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Influence of the repetition rate and pulse duration on the incubation effect in multiple-shots ultrafast laser ablation of steel

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Abstract

We report on an experimental study of the incubation effect during laser ablation of stainless steel with fs- and ps-pulses at high repetition rates. Ablation thresholds for multiple pulses N have been estimated. As expected, the ablation threshold decreases with N due to damage accumulation. The related incubation coefficient has been determined at different repetition rates, from 50-kHz to 1-MHz and two pulse durations: 650-fs and 10-ps. Results show that the incubation effect is lower for fs-pulses below 600 kHz. At higher repetition rates incubation is more pronounced regardless of the pulse duration, probably due to heat accumulation.

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1. Introduction

Ultrafast laser micromachining is an emerging technology enabling fabrication of precise microstructures via laser ablation on a variety of materials. Despite its unique capabilities in terms of flexibility, accuracy and reliability, this technology still needs to boost its process efficiency to be established on an industrial scale. A

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number of studies have already been devoted to maximize the volume ablation rate by optimizing the process parameters settings for different materials and applications [1-4]. Recent development of fs and ps laser sources operating at repetition rates of hundreds of kHz up to a few MHz and delivering several tens of watts of average power, has paved the way towards the scaling of productivity. Nevertheless, many studies have revealed that by increasing the repetition rate, plasma and/or particle shielding and heat accumulation may limit the laser ablation efficiency and the achievable precision, respectively [2,5]. Using femtosecond pulses instead of picosecond ones may help to prevent heat accumulation at high repetition rates, mainly in case of metals with a relatively low thermal conductivity like stainless steel [6]. Nonetheless, as far as the pulse energy is increased, melting, which is detrimental to accuracy, cannot be avoided in multi-pulses femtosecond laser ablation processes.

It has been experimentally demonstrated that the ablation threshold of metals irradiated by bursts of fs or ps pulses is lowered as far as the number N of consecutive pulses impinging on the same spot area is increased [4,7-10]. This effect has been defined in literature as incubation but the physical mechanisms responsible for it are still debated among the scientific community. Several interpretations have been proposed. For metals, incubation effect was ascribed to the accumulation of laser-induced chemical and/or structural changes of the material and/or plastic deformation of the surface [7]. Such defects, generated by the first impinging pulses, facilitate absorption of the next coming laser pulses, thus enhancing ablation and material removal. In case of bursts with a significantly high number of pulses N , heat cumulative processes could be responsible for a reduction of the ablation threshold as well. This effect could be even more crucial as far as the repetition rate approaches the MHz regime. Here, the very short time interval between subsequent pulses prevents heat dissipation into the bulk material through thermal conduction. As a result, the local temperature rise caused by the first pulses could generate better absorption conditions for the following ones thus lowering the ablation threshold. The dependence of the ablation threshold on N is modelled by a power law equation defining an incubation coefficient S [8]. Incubation is absent for $S=1$ and the ablation threshold stays constant irrespective of the number of pulses. As far as the damage accumulation mechanism reduces the ablation threshold, the incubation coefficient gets lower [7-10]. Therefore, the incubation effect is expected to have a crucial role in enhancing ablation efficiency when using high-repetition-rate ultrafast lasers. Previous studies have investigated the incubation effect using femtosecond and picosecond laser sources operating at repetition rates ranging from 100 Hz [7] up to several kHz [4]. At such a low repetition rates the heat accumulation mechanism contribution to the incubation effect cannot be accurately evaluated.

In this work, we present a systematic study of the S coefficient during multiple shots laser ablation of stainless steel targets with femtosecond and picosecond pulses at repetition rates from 50 kHz up to 1 MHz. The influence of the repetition rate and pulse duration on the incubation mechanisms responsible for the reduction of the ablation threshold has been investigated, focusing on potential contributions of heat cumulative processes.

