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Ultrashort pulsed laser ablation of stainless steels

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ABSTRACT

This research investigated the ablation process of commercial stainless steels with ultrashort pulsed laser. Square hollows were ablated on stainless steel sheets using a picosecond laser with pulse duration of 0.25, 1 and 10 ps, respectively, and fluence ranging from 0.125 J/cm² to 5 J/cm². For each setting the surface quality and ablation efficiency were determined by optical microscopy and scanning electron microscopy (SEM). Processing windows producing high quality surfaces at high ablation efficiency were found at a fluence around 0.75 J/cm² for the two shorter pulse lengths tested. Individual cones and periodic cone-like protrusions were found in the low fluence regime (< 0.625 J/cm²) and high fluence regime (> 0.875 J/cm²), respectively, both of which make the ablated surface rough. Emphasizing on the individual cones, it is found that the cones are caused by inclusions in the base materials, attributed to a higher fluence threshold required for ceramic-like inclusions. This novel theory explaining the creation mechanism is verified by multiple analysis methods like SEM and energy dispersive X-ray spectroscopy and stop-motion SEM imaging.

1. Introduction

Ultrashort pulsed (USP) laser ablation is an advanced processing technique that, for its cold-ablation capabilities and high accuracy, is increasingly used in industrial applications, such as micromachining, surface micro-patterning and 3D fabrication of a wide range of materials including metals, ceramics, semi-conductors and plastics [1–4]. Extensive research has led to a better understanding of the occurring physical phenomena along with improved ablation efficiency, reliability and decreasing investment costs [5,6]. Much of the current research on USP laser ablation focuses on understanding the mechanisms governing the ablation of material.

Laser ablation is the removal of material from a substrate by direct absorption of the laser energy. This can be achieved by intense continuous wave irradiation; however, pulsed lasers are more suitable to achieve ablation instead of melting [7]. Ablating metallic targets, known for their large thermal diffusivities and low melting temperatures, with longer pulses (ns) will result in large heat affected zones (HAZ) and molten regions [8]. This research uses an USP laser to ablate stainless steel. The ultrashort pulses used in this research deposit the energy of the laser pulses within a short excitation time, which has two effects. Since the excitation time is shorter than the thermalization time in the material, non-thermal ablation mechanisms can occur that

restrain the material to reach its thermodynamic critical temperature and thus explosive boiling called phase explosion [7]. Secondly, the volume of material directly excited by USP laser has less time to transfer energy to the surrounding material before being ejected [7]. Because of these two effects, USP lasers are capable of high efficiency ablation with minimal HAZ producing micro-sized high quality structures even in metallic targets [9,10].

A large body of research was focused on the study of laser induced periodic surface structures (LIPSS). LIPSS are USP induced surface structures that form a regular pattern on the surface of the treated material. The wavelength of LIPSS can be either in the same order as the wavelength of the used lasers (λ), which is then named *low spatial frequency LIPSS* (LSFL). Or the wavelength of LIPSS is significantly smaller than that of the laser wavelength, referred as *high spatial frequency LIPSS* (HSFL). The LIPSS are usually oriented perpendicular to the laser polarization direction [2,11]. Currently there is agreement on the mechanism governing the occurrence and behavior of LSFL, i.e. these structures are generated by interaction of the incident laser beam with an electromagnetic wave scattered at the rough surface [12]. HSFL is usually formed in the femtosecond pulse duration range and the fluence has to be close to the ablation threshold [12,13]. At present there is no agreed view on the mechanism causing HSFL. Some of the current hypotheses are self-organization as suggested by Reif et al. [14]

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or the ablation of oxidation of the surface layer suggested by Li et al. [15]. LIPSS can be a desired outcome, e.g. to change the hydrophobic properties [16,17], frictional properties [18,19] or color of a surface [20]. However, in the case of laser ablation of highly accurate, high surface quality components, LIPSS is undesired.

Several works have reported a gradual change from LIPSS to cones and/or holes in the surface upon increasing fluence, called *cone-like protrusions* (CLP) [21,22] or periodical spikes [23]. Compared to LIPSS, the scale of CLP is much larger in both of height and periodicity, i.e. micrometers sized instead of (sub-)wavelength of the incident radiation. In some cases CLP is beneficial, e.g. for multi-scale structuring of surfaces, involving both LIPSS and CLP, to create super-hydrophobic properties [24]. However, for the targeted application, CLP is unwanted, since it makes it impossible to machine flat surfaces and sharp edges at high precision [17,21,22,25–27].

