optimum fluence for single pulse ablation for these materials which would be ~ 7.4 (e²) times the threshold fluence. This increase can probably be attributed to shielding effects that reduce the effective fluence reaching the sample surface, shifting the fluence of optimum ablation efficiency to higher values. Moreover, as already mentioned the processing fluences were not only chosen regarding the efficiency but also for optimization of the surface quality of all burst processes which is why the most efficient fluence for single pulse ablation was not chosen exactly.



$$\begin{split} \lambda &= 1030 \text{ nm} \mid \tau = 2 \text{ ps} \mid F_0 = 1.6 \text{ J/cm}^2 \mid f_{\text{rep}} = 400 \text{ kHz} \mid f_{\text{burst}} = 40 \text{ MHz} \\ 2w_0 &= 50 \text{ } \mu\text{m} \mid \text{v}_{\text{scan}} = 6 \text{ m/s} \mid \text{line pitch} = 15 \text{ } \mu\text{m} \\ 1\text{-8 pulses per burst} (\text{PpB}) \mid \text{material: stainless steel} \end{split}$$

Fig. 4 Surface images, roughness and ablation efficiency of a previous parameter study to determine suitable ablation parameters for stainless steel.



$$\begin{split} \lambda &= 1030 \text{ nm} \mid \tau = 2 \text{ ps} \mid F_0 = 5.0 \text{ J/cm}^2 \mid f_{\text{rep}} = 400 \text{ kHz} \mid f_{\text{burst}} = 40 \text{ MHz} \\ 2w_0 &= 50 \text{ } \mu\text{m} \mid v_{\text{scan}} = 6 \text{ m/s} \mid \text{line pitch} = 15 \text{ } \mu\text{m} \\ 1\text{-8 pulses per burst (PpB)} \mid \text{material: copper} \end{split}$$

Fig. 5 Surface images, roughness and ablation efficiency of a previous parameter study to determine suitable ablation parameters for copper.

In Fig. 6, the ablation efficiency as a function of the number of pulses per burst is shown for the two sample materials stainless steel and copper. For stainless steel the ablation efficiency is decreasing with increasing number of pulses per burst. The decrease can probably be attributed to shielding effects that reduce the effective fluence of the later pulses in a burst train and is consistent with results obtained by Kramer et al. [5] and Neuenschwander et al. [11] and is also consistent with the previously mentioned effect which increases the optimum fluence. However, for copper another behavior of the ablation efficiency can be observed. Here, a low efficiency is measured for even numbers of pulses per burst and a significantly higher efficiency for all investigated odd numbers of pulses is observed a finding that is also reported by Neuenschwander et al. [11] and Jäggi et al. [3]. A possible explanation for this behavior is that the first and all other odd numbered pulses ablate material that shields the even numbered pulses. Therefore, these pulses ablate less material that reduces the shielding effect for the subsequent pulse. As a consequence, the following pulses (odd numbered) reach the surface without being shielded and ablate material efficiently.

While evaluating the ablation efficiency of bursts, it should be noted that the ablation rate (mm³/min) nevertheless increases with the number of pulses per burst as the used average power increases. Therefore, with well-adjusted burst parameters, a higher throughput compared with single pulse processes can be established in many cases of application which is the reason why bursts are used in the first place [10].



Fig. 6 Ablation efficiency for pulse bursts on copper and stainless steel. ($\lambda = 1030 \text{ nm}$, $\tau = 2 \text{ ps}$, $f_{rep} = 400 \text{ kHz}$, $f_{burst} = 40 \text{ MHz}$, $F_{0_copper} = 5 \text{ J/cm}^2$, $F_{0_steel} = 1.5 \text{ J/cm}^2$)

In order to examine the shielding of the single pulses of a burst the ablated volume per pulse is calculated. On the assumption that the n-1 pulses of a burst with n pulses behave like a burst with n-1 pulses the equation (2) can be used for this calculation. Where n is the number of pulses per burst, η_{burst_n} is the ablation efficiency of the burst with n pulses, η_{pulse_i} is the ablation efficiency of the i-th pulse of a burst which is calculated iteratively with (1), $\overline{P}_{\text{burst}_n}$ is the average power applied and f_{rep} is the repetition rate of the bursts. Additionally, it has to be taken into account that the equation (1) is based on an equal energy distribution between the pulses in the burst which is almost the case with the used laser system (see Fig. 2). In Fig. 2 a loss of energy of the last pulse of each burst can be noted. Since the fluence in the experiments is set via the average power, this has an underestimation of the first n-1 pulses and an overestimation of the n-th pulse of an n-burst as consequence. This energy deviation was calculated, compared to the determined fluencedependent efficiency for single pulse ablation of the materials and compensated by correction factors for further calculation.