2. Experimental

Ablation experiments were carried out using a fiber laser amplifier from Active Fiber Systems GmbH, based on the chirped pulse amplification technique (CPA). This laser source delivers an almost diffraction limited beam ($M^2 \sim 1.25$) at a wavelength of 1030 nm, with laser pulses of tunable widths in the range from 650 fs to 20 ps and repetition rates programmable from 50 kHz to 10 MHz. At lower repetition rates the maximum pulse energy is limited to 100 μ J, while at higher repetition rates the maximum available average power is 50 W. The laser source is equipped with an external Acousto-Optic Modulator (AOM) that allows the generation of bursts of any desired number of pulses. The linearly polarized beam exiting from the laser source was first converted to circular polarization by means of a quarter-wave-plate and then expanded to a

diameter of about 10 mm. We employed a computer interfaced galvoscanner equipped with a F-Theta lens with a focal length of 100 mm to focus the laser beam onto the surface of 2-mm-thick stainless steel (AISI 304) targets that had been previously cleaned with acetone. The experimental setup is sketched in Figure 1.

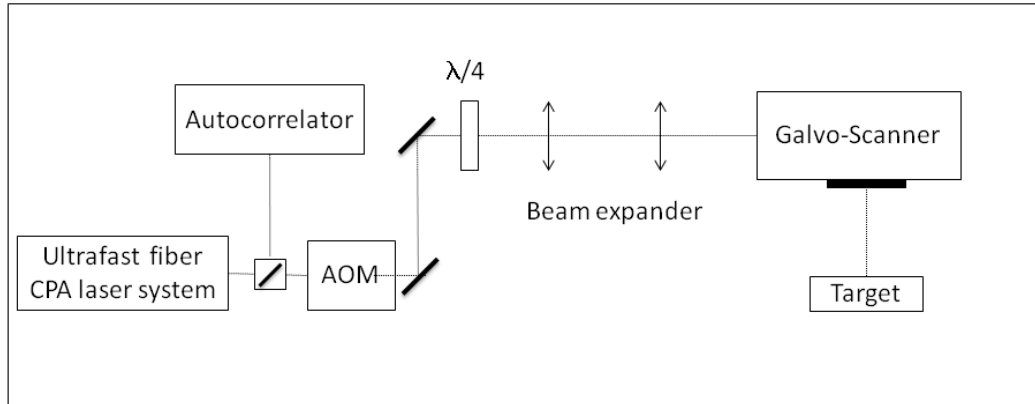


Fig. 1. Schematic of experimental set-up.

Ablation experiments were performed in ambient air without any gas shielding, by irradiating the sample surfaces with selected number of pulses ($N= 2, 5, 10, 25, 50, 250, 500, 2500, 25000, 250000$) at different pulse energies from $1 \mu\text{J}$ to $50 \mu\text{J}$ and repetition rates ranging from 50 kHz up to 1 MHz. The same experiments were replicated for two different pulse durations: 650 fs and 10 ps, as measured by an autocorrelator. Five craters were produced for each combination of process parameters and their diameters and morphologies were measured and observed by scanning electron microscopy. Thus, we calculated a crater diameter average value and an associated variance for every different set of operating conditions used in our experiments.

3. Results and discussion

The focused laser spot size on the sample surfaces w and the ablation threshold fluence Φ_{th} was estimated for each number of pulses N , using the method proposed by Liu [11], starting from the measurement of the craters diameters, being supposed a Gaussian beam profile for our laser.

The squared diameters of the ablated area D^2 are related to the peak laser fluence Φ_0 on the sample surface by the following equation:

$$D^2 = 2w^2 \ln\left(\frac{\Phi_0}{\Phi_{th}}\right) \quad (1)$$

where the peak laser fluence is defined by:

$$\Phi_0 = \frac{2E_p}{\pi w^2} \quad (2)$$

with E_p being the incident laser pulse energy. Combining equations (1) and (2) it is possible to relate the crater diameters, directly measured onto the sample surfaces, to the applied laser pulse energy. The linear

fitting of the squared diameters D^2 versus the logarithm of the pulse energy E_p allows determining the laser spot radius and the threshold fluence from the slope and the ordinate-intercept of the fitting line, respectively. This method for estimating the ablation threshold [11] is particularly advantageous thanks to its simplicity and because it does not require a characterization of the laser beam. Furthermore, the laser spot size on the sample surface can be determined regardless the specific optical arrangement.