In the case of fast production of accurate products with high quality surfaces, both LIPSS and CLP are an unwanted side-effect. However, several authors have shown that these effects are very pronounced for stainless steel, occurring in a wide range of fluences and pulse-widths [16–18,20,23–25,28]. Furthermore, this research has shown the occurrence of what we call *inclusion induced cones* (IIC), which are large macroscopic defects in the form of protruding cones on the ablated surface. The IIC, LIPSS and CLP create a challenge in the form of finding a suitable process window, with which accurate and high-quality surfaces can be ablated with high efficiency. In particular, there is no report and a lack of understanding about the cause of IIC and how to prevent it.

Therefore, the current research investigates the ablation behavior of stainless steels AISI 301 and 316 VR using USP ablation. Amongst others, fluence and pulse duration are varied to find the laser settings that produce high quality surfaces at high ablation efficiency. Furthermore, the occurrence of IIC is investigated using multiple methods and a novel theory is proposed for the creation mechanism of this surface effect. The findings of this research can aid in the selection of a suitable USP-laser source to improve the adoption of USP-technology in industrial processes.

2. Experimental details

2.1. Material

For the ablation process, two kinds of stainless steel sheets were used, namely AISI 301 stainless steel sheet 0.4 mm thick and AISI 316 VR stainless steel sheet 0.4 mm thick. The latter has a comparable chemical composition and has received a vacuum remelting treatment to reduce the amount and size of non-metallic inclusions [29,30]. The chemical compositions of the two steels used for this research are listed in Table 1. The cold rolled steel sheets had a surface roughness R_a between 0.145 and 0.197 μm .

2.2. Picosecond pulse laser and ablation

A Pharos 10 W ultrashort pulse laser from Light Conversion was used for ablation processing. It was capable of generating pulses tunable between 0.2 and 10 picoseconds. The repetition rate was tunable between single pulse and 1 MHz. The wavelength of the laser beam was 1030 nm, and the second harmonic (515 nm) was used for this research.

The wavelength conversion was performed extra cavity by harmonic generation using non-linear optics. The maximum average output power was approximately 6 W after wavelength conversion, as was confirmed by a Coherent Powermeter. The laser light was linearly polarized and directed to the galvoscaner without using a beam expander. A beam scanner Hurriscan II from Scanlab with a maximum scan speed of 2.5 m/s was used for this research. An F-Theta lens made by Sill Optics designed for wavelengths between 515 nm and 545 nm focused the laser beam with a focal length of 163.9 mm. The beam profile was measured using a Primes MicroSpotMonitor that is able to measure the real spot diameter at multiple points in the z-direction. This showed that the beam had a Gaussian profile with $M^2 < 1.2$ and $1/e^2$ spot diameter (D) of 27 μm .

To investigate the effect of laser pulse duration on the ablation quality and efficiency, three pulse durations 0.25, 1 and 10 ps were used for ablation processing. A range of laser beam fluence was investigated by tuning the output power of the laser with the fixed beam spot-size D . The fluence was calculated by dividing the pulse energy by the effective spot area ($\pi D^2/4$). The pulse energy was defined by the average laser power divided by the repetition rate (RR). The surface of the steel sheets was ablated with multiple scans until a 1 mm square hollow with a depth of approximately $D_p = 150 \mu\text{m}$ was created, the exact depth was determined using optical 3d metrology using a Alicona InfiniteFocus. This depth was used for efficiency calculations where protruding surface effects were ignored for their impact on the ablated volume was deemed insignificant. By scanning, the laser beam followed a single direction raster scan pattern with a hatch distance of 5 μm . Every layer was rotated with 90° with respect to each other, after the 4th layer, the 90° was increased by respectively 2° , 3° , 1.5° and 2.5° (i.e. yielding a total rotation of layer 5 of 92° with respect to layer 4 and of layer 6 of 93° with respect to layer 5 and so on). This pattern rotation was performed to prevent periodicity in the processed surface. The repetition rate of the laser was set to 200 kHz with a scan speed of 0.5 m/s, which results in a pulse overlap of 90%. To reach higher fluences the repetition rate was decreased to 50 kHz because of the power limitation of the laser. The tested repetition rates (50 kHz and 200 kHz) were relatively low and therefore pulse-to-pulse interaction in the form of particle shielding or heating effects were insignificant [2,6]. When using the lower repetition rate of 50 kHz, the scan speed was accordingly decreased to 0.125 m/s to ensure the same pulse overlap. The laser ablation settings used for the experiments are listed in Table 2.