$$\eta_{pulse_i} = n_i \cdot \eta_{burst_{n_i}} - \sum_{j=1}^{n_i-1} \eta_{pulse_j} \quad (1)$$
$$\Delta V_{pulse_n} = \left(n \cdot \eta_{burst_n} - \sum_{i=1}^{n-1} \eta_{pulse_i}\right) \cdot \frac{\overline{P}_{burst_n}}{f_{rep} \cdot n} \quad (2)$$

The in this way calculated ablation volumes are shown as a function of the pulse number in Fig. 7 for stainless steel and Fig. 8 for copper. Additionally, the integrated intensity of the process luminescence 24 ns after the related pulse is plotted. For this purpose, the backgrounds taken before each image without the laser process were subtracted from the images, the intensity of the so processed images has then been integrated and averaged over the three images taken for each point in time. The intensities of the ablation products as determined this way lie above the intensity of the background illumination and therefore, it is called luminous matter in the following. The 40 MHz seed frequency of the laser with which the pulses are emitted results in a temporal distance of 25 ns between the pulses. Knowing that, the plotted intensity in Fig. 7 and Fig. 8 shows the value 24 ns after the related pulse and also the value 1 ns before the subsequent pulse arrives. Therefore, it can be assumed that, if the luminous matter shields the following pulse by absorption, scattering or reflection, the intensity count should correspond to the extent of shielding for the upcoming pulse. This assumption is based on the hypothesis that regardless of whether the luminous matter consist of plasma, vapor, particles or a mixture of these the intensity should correlate with its density and therefore a high intensity before a pulse should indicate a pronounced shielding effect.



Fig. 7 Ablated volume per pulse for stainless steel compared with the integrated intensity of the process luminescence 24 ns after each pulse. ($\lambda = 1030 \text{ nm}, \tau = 2 \text{ ps}, f_{rep} = 400 \text{ kHz}, f_{burst} = 40 \text{ MHz}, F_{0 \text{ steel}} = 1.5 \text{ J/cm}^2$)

In either case the intensity 24 ns after the pulse is rising steadily without significant conspicuity while the ablated volume per pulse alternates with the number of pulses. The even numbered pulses ablate less than the odd numbered ones. For stainless steel the fluctuation is less pronounced than for copper, so the burst efficiency in Fig. 6 is barley affected for steel. Nevertheless, by evaluating the ablated

volume per pulse, a similar trend for burst ablation as for copper can be identified for which it is already known. This correlates with the observations of Sailer et al. [12] for double pulse ablation of steel. Thus, the unconventional behavior for the ablation of copper for even number of pulses within a pulse train can be identified for the processing of steel as well. However, the effect is less pronounced and is visible only if the ablation efficiency is calculated for the single pulses within a burst of pulses. For copper, on the other hand, this effect is so strong that the calculated ablated volume becomes negative for even numbered pulses and is increasing for the second and third pulse compared with the first pulse. Carefully considered, the negative values may be a hint of redeposition of ablated material as it is already suspected in the technical literature sometimes [6, 13]. This would also reflect the conspicuous behavior of the ablation efficiency for copper in Fig. 6. The increase of the ablated volume per pulse on the other hand, could probably be explained by a higher absorption of a molten layer of copper on the surface [11] which therefore could overcompensate the loss of ablation of the even numbered pulses and be the reason for the highest ablation efficiency measured for five pulses per burst on copper (see Fig. 6). In addition, an increase in the evaluated intensity should be observed if shielding by absorption takes place which is not the case either.



Fig. 8 Ablated volume per pulse for copper compared with the integrated intensity of the process luminescence 24 ns after each pulse. ($\lambda = 1030$ nm, $\tau = 2$ ps, $f_{rep} = 400$ kHz, $f_{burst} = 40$ MHz, $F_{0 \text{ copper}} = 5 \text{ J/cm}^2$)

Another finding that suggests that there is no direct excitation of the luminous matter by the incident laser radiation is shown in Fig. 9 where the recorded images itselves are evaluated. In Fig. 9 three images of a pulse burst with eight pulses for three different times are shown. One nanosecond before the fourth pulse arrives on the surface, the moment of the incoming pulse and one nanosecond after the pulse has arrived. Two spatial areas with different behavior of the ablated matter can be identified:

- 1.) The long-distance luminous matter which comes to a standstill at a height round about $200 400 \,\mu m$ with a high lifetime.
- 2.) The surface-near luminous matter which expanse and propagates fast from the surface.