The same procedure has been applied to determine the ablation threshold for each number of pulses N , repetition rate and pulse duration. Among all the examined process parameters combinations, we report in Figures 2a,b,c and d, as representative, the plot obtained for $N=5, 50, 500, 25000$, repetition rates of 200 kHz and 1MHz and pulse widths of 10ps and 650fs. It is clearly evident that the crater diameters increase either by increasing the pulse energy or the number of pulses. These results are consistent with the theory of damage and/or deformation accumulation mechanisms in multi-shots irradiation [8].

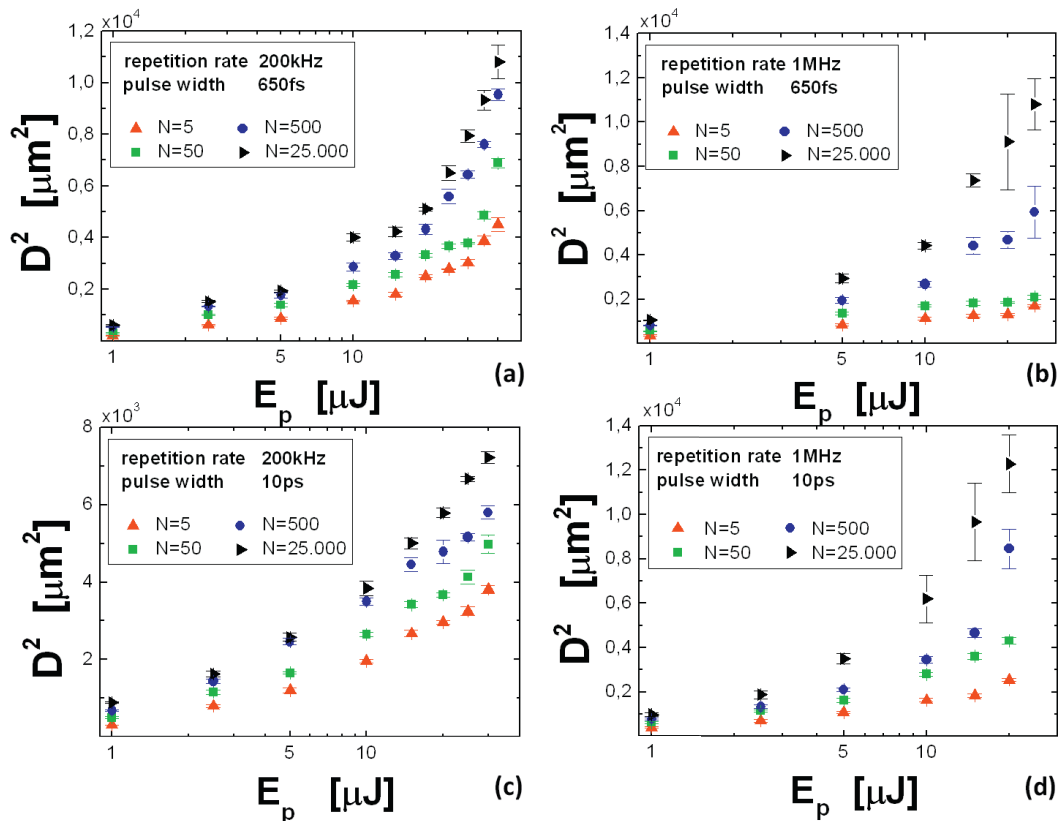


Fig. 2. The squared diameter of the ablated craters in stainless steel (AISI 304) versus pulse energy for $N=5,50,500,25000$ at (a) 200kHz and 650fs; (b) 1MHz and 650fs; (c) 200kHz and 10ps; (d) 1MHz and 10ps.

