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Near-THz bursts of pulses – Governing surface ablation mechanisms for laser material processing

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Title:

Near-THz bursts of pulses – governing surface ablation mechanisms for laser material processing

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Abstract:

Laser operation in bursts of pulses has recently attracted a lot of attention, as high average powers and pulse energies have become available. Bursts of pulses have become a mean to achieve different laser-matter interaction regimes with a single laser source. We have used an ultra-short pulse laser source in combination with an external array of birefringent crystals to generate near-THz bursts of single-picosecond pulses. Variability of the setup and high single pulse energy were exploited to generate bursts of up to 16 pulses at different delays between consecutive pulses. The experimental setup was applied for surface processing of different industrially relevant materials, including metals, ceramics, and polyimides.

The goal of this study was to find out if the reduced ablation thresholds using near-THz bursts of pulses as reported in previous studies of other group could be utilized for increased ablation efficiency. Our findings show that near-THz bursts of pulses interact with materials in a similar fashion as a single, albeit longer, pulse would. We observed a clear trend on all the tested materials, showing a declining ablation efficiency with increasing total burst duration at a fixed pulse repetition frequency. Findings contradict the results of other groups concluding on an improved efficiency of laser to material energy transfer using bursts at near-THz intra-burst repetition rates. A simple model describing ablation efficiency decline with pulse prolongation was used to compare the experimental data with theoretical predictions. Detailed analysis of surface ablation effects on metallic materials also revealed a presence of melting processes typically associated with laser pulses above 10 ps in duration as opposed to original 1.5 ps pulses.

Keywords:

bursts of pulses; ultra-short laser processing; surface effects; ablation efficiency

1. Introduction

Ultra-short pulsed lasers with pulse durations around 1 ps or shorter have become widely used in recent years in different industrial and scientific settings, from laser ablation processing applications [1–3], surface modification [4,5], and additive manufacturing [6] to two-photon imaging setups [7]. Numerous advantages were recognized in the use of ultra-short pulses, being either enhanced nonlinear effects, nearly non-existent heat effects on the material surrounding target zone, or the wide range of possible applications with a single laser source. Lately, a lot of attention has been focused on generating and applying bursts of pulses [8–13] as high average powers and pulse energies have become available. Those properties can be by itself hard to fully exploit due to increased thermal effects or plasma generation [2] and bursts of pulses are a possible solution.

Bursts of pulses are typically obtained either by splitting a single pulse [10,13] or by grouping consequent pulses together [9,11,12]. Different approaches have shown similar results (i.e. significantly lower ablation threshold per pulse compared to a single pulse) albeit explained with different physical mechanisms, such as heat accumulation [11], incubation [14], or ablation-cooling [9]. Dual and quad pulse bursts, achieved with single pulse splitting, were already used for different applications, including as a probe for energy absorption mechanisms [15], micromachining [16], laser-induced periodic surface structure formation [17] and multi-photon absorption enhancement [18]. The burst generation mechanism majorly affects the resulting intra-burst repetition frequency. While grouping of pulses typically results in intra-burst repetition frequencies up to 100 MHz, equal to the frequency of the mode-locked oscillator used, splitting a pulse in multiples may result in much higher frequencies. Bursts of pulses are gaining track also as an integrated mode of operation on newly developed laser sources in MHz repetition rates. In our experiments, bursts of 1 to 16 pulses at near-THz repetition rates (up to 440 GHz) were generated via series of birefringent crystals.

Materials of different compositions, from metals, through ceramics to polyimide materials, were used in our experiments as representatives of different widely used industrial applications. We have focused our research on surface ablation processing of five different materials, copper, Kapton, low-temperature co-fired ceramics material (LTCC), alumina ceramics Al_2O_3 , and Kovar, all of them widely used for scientific applications and relevant in the printed circuit board industry.

The goal was to design a new approach to fast and efficient material processing with ultra-short pulses. Other groups have reported on more efficient deposition of the laser energy into the material when bursts with a high number of sub-pulses are used [13]. Following from other studies [9–11,14,15], we have also predicted a possibility that a THz burst exhibits a similar interaction with the material as a proportionally longer single pulse under certain conditions. The latter assumption is in a disagreement with recent results of other groups on metal processing applications [13]. In order to evaluate the effect of the number of sub-pulses in a burst on ablation efficiency, we kept all other processing parameters constant. Laser parameters, scanning parameters and focusing optics were thus unchanged throughout the tests. The ablation efficiency was observed as a measure of the maximum possible total throughput and the resulting structure quality (surface roughness, signs of heat-related processes) as a measure by itself. Surface melting was correlated with an increased total duration of a burst and thus total duration of laser energy deposition.