It becomes evident that no change in the intensity of the long-distance luminous matter is observed while the incident laser radiation of the fourth pulse reaches the sample surface. This implies no direct excitation by absorption of the radiation in the ablated matter which would probably lead to emission of black body radiation of the plasma or hot particles/vapor as it was observed by A. Semerok [14] for plasma shielding. Nevertheless, shielding is obviously happening because of the reduced ablation for both materials. This is why it must be assumed that the shielding performed by nonluminous matter which cannot be seen in this images.



 $\lambda = 1030 \text{ nm} | \tau = 2 \text{ ps} | F_0 = 5 \text{ J/cm}^2 | f_{rep} = 400 \text{ kHz} | f_{burst} = 40 \text{ MHz}$ 8 pulses per burst | material: copper

Fig. 9 ICCD images 1 ns before the fourth pulse, at the time of the fourth pulse and 1 ns after the fourth pulse of a burst with eight pulses on copper. The sample surface is marked by the red horizontal line. Furthermore, the area of the long-distance and surface-near luminous matter is marked in the images.

Despite the lack of direct excitation of the long-distance luminous matter the intensity of the area is increasing over the time of the burst. The reason for this is shown in Fig. 10 where four different times after the fourth pulse of an eight pulse burst on copper are imaged. 2, 12, 18 and 24 ns after the previous pulse the expansion of the surface-near and long-distance luminous matter can be seen. In the last image the beginning of the superposition of the two matter accumulations is imaged and the intensity is increasing locally. It becomes apparent that the long-distance luminous matter consists of the surface-near matter that expanse and propagates to this height which leads to an accumulation of this luminous matter and therefore indirectly excites this area.



 $t_{4th_pulse} + 2 \text{ ns}$ $t_{4th_pulse} + 12 \text{ ns}$ $t_{4th_pulse} + 18 \text{ ns}$ $t_{4th_pulse} + 24 \text{ ns}$

 λ = 1030 nm | τ = 2 ps | F_0 = 5 J/cm² | f_{rep} = 400 kHz | f_{burst} = 40 MHz 8 pulses per burst | material: copper

Fig. 10 ICCD images 2, 12, 18 and 24 ns after the fourth pulse of a burst with eight pulses on copper. The sample surface is marked by the red horizontal line.

To get a first look of this indirect excitation effect the front propagation velocity of the surface-near luminous matter was determined and is shown in Fig. 11. Therefore the time range from the arrival of the pulse to 10 - 20 ns after this moment was used for calculation, depending on how long the propagating front was clearly distinguishable before it merged with the long-distance luminous matter.



Fig. 11 Front expansion velocity of the surface-near luminous matter perpendicular to the sample surface for stainless steel and copper as a function of the pulse number.

For copper, the propagation velocity is lowest for the first pulse, increases until pulse number three and is levelling off to round about the value of the second pulse. A similar behavior can be assumed for steel, where the overshoot of the third velocity is less pronounced. One possible explanation of this behavior is that the first three pulses see another medium of propagation as the last pulses due to the influence of the ablation process to the atmosphere above the area of ablation. This is because of the slow propagation of longdistance luminous matter, which is still close to the surface for the first pulses of the burst as it can be seen in Fig. 12. For the last five pulses, the long-distance area has propagated far enough that these pulses see a less influenced medium of propagation so the propagation velocity levels out to a constant value. For single pulse ablation of copper with a pulse duration of $\tau = 50$ fs, wavelength of $\lambda = 800$ nm and a pulse energy of $E_p = 20 \mu J$ a similar behavior in a comparable order of magnitude was observed, with a velocity of propagation of 5.6 km/s in the first 15 ns which was then slowing down to approximately 1 km/s [14]. The reported slowdown could not be measured in this experiment because the front of the expansion was no longer clearly discernible and additionally after 25 ns the next pulse generated new ablation products.