Furthermore, the slope of the linear fitting becomes steeper when the pulse energy exceeds a typical value of $15\mu\text{J}$. The slope change is more evident for $N > 500$ incident laser pulses. This behavior has been already observed in previous analogous experiments [4,7] and was ascribed to a transition from a “gentle” to a “thermal” ablation regime. Figure 3 shows the SEM images of the ablated craters produced for a high number of laser shots (25000) at two repetition rates, 200kHz and 1 MHz, and different pulse energies. At the lowest

pulse energy of $1\mu\text{J}$ the ablated areas are characterized by rippled structures inside the craters for both the repetition rates (Fig. 3a and Fig. 3c), thus indicating that no melting occurs originated by heat accumulation, despite a reduction of a factor of five of the time interval between subsequent pulses from 200 kHz to 1 MHz. As far as the pulse energy is increased the thermal ablation regime is established and the craters morphology changes, exhibiting smoother inner surfaces and molten and resolidified material on the edges. Figure 3b shows that such a transition is observed at $30\mu\text{J}$ of pulse energy and 200kHz of repetition rates. Due to heat accumulation, melting already starts at $10\mu\text{J}$ when the repetition rate is increased up to 1 MHz, as showed in Figure 3d.

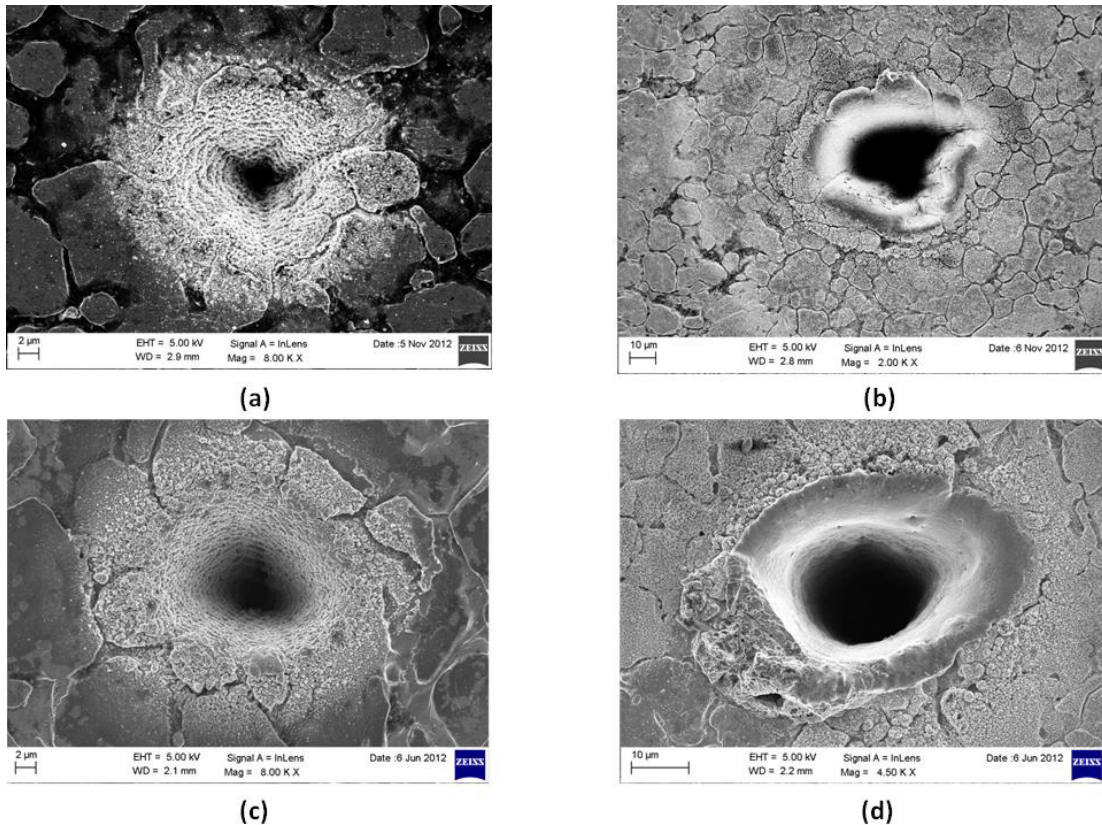