In this study, we have successfully evaluated the effects of splitting a single high-energy ultra-short laser pulse into a near-THz burst. Within the range of experimental conditions, for bursts of 1-16 pulses at 50 GHz to 440 GHz repetition rates, we have identified the best conditions for both ablation efficiency and surface quality of the processed material. Results are in general showing a uniform tendency that an increased burst duration reduces the ablation efficiency at a constant burst energy. The findings are important for further integration of ultra-short pulsed laser sources in various industrial settings, where both throughput and final surface quality are significant. While near-THz bursts of pulses may be beneficial for additive manufacturing or nonlinear imaging, their generation with single-pulse splitting shows limited usability for material processing. Near-THz burst generation could be preferred in certain applications, if high peak power of a single pulse causes unwanted effects, but tends to lower the resulting structure quality due to unwanted surface phenomena.

2. Materials and methods

The experimental setup was designed and used in a way that mimicked actual industrial-environment approach to laser-based material processing. The experimental setup was at the same time isolating the effects of a changing number of pulses in a burst from all other variables.

We have used a NIR pulsed laser emitting 1.5 ps long pulses at a 1064 nm wavelength, operating at a 300 kHz repetition rate (labelled f in **Error! Not a valid bookmark self-reference. a**) at an average power of 16.0 W as measured on the sample. The result was a constant burst energy of about 50 μ J. The experimental setup was further formed by an array of four birefringent α -cut yttrium orthovanadate (YVO₄) crystals with a $\lambda/2$ waveplate in front of each crystal. With fine control of the incident pulse polarization each crystal can be set to either split a pulse into two equal sub-pulses or transmit the original unmodified pulse. The delay between sub-pulses depends on the different group velocities of the two orthogonal polarizations, one parallel to the ordinary crystal axis and the other parallel to the extraordinary crystal axis [19]. We have used four crystals of different thicknesses (2.9 mm, 5.75 mm, 11.5 mm, and 23 mm) in order to produce twice as long delays in every subsequent crystal compared to the previous one. Regardless of the number of active crystals, there are always two orthogonal polarizations present within a pulse burst of minimally two pulses. We have calculated the delays between sub-pulses in correspondence with material properties of the birefringent crystals. The difference between the ordinary refractive index $n_o(\text{YVO}_4 @ 1030 \text{ nm}) = 1.959$ and the extraordinary refractive index $n_e(\text{YVO}_4 @ 1030 \text{ nm}) = 2.168$ gives a time delay per unit of crystal length between the ordinary and extraordinary polarizations of about 700 fs/mm. We confirmed the predicted delays with an autocorrelator setup (example in **Error! Not a valid bookmark self-reference. d**, device used is pulseCheck SM 1600, APE Angewandte Physik & Elektronik GmbH).

The experimental setup allowed us to control the splitting of a single linearly polarized picosecond laser pulse into 1-16 sub-pulses forming a burst of pulses. Each sub-pulse has the same pulse duration as the original incident pulse and the pulse energy is equally distributed among sub-pulses. Different combinations from a single pulse to a near-THz burst of 16 pulses were formed, depending on the individual positions of $\lambda/2$ waveplates. Sixteen different sub-pulse configurations were generated, as shown in **Error! Not a valid bookmark self-reference. b**. Total burst duration is defined as a sum of single pulse FWHM and the longest sub-pulse delay measured peak-to-peak. Intra-burst repetition frequencies (labelled f_i in **Error! Not a valid bookmark self-reference. a**) achieved in the setup varied from 50 GHz to 440 GHz. Detailed schematics of 8-pulse burst generation at a 440 GHz repetition rate is shown in **Error! Not a valid bookmark self-reference. c**. Additional details about the preparation of the birefringent crystal array setup can be found in existing publications of other groups [19,20].

Bursts of 1-16 pulses were further guided to fast x-y galvo-scanners and were focused through a 163 mm f-theta lens onto the sample surface, resulting in a calculated laser spot size of approximately 20 μ m and a total burst fluence of about 16.5 J/cm². Scanners were set to generate long straight lines with a fixed scanning speed of 3000 mm/s, resulting in an approximately 50 % overlap of consecutive burst positions on material at an estimated laser spot size of 20 μ m. We kept a fixed average laser power, the laser repetition frequency, NIR wavelength, scanning speed, and focal distance throughout all the tests.