As mentioned above, Fig. 12 shows the front propagation of the long-distance luminous matter as a function of time after the first pulse of the burst train. As a guide for the eye the points in time when the burst pulses arrive at the surface are marked with arrows. It can be seen that the front is pushed up a certain distance after a pulse has ablated material and settles to a slightly lower value just before the next pulse arrives. This behavior can be seen for all eight pulses until 50 ns after the last pulse when the front starts to propagate away from the sample surface again. This motion can be attributed to the expansion of the long-distance luminous matter and levels off to round about 400 µm for copper. For stainless steel a similar but less pronounced behavior is observed and the front comes to a halt at round about 300 µm instead. This weaker pronouncement is probably due to the lower single pulse fluence used for steel.



Fig. 12 Front expansion of the long-distance luminous matter perpendicular to the sample surface over time. The times of the burst pulses are marked as a guide for the eye. The error bars have not been drawn for better visibility, but are always below or equal to 5 % of the measured position.

In Fig. 12, a clearly recognizable front of the long-distance luminous matter was evaluated up to round about 375 ns after the first pulse applied. To evaluate the lifetime of this ablated material the image intensity of 90 µm above the sample surface and higher was integrated and applied over the process time as shown in Fig. 13. An exponential decay of the intensity can be observed and fitted. This allows the half-life of the process luminescence to be determined for both materials which are 117 ns for copper and 120 ns for stainless steel. The intensity of the process luminescence has therefore almost the same half-time even though the applied fluence for copper was round about 3.3 times the fluence used for stainless steel. This indicates that the more pronounced shielding and redeposition effects for copper are not represented by the intensity and lifetime of the process luminescence.



Fig. 13 Intensity decay behavior of the long-distance luminous matter for stainless steel and copper after a burst with eight pulses. ($\lambda = 1030 \text{ nm}, \tau = 2 \text{ ps}, f_{rep} = 400 \text{ kHz}, f_{burst} = 40 \text{ MHz}, F_{0_copper} = 5 \text{ J/cm}^2$, F_{0 steel} = 1.5 J/cm²)

4. Conclusion and outlook

In order to gain a better insight into the ablation and shielding effects during processing with ultrashort pulsed laser bursts in-situ imaging was performed to examine the process luminescence. In contrast to copper, stainless steel

shows only a weakly pronounced ablation efficiency decrease for an even number of pulses in the burst, whereas copper has a significant drop of ablation efficiency for an even number of pulses. For a more precise evaluation the ablated volume per pulse was calculated for both materials. For copper, a strongly pronounced fluctuation of the ablated volumes was observed which were even negative for even numbered pulses. For steel, an unexpected alternation in the ablated volume was found, as well. However, the observed effect is not strong enough to have a significant effect on the ablation efficiency. This behavior is expected to be the effect of shielding through ablation products of the previous pulse which are removed or pushed to the surface by the even numbered pulse so the next pulse is less shielded or not shielded at all. This leads to less ablation of the so shielded even numbered pulse or even to redeposition of material which correlates with the calculated ablation volumes per pulse in this work and would both decreases the overall burst efficiency. For copper this effect is already reported ([6, 11]) but that the ablated volumes per pulse of steel during burst processing show a similar but less pronounced behavior like copper is a new observation. This could indicate that the behavior observed for copper could be specific for metals in general and not only for copper.

For burst processing two spatial areas in process luminescence can be identified. The long-distance luminous matter regime with a high lifetime (120 ns half-time for eight pulses per burst) and a surface-near luminous matter which expanse and propagates fast from the surface. However, there is no visible direct excitation of the long-distance luminous matter by laser radiation. Only an indirect excitation takes place in which the surface-near ablated matter propagates into the long-distance area.

Contrary to all expectations, it was found that the intensity of the process luminescence is not correlated to the shielding effects and the ablated volume per burst pulse. Therefore the shielding effect that obviously takes place is not attributable to absorption in the process plasma. So the shielding of the pulse has to be attributed to reflection or scattering on plasma or particles. Also absorption of the pulse in ablated particles which is still not powerful enough for black body radiation is conceivable.

Furthermore, it should be kept in mind that the drop of efficiency measured on processed samples must not be only influenced by effects observable during a single pulse burst. Also cumulative effects can play a role that only appear during a complete structuring process and are not present in a single burst.

Further investigations for more precise statements are to be carried out. This includes, among others, in situ shadowgraphical imaging concentrating on the particle plumes and further investigations of the process luminescence with neutral density filters and shorter exposure time to examine a broader spectrum. Moreover, an in situ investigation of a complete structuring process could provide further insight.

Acknowledgments and Appendixes

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