Fig. 3. SEM images of craters produced in stainless steel (AISI 304) at 25,000 shots, 650fs pulse width, (a) 200kHz and $1\mu\text{J}$; (b) 200kHz and $30\mu\text{J}$; (c) 1MHz and $1\mu\text{J}$; (d) 1MHz and $10\mu\text{J}$.

In order to better estimate the ablation thresholds for each operating condition, we excluded from the linear fit of the experimental data the craters affected by a significant amount of molten material that indicates a thermal ablation regime.

The multi-shots threshold fluences were thus determined for each number of incident laser pulses, pulse duration and repetition rate. Figure 4 reports the ablation thresholds versus the number of applied laser shots, at the lowest and highest repetition rates (50kHz and 1MHz) and two different pulse widths. It was found that the ablation threshold is lower for higher number of pulses and the influence of pulse duration is negligible. Such a trend has been observed for all the repetition rates. This behavior is consistent with an incubation

model. It is worth noting that for $N > 1000$, a saturation of the incubation effect occurs and the multi-shots threshold fluence does not decrease by further increasing the number of laser shots.

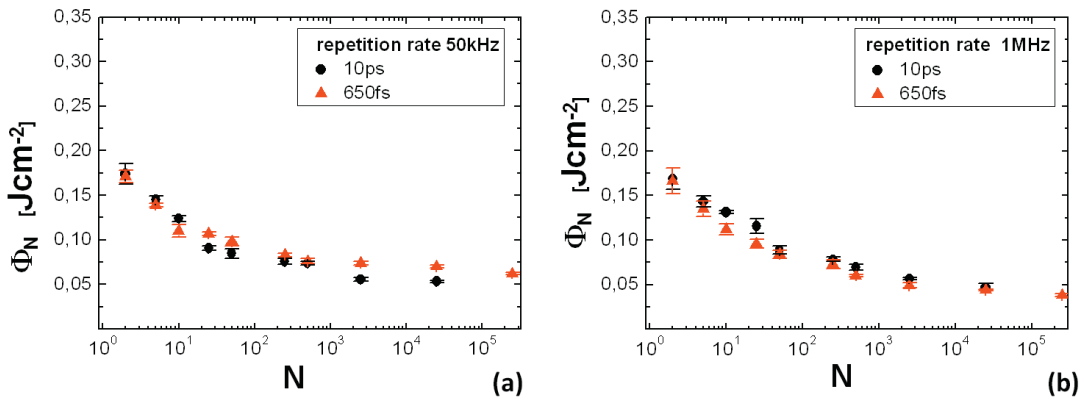


Fig. 4. Multi-shot threshold fluence versus the number of applied laser pulses in stainless steel (AISI 304) sample at two pulse widths of 650fs and 10ps at (a) 50kHz; (b) 1MHz.

Figure 5 represents the multi-shots threshold fluence as a function of the repetition rate, for a relatively high number of pulses ($N > 250$) and two diverse pulse durations.