Each line was scanned multiple times until the resulting channel formed a cut through the complete material thickness. The number of scans was increased in steps of one until a through cut was achieved. Cutting was performed on five separate lines at each setting to eliminate possible measurement errors due to local material defects or possible laser instability. Relatively low overlap between consecutive burst positions on material was set to ensure minimal energy incubation between bursts.

Fig. 1: a) Experimental setup schematics. b) Different temporal distributions of 1 to 16 pulses within bursts, as used in experiments. c) Detailed schematics of single pulse to an 8-pulse burst transformation with the array of birefringent crystals. In this case, input polarization to Crystal 1 is set parallel to crystal's ordinary axis to render it inactive. d) Autocorrelation curve of a two-pulse burst at around 200 GHz repetition rate. Note that peak width in an autocorrelation curve is approximately 1.54-times wider than the actual pulse width and that a double-pulse burst translates to a triple-peak shape due to the nature of autocorrelation calculation.

Materials chosen for the laser processing experiments were metals (5 μm thick copper layer on a dielectric substrate and 20 μm thick Kovar-alloy metal foil), ceramics (200 μm thick un-fired green LTCC and 260 μm thick Al_2O_3), and a polyimide film material (150 μm thick Kapton). Thermal properties of those materials are gathered in Table 1. Thermal diffusivity is a measure of heat wave dissipation. It can be reasonably expected that a higher value of thermal diffusivity will result in a larger heat-affected zone in a homogeneous material under the same heating conditions. While this approximation does not hold true on a single picosecond time scale, it can be used at longer time scales, where heat affected zone is roughly the maximum of optical penetration depth and thermal diffusivity length [21]. Thermal diffusivity thus provides a guideline which materials will exhibit a greater drop of ablation efficiency with an increased pulse duration. This measure predicts that the greatest drop would be observed on Al_2O_3 , followed by Kovar, LTCC, and copper, with Kapton being affected the least. Ablation efficiency comparison was calculated from the number of scanning passes needed to create a cut, corrected for cut dimension changes where observed.

Material	Density (g/cm ³)	Specific heat capacity (J/kgK)	Conductivity (W/mK)	Thermal diffusivity $\cdot 10^5$ (m ² /s)
LTCC (DuPont data)	3.1	730	3-5	130-220
Al_2O_3 [22]	3.7-4.0	755	33	1090-1180
Copper [23]	8.95	383	386	11.2
Kovar	8.1	460	17	470
Kapton (DuPont data)	1.4	1090	0.12	7.7

Table 1: Overview of materials used in experiments and their thermal properties.

We have measured the resulting structures with high magnification optical microscopy in both bright field and dark field imaging modes (Olympus BX53M microscope with Olympus MPlanFL 50x objective). The optical measurements with focus plane recognition resulted in high vertical position sensitivity that were used for channel dimension measurements (ablation efficiency measurements in Fig. 2 and

Fig. 3) and edge roughness estimation (Fig. 4). All findings from optical observation have been further analysed and confirmed using scanning electron microscopy (SEM) imaging on Jeol JSM 5800 microscope. The increased resolution and magnification allowed us to recognize sub-micron sized debris deposition and to observe substrate surface topography.

3. Theory/calculation

We used a combination of two simple ultra-short pulse ablation models of metals to compare the measured relative ablation efficiency data with theoretical predictions. Ablation efficiency was defined as a ratio of the ablated volume V and the total input energy E needed:

$$\eta = \frac{V}{E}.$$

As the total energy per burst was kept at a near constant value during the experiments, the changes of the ablation efficiency directly reflect the effect of varying the total burst duration.

The first model predicts the dependency of E_{th} on the pulse duration for metallic targets:

$$E_{th} \propto \beta \tau^{1/4}$$

where β is a constant parameter and τ is the pulse duration. The equation is valid under an assumption of measurements done under a strong ablation regime, i.e. with pulse fluence above 1 J/cm² [24]. While the typical scaling law for pulses longer than 40 ps would be $E_{th} \propto \beta \tau^{1/2}$ [25], the ablation threshold changes at a lower rate in the intermediate pulse durations between 1 ps and 40 ps.