The bottom face of the hollows was investigated using a scanning electron microscope (SEM, Philips XL30F equipped with a field emission gun) and X-ray energy dispersive spectroscopy (EDS) (EDAX, type XL30/40 132–10, equipped with a super ultra-thin window (SUTW 3.3) and an active area of 10 mm², amplifier model 204b) to quantify the ablation quality, efficiency and composition, and thereby determine the optimal settings for the ablation of stainless steel AISI 301. CLP average height and spacing were determined on the cross-sections of the samples (prepared parallel to the polarization direction). The height of individual cones was measured by measuring the distance from the tip of the cone to the bottom on both sides of each cone. The spacing was defined as the distance from the center of an individual cone to the center of the adjacent cones. An example of CLP-spacing and height measurement can be found in the insert of Fig. 2b. For each ablation setting, a section of 168 μm width were measured to get the average height and spacing of CLPs.

Table 1
Chemical composition of used material according to ISO 15510:2014.

Composition (%)	C	Si	Mn	P	S	Cr	Ni	Mo	N
AISI 301	0.09	1.15	1.21	0.026	0.001	16.0–19.0	6.0–9.50	0.80	0.10
AISI 316 VR ^a	0.08	1.00	2.00	0.045	0.030	16.0–18.0	10.0–14.0	2.00–3.00	0.10

^a Vacuum remelted.

Table 2
Laser beam ablation settings.

Fluence [J/cm^2]	Pulse duration, τ [ps]	Repetition rate [kHz]	P_{avg} [W]
0.25	0.25, 1, 10	200, 200, 50	0.29, 0.29, 0.07
0.5	0.25, 1, 10	200, 200, 50	0.58, 0.57, 0.15
0.75	0.25, 1, 10	200, 200, 50	0.84, 0.88, 0.21
0.875	0.25, 1, 10	200, 50, 50	1.00, 0.25, 0.25
1.0	0.25, 1	50, 50	0.31, 0.30
2.5	0.25, 1	50, 50	0.69, 0.71

3. Results

The results section is structured around the main research questions, i.e. what are the optimal USP settings to achieve high quality surfaces with high ablation efficiency and what is the cause of IIC.

3.1. Optimal laser ablation settings

To determine the optimal settings for USP laser ablation of stainless steel, the ablation efficiency and quality of processed surfaces are first examined. Two laser parameters, pulse duration and fluence, primarily determine the quality and efficiency of ultrashort laser ablation. Fig. 1 shows the overview of the bottom surface of the ablated hollows as the fluence of the laser beam increases from 0.25 to 2.5 J/cm^2 from left to right each row, from top to bottom of the figure the pulse duration changes from 0.25 ps, 1 ps to 10 ps. Note that the dramatic change in the morphology of ablated surface and thus the ablation quality largely depends on the applied laser fluence, considering the view area of the figures is equal.

The SEM micrographs clearly show that, in the *low* fluence regime ($< 0.75 \text{ J}/\text{cm}^2$), a large amount of micron-sized cones are formed on the ablated surface. The spatial density of the cones is determined by counting the amount of cones in each SEM micrograph which corresponds to a sample area of $400 \times 500 \mu\text{m}^2$. Cones along the border of the inspected area that are visually estimated to have 50% or more of their respective top-view area inside the field-of-view (FOV) are counted, while the cones with less than 50% of their respective area inside the FOV are disregarded. Accordingly, a sharp decrease in the density of cones is found with increasing laser fluence to around $0.625 \text{ J}/\text{cm}^2$, see Figs. 1 and 2a. According to the inspection it is observed that the size of the cones also decreases sharply, with increasing laser fluence. The influence of pulse duration on the spatial

density and size of the cones appears to be small. In the *high* fluence regime ($> 0.875 \text{ J}/\text{cm}^2$ for 0.25 and 1 ps pulses and $> 0.75 \text{ J}/\text{cm}^2$ for 10 ps pulse), fine cone-like protrusions (CLPs) are formed on the ablated surfaces. In this fluence regime USP laser ablation creates periodic trenches of tens of microns deep. These features start to occur even at lower fluences as the laser pulses get longer, e.g. at $0.625 \text{ J}/\text{cm}^2$ by 10 ps pulse ablation compared with $0.875 \text{ J}/\text{cm}^2$ by 1 ps pulse ablation and $1.0 \text{ J}/\text{cm}^2$ by 0.25 ps pulses ablation. These findings are more clearly summarized in Fig. 2b and c, where the spacing and height of the cones and CLP are presented as a function of laser fluence and pulse duration. It is noteworthy that the processing window with which a flat surface without the two surface structures (IIC and CLPs) can be ablated, is the largest ($0.625\text{--}0.875 \text{ J}/\text{cm}^2$) for the shortest pulse duration tested, and reduces with increasing pulse duration. Only very narrow or nearly no suitable processing window exists for flat ablation for 10 ps pulse laser; namely, either cones or CLPs occur on the ablated surface.