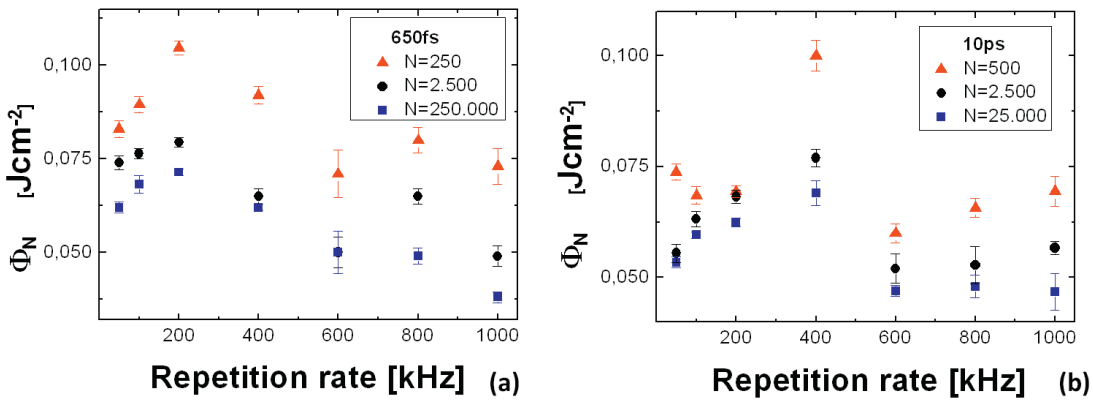


Fig. 5. Multi-shot threshold fluence as a function of repetition rate at three different number of incident pulses in stainless steel (AISI 304) sample for pulse width of (a) 650fs; (b) 10ps.

An increase of the ablation threshold is observed from 50 kHz to 200-400 kHz. This behavior could be ascribed to the particle shielding mechanism. This effect has been already observed in previous experiments performed under analogous experimental conditions [5,6]. Here, the particles ablated and ejected by the first impinging laser pulses reside on top of the irradiated area and do not fade before the coming of the subsequent pulses, thus scattering, absorbing or reflecting part of their energy. In this way, the ablation threshold is enhanced. This effect is even more critical in case of a higher number of incident pulses. For repetition rates higher than 400kHz a decrease of the multi-shots ablation threshold is found, corresponding to the onset of heat accumulation effects overbalancing particle shielding. Melting originated by heat cumulative processes is

facilitated by longer laser pulses, as clearly shown in Figure 6, where SEM images of two craters obtained under similar process conditions and different pulse durations are depicted. In case of picosecond pulses, melting and resolidification around the crater edges (Fig. 6b) are noticeable if compared to the femtosecond case (Fig. 6a).

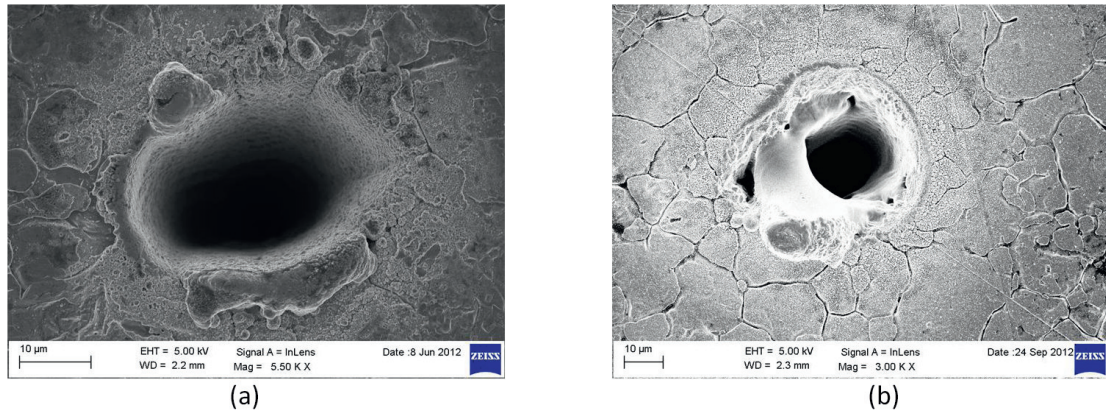


Fig. 6. SEM images of craters produced in stainless steel (AISI 304) at $N=2500$ shots, pulse energy of $10\mu\text{J}$, repetition rate of 800kHz and pulse width of (a) 650fs and (b) 10ps .