The second model predicts the ablation depth d as a function of the pulse energy:

$$d(E_p) = \frac{1}{\alpha} \ln \left(\frac{E_p}{E_{th}} \right),$$

where α is the optical absorption or heat penetration coefficient, E_p is the pulse energy, and E_{th} the threshold energy for ablation [26]. In this regime, both lateral dimensions of the ablation crater have the same functional dependency as the ablation depth [24].

Combining the equations above gives us the model equation to be used on our experimental data, treating a whole burst of pulses as a single longer pulse. We have used a normalized relative ablation efficiency, set to 1 for the single pulse measurements, for easier comparison of results measured on different materials. If the assumption to treat a whole burst as one pulse was correct, the graph would follow the functional shape of:

$$\eta_R = \ln^3 \left(\frac{A}{B + \tau^{1/4}} \right),$$

where recalculated constants are used as $A = E_p/\beta$ and $B = E_{th0}/\beta$ for purposes of nonlinear fitting algorithms. The assumption above is expected to hold true as long as the total burst duration stays short enough that plasma shielding is not significant yet and the surface of the material can still be considered stationary. This critical time scale can be approximated at about 100 ps [27,28], which is also above the 40 ps limit imposed by the first model assumptions.

Other groups have done calculations based on the two-temperature model and those results can be either reinterpreted or appropriately commented-on in light of our experimental results. Numerical results from Gaudio, et. al. [13], show that melting is achieved before the irradiation stops in case of multiple pulses in a burst, on the contrary to the results calculated for a single 650 fs laser pulse. This result by itself supports the hypothesis that a near-THz burst of pulses could act similarly to a single pulse with a matching total duration. Simulations done by other groups [29] on 10 ps pulses show an

equivalent behaviour – lattice temperature exceeds the melting temperature already during the laser pulse.

4. Results

The most uniform splitting of laser energy was achieved with 1.5 ps pulses repeating every 2.3 ps or at a 440 GHz intra-burst repetition rate. At a fixed intra-burst repetition rate, the changing number of pulses in a burst remains the only changing parameter in the experiment. Results presented in Fig. 2 are showing a uniform tendency that an increased burst duration reduces the ablation efficiency while keeping a constant burst energy. Thermal properties gathered in Table 1 suggest that at the longest available timescale in our experiments, around 40 ps, ablation efficiency would be reduced the most comparing to a single 1.5 ps pulse for Al_2O_3 , followed by Kovar, LTCC, copper, and Kapton. Comparison with the results gathered in Fig. 2 shows that this crude approximation holds true except for the LTCC material. Material processing of LTCC with a laser may also induce a localized co-firing process, thus altering the expected response of material to either a single-picosecond or a longer pulse.

Fig. 2: Graph of relative ablation efficiency as a function of burst duration at a 440 GHz intra-burst repetition rate for all five materials tested.

Further analysis was performed on metal targets that were chosen for high thermal conductivity, high rates of heat diffusion [23] and a well-defined transition in a molten state on surface [30]. Also, the process of electron-phonon energy transfer was measured to be on a typical time scale of $\tau=1-4$ ps depending on the laser pulse fluence used [31]. This is a good indicator that heat-related surface effects will become observable for time scales of energy deposition in the material longer than τ but shorter than the mechanical lift-off time. Simulation results show lift-off time of at least 35 ps for the spallation process in copper [32]. Detailed relative ablation efficiency data gathered on both metallic targets (copper and Kovar) is presented in Fig. 3. Best fits of the model function described in previous chapter are included. A clear trend of gradual efficiency decline is visible, albeit with some outlying points. The greatest outliers in both data series, points at duration values of 10.5 ps and 19.5 ps, were obtained by using a dual-pulse burst at a 110 GHz and 55 GHz respectively. Those two data point represent the least uniform regimes of energy deposition and larger deviations from the model can thus be expected. In this range of total burst durations the simple model describes the general trend of our experimental data rather well. We added renormalized experimental data for comparison as originally presented by Neuenschwander et. al. [33] on copper ablation with 500 fs to 10 ps single pulses. Our experimental data matches both the model curve and data from comparative research

within 20 % uncertainty range. This match is a confirmation that near-THz bursts of pulses interact with metallic materials similarly to extending a single laser pulse.