3.2. Ablation efficiency

Ablation efficiency is defined by the ablated volume of the hollows divided by the process time and average power required. Fig. 3 shows the efficiency of the ablation process as a function of laser fluence and pulse duration. Higher ablation efficiency was obtained in the *low* fluence regime and especially when using shorter pulse durations, e.g. 0.25 ps and 1 ps pulses. This is consistent with the findings of previous work [31,32]. The use of shorter pulse durations results in the pulse energy being deposited in less time. This leaves less time for energy dissipation, or heating of the surrounding material. An inferior surface quality was found for fluences under $0.625 \text{ J}/\text{cm}^2$ and above $0.75\text{--}1 \text{ J}/\text{cm}^2$ (depending on pulse length) due to generally unwanted surface effects. This shows that the pulse length as well as laser fluence is of significant

Fig. 1. SEM micrographs showing the bottom surface of ablated hollows 150 μm deep with three pulse durations: 0.25 ps (top row), 1 ps (middle row), and 10 ps (bottom row). Six fluence levels were used (from the left to the right column): 0.25, 0.5, 0.75, 0.875, 1 and $2.5 \text{ J}/\text{cm}^2$, respectively.

Fig. 2. (a) The number density of IIC as a function of laser beam fluence for three different pulse durations. The average spacing (b) and height (c) of CLP as a function of laser beam fluence at three pulse durations, with insets showing the cross sectional profile of CLPs created with laser fluence 0.625 and 0.825 J/cm^2 at three pulse durations.

impact on the ablation efficiency and obtained surface quality. As shorter pulse lasers are more costly, a balance between ablation efficiency, quality and cost may be made for different industrial applications.

Fig. 3. The ablation efficiency when creating a hollow in AISI 301 with a depth of approximately 150 μm . Tests were performed using a wide range of fluences and three different pulse durations. The shaded regions show non-feasible process settings because of unwanted surface effects (IIC in the gray region and CLP in the yellow region). Note the narrowed process window with increasing the pulse duration.

3.3. Formation of inclusion induced cones

The CLP formed in the *high* fluence regime had been well reported in the literature [21,27]. In this study we have additionally observed individual cones formed in the *low* fluence regime and tried to understand the formation mechanism of these cones and possible prevention.

The appearance of the cones found in the *low* fluence regime in this research, see Fig. 4, shows some similarity with the individual cones reported by a few researchers [17,25,26]. However, their formation mechanism is completely different. Previous hypotheses on the origin of individual cones suggested that they were formed by molten and solidified ablation debris particles or oxides [25]. It was stated that the debris absorbed the energy of subsequent laser pulses with less efficiency and would thus not be ablated. Beneath this cluster of debris, cones of bulk material would be shielded from ablation and remain intact for multiple scans.

Our investigation proposes that the cones found in this research are caused by inclusions in the material having a higher threshold of ablation energy than that of the surrounding bulk material. From Fig. 4a, it can be seen that a dark particle is always present at the summit of the cones. For this reason these particular cones are termed inclusion induced cones. When formed, the cones can be gradually ablated at longer ablation time or if the laser beam fluence is increased, see Fig. 4c.

3.3.1. Fluence dependence of IIC formation

To study the fluence dependence of IIC formation, the *low* fluence regime received in-depth investigation with small incremental steps of laser fluences from 0.0875 to 0.75 J/cm^2 at a pulse duration of 0.25 ps and repetition rate of 200 kHz. The detailed settings are shown in Table 3. The results show that the spatial density and average height of IIC decreases with increasing laser fluence. The energy of laser beam at the lowest laser fluence used was not high enough for efficient ablation as shown in Fig. 5a. The hollow made with this setting was significantly less deep compared to the hollows made at higher laser fluences, and the ablated surface was full of shallow cones. Such rough surfaces, comprising a high spatial density of short cones, might be useful for hydrophobic properties [28,33,34], although it is out of the scope of this work. Fig. 5b shows the surface made at the laser fluence of 0.125 J/cm^2 . The

Fig. 4. Inclusion-induced cones on the ablated surface of AISI 301: (a) top view and (b) cross sectional view with a double side arrow indicating the polarization of the incident laser beam; (c) cones being flattened as indicated by the arrows once the inclusion is fully ablated.