Our experimental results confirm that damage and/or heat accumulative mechanisms are responsible for a lowering of the multi-shots ablation threshold with increasing number of incident pulses N . This effect has been described in literature by an incubation model [8] relating the multi-shots threshold fluence Φ_N to N by the following law:

$$\Phi_N = \Phi_1 N^{S-1} \quad (3)$$

where Φ_1 is the ablation threshold for $N=1$ and S is the so-called incubation coefficient. Equation 3 can be expressed in the following form:

$$\ln(N\Phi_N) = S \ln(N) + \ln(\Phi_1) \quad (4)$$

from which it is possible to calculate the S coefficient and the single pulse ablation threshold by plotting the first member of Equation 4 versus the logarithm of N for all number of pulses. The incubation coefficient represents the slope of the best fitting line of the data, while the single pulse ablation threshold can be calculated from the ordinate-intercept.

Typical values of S in the range between 0.8 and 0.9 were found with this method in case of multi-shots laser ablation of metals at relatively low repetition rates ($< 100\text{ kHz}$) [4,7]. We have calculated the incubation coefficient also at higher repetition rates aiming to see if in the MHz regime the heat accumulation effect may affect the incubation mechanism. We have also compared S -values obtained with femtosecond and picoseconds pulses.

The plots of the experimental data obtained at two different repetition rates of 50kHz and 1MHz and pulse durations of 650fs and 10ps are shown in Figure 7 with a calculated linear fit regression coefficient R^2 of 99.8%, thus confirming the validity of the incubation model for ultrafast laser ablation of metals described by Equations 3 and 4.

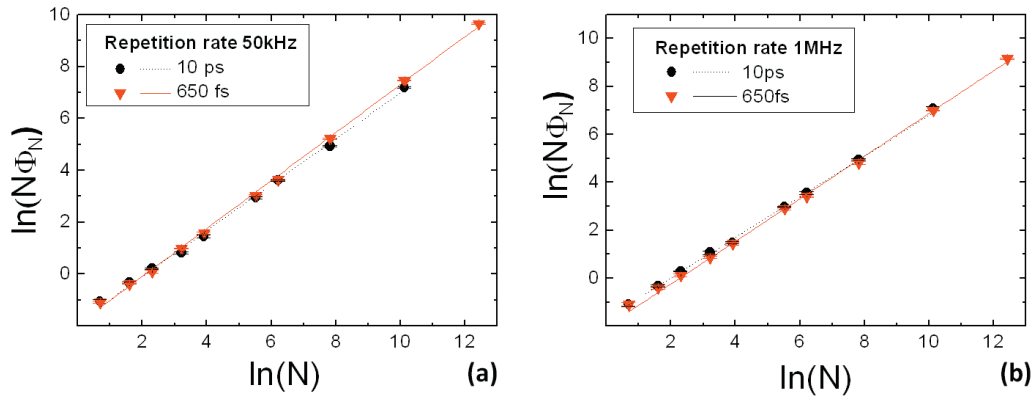


Fig. 7. Accumulation curve for stainless steel (AISI 304) sample for 650fs and 10ps pulse width and at (a)50kHz and (b)1MHz. The solid line represents the linear best fit of the experimental data according to equation 4, with a calculated R^2 -value of 99,8%.

The estimated incubation coefficients S as a function of the repetition rate, for the two different pulse widths, are presented in Figure 8. The S value is lower than 1 in all cases, which indicates a decrease of multi-shots threshold fluence with the rise of the number of laser pulses. In some cases, the damage threshold at several tens of thousands pulses is 3 or even 4 times lower than the single-shot ablation threshold.

By comparing results obtained with picoseconds and femtosecond pulses it can be noticed that at lower repetition rates, below 600 kHz, the incubation effect is less evident in case of shorter pulses.