Optical surface analysis has also revealed typical signs of melting after processing with a burst of two or more pulses, while such signs were not present when using single pulses. An array of images in Fig. 4 a, corresponding to data points presented in Fig. 2, shows the resulting structure edge after processing. Dotted red rectangles are added to focus attention on the top edge, which exhibits a dark stripe visible on every image except for the first one. The dark stripe consists of solidified molten copper debris and is an indicator of surface melting processes taking place at all burst durations beyond the shortest one.

Fig. 3: Graphs of relative ablation efficiency as a function of total burst duration measured a) on Kovar and b) copper. Both graphs have added best fit of model function. Our data on copper ablation is compared with renormalized experimental data on copper ablation with various single pulse durations published in [33]. Due to different laser fluences used in the experiments, we set the relative efficiency to 1 at 1.5 ps pulse duration for comparison.

The surface roughness of structured slopes was also taken into account. While at sub-picosecond or single-picosecond pulse durations the graininess of surface is very fine (below 500 nm peak-to-peak), it becomes coarser due to surface tension effects if melting effects are present (above 2 μ m peak-to-peak). Comparison uses the structures done with the longest burst duration and the shortest respectively (Fig. 4 b).

Fig. 5: a) Effects of surface melting during burst-mode processing of copper for different burst durations with 440 GHz intra-burst repetition rate. Darkfield imaging, scale bar equals to 100 μm . b) Detailed comparison between the effects of the shortest and the longest burst durations from part a. Increased surface roughness is visible in highlighted regions at the longer burst duration. Darkfield imaging, scale bar equals to 50 μm .

SEM imaging reveals that signs of melting are virtually not present when using a single 1.5 ps pulse. With splitting of pulses in progressively longer near-THz bursts of pulses, signs of surface melting, melt ejection, and subsequent re-solidification on nearby surfaces are becoming increasingly common, as seen on SEM images in Fig. 5. Effects on the underlying substrate material are also changed due to an increased burst duration, going from visible cracks at 1.5 ps and 3.8 ps to pronounced melting effects beyond 17.3 ps (zoomed view in bottom right panel of Fig. 5). Crack formation may be related to high temperature copper plasma formation above the substrate. Heat effects are correlated directly to energy dissipation during illumination with high-energy laser light.

Fig. 4: SEM images of laser processed traces at different total burst durations. Orange dashed lines denote the total channel width at the top surface. Effects of copper surface melting and heat effects on the substrate become clearly visible at longer durations as re-deposited debris outside the orange lines and as thermal damage to the substrate (zoomed images in bottom right for comparison).

5. Discussion and conclusions

In order to understand the discrepancy between our results and conclusions on more efficient deposition of the laser energy into the material with longer bursts by Gaudio et. al. [13], we have done an extensive search through literature to identify comparable experimental setups and results. To our best knowledge, comparable experiments were mostly not done yet, with a notable exception of Chowdhury et. al. [16] and Gaudio et. al. [13]. Chowdhury et. al. used quad pulse bursts with 1 ps intra-burst delays in the experiments. Results showed similarly shaped structures during laser ablation processing of stainless steel with quad pulse bursts and with single pulses at equal combined energy, albeit the bursts exhibited a lower ablation efficiency. Also, results presented by Gaudio et. al. on threshold fluence as a function of the total number of sub-pulses can also be recalculated from sub-pulse threshold fluence currently presented to the total burst fluence. The stated 93 % ablation threshold energy reduction when using a 32-pulse burst with 1.5 ps delay between consecutive pulses becomes a 110 % increase from a perspective of a burst as a whole. This perspective is in line with our findings unlike the finding of more efficient energy deposition from the same article.

Individual results of other groups, mentioned before, are in line with results of our extensive testing of different materials and different delays between sub-pulses, altogether showing a decreased ablation efficiency with different kinds of near-THz bursts. While any kind of splitting a single ultra-short pulse in a burst does not appear beneficial for either the resulting structure quality or ablation efficiency optimization, a laser source natively capable of operation at given conditions might be beneficial. In such case, near-THz bursts of pulses enable an increase of overall energy delivered to the material and thus increase the total throughput albeit at a lower efficiency of operation.

We have demonstrated that splitting a single pulse into a burst of 2-16 sub-pulses does not increase the ablation efficiency nor the final structure quality as longer bursts in general appear to interact with both metallic and non-metallic materials similarly to a single, proportionally longer pulse.

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