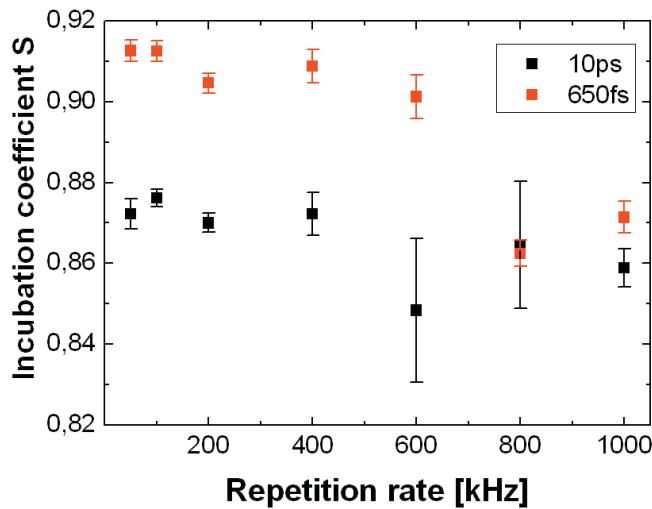


Fig. 8. Incubation coefficient S as a function of the repetition rate for multi-pulse laser ablation of stainless steel (AISI304) targets with 650-fs and 10-ps laser pulse widths.

As far as the repetition rate is increased above 600 kHz, the damage accumulation mechanism is even more pronounced and a significant drop of the incubation coefficient was observed for both pulse durations even though for longer pulse durations the decrease of S takes place at lower repetition rates. Nonetheless, above

800 kHz, when the heat accumulation effect prevails, similar values for S were found for picoseconds and femtosecond pulses. Although a detailed investigation of the microscopic origin of the incubation effect during multi-shots laser ablation of metals was beyond the scope of this work, our results suggest that in case of very high repetition rates, heat accumulation plays an important role in the incubation mechanism whatever its origin, whether due to damage accumulation or rather to plastic deformation [4].

4. Conclusion

We performed a systematic study of the incubation effect during multiple-shots ultrafast laser ablation of steel target. This effect leads to a lowering of the ablation threshold with increasing number of incident pulses. The physical mechanisms behind this effect are still not clear. Some authors ascribe incubation to plastic deformations [7], others to laser induced defect accumulation [4]. We focused our experimental investigation on the influence of the repetition rates, in the range from 50 kHz to 1 MHz, on the incubation effect. A comparison of the results for two different pulse durations of 650-fs and 10-ps was also reported. Ablation thresholds for several numbers of laser shots, from N=2 to N=250000, have been estimated. It was confirmed that the multi-shots threshold fluence decreases with growing number of incident laser pulses.

This behavior is described by the incubation law defining the so-called incubation coefficient S. We studied the dependence of the S coefficient from the repetition rate, finding that it is nearly constant at low repetition rates while it decreases approaching the MHz regime, thus indicating that, at such high repetition rates, incubation mechanisms are facilitated resulting in a reduction of the ablation threshold of a factor of 3 or 4, when using several thousand pulses instead of single-shot. The incubation effect is less evident in case of shorter pulses, although for repetition rates higher than 800 kHz S-values calculated for femtosecond and picoseconds pulses are similar. SEM analyses of the craters morphology showed that at such a high repetition rates a thermal ablation regime establishes and the ablated areas are characterized by a huge amount of molten and resolidified material originated by heat accumulation. Therefore, it can be argued that under these operating conditions, heat accumulation plays an important role in the incubation mechanism.

Further investigations are still needed to better understand the microscopic and physical origin of the incubation effect. For this purpose, analyses with a confocal microscopy are currently in progress on the ablated craters. Nonetheless, our results are very interesting from an application point of view, because a reduction of the ablation threshold by multi-shots irradiation might be useful to increase the material removal rate during industrial laser milling processes using high repetition rate ultrafast laser sources.

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