Table 3

Laser settings for the investigation of low-fluence ablation effects.

Fluence [J/cm^2]	Pulse duration, τ [ps]	Repetition rate [kHz]	P_{avg} [W]
0.0875	0.25	200	0.11
0.125	0.25	200	0.15
0.1875	0.25	200	0.22
0.25	0.25	200	0.29
0.5	0.25	200	0.58
0.75	0.25	200	0.84

depth of this hollow and the height of the cones are in line with the rest of the experiments. The maximum height of the cones is visually estimated over $40\ \mu\text{m}$. With increasing laser fluence, much less and shorter cones are formed as can be seen in Fig. 5c and d. Only a few IICs are formed by ablation at $0.5\ \text{J}/\text{cm}^2$ (Fig. 5e). At

Fig. 5. Fluence dependence of IIC in AISI 301: (a) $0.0875\ \text{J}/\text{cm}^2$, (b) $0.125\ \text{J}/\text{cm}^2$, (c) $0.185\ \text{J}/\text{cm}^2$, (d) $0.25\ \text{J}/\text{cm}^2$, (e) $0.5\ \text{J}/\text{cm}^2$, and (f) $0.75\ \text{J}/\text{cm}^2$ ($\tau = 0.25\ \text{ps}$, repetition rate $200\ \text{kHz}$).

Fig. 6. Comparison of bottom surfaces of ablated hollows $75\ \mu\text{m}$ deep in AISI 301 (top row) and AISI 316 VR (bottom row), respectively, processed at laser fluence of $0.185\ \text{J}/\text{cm}^2$ (left) and $0.25\ \text{J}/\text{cm}^2$ (right) ($\tau = 0.25\ \text{ps}$, repetition rate $200\ \text{kHz}$).

laser fluence of $0.75\ \text{J}/\text{cm}^2$, no IIC is formed but fully flat surface is achieved by USP laser ablation as seen in Fig. 5f.

3.3.2. Cause of IIC formation

Two stainless steels of similar composition but different contents of inclusions were used to verify if the presence of inclusions is the cause of IICs. Fig. 6 shows the resulting surfaces of AISI 301 and AISI 316 VR ablated at two different fluences ($0.185\ \text{J}/\text{cm}^2$ and $0.25\ \text{J}/\text{cm}^2$). Clearly, the stainless steel containing fewer inclusions (AISI 316 VR) has a lower concentration of IICs after ablation with the same laser settings. These findings indicate that the creation of IICs at the low fluence regime can indeed be attributed to the presence of inclusions in the base materials.

The IICs were further scrutinized with SEM characterization (Fig. 7). The dark spot in the summit of the cones indicate a spherical particle of

Fig. 7. SEM-picture of IIC created when ablating AISI 301 with 0.25 J/cm^2 . (1) EDS spot on bulk material. (2) EDS spot on the tip of an IIC.

Table 4

EDS measurement data for AISI 301 and AISI 316 VR steels.

Element	AISI 301 [wt.%]	AISI 316 VR [wt.%]
Si	1.3	0.7
Cr	17.6	17.3
Mn	1.5	2.1
Fe	72.6	62.7
Ni	7.0	14.0
Mo	0	3.2

Table 5

EDS measurement data of the inclusions in AISI 301.

Element	Content [wt.%]
O	36.1
Mg	11.8
Al	29.5
Si	5.5
Ca	1.6
Ti	0.9
Cr	4.7
Mn	7.6
Fe	2.2

a few micrometers in diameter. EDS analyses were made to reveal the difference in chemical composition between the tip particle and the cone bulk. Fig. 7 shows the spots where the EDS analysis was performed. Position 2 was placed on the dark spot in the summit of an IIC, and position 1 on a flat surface of bulk material and a third EDS analysis was performed on the cleaner AISI 316 VR. Comparison between the three EDS results, see Table 4 and Table 5, shows the different composition of the inclusion and the similarity between the two stainless steels. The EDS analysis of AISI 301 bulk, shows high iron content with chromium. The EDS analysis of AISI 316 VR shows a small but noticeable difference in terms of higher molybdenum-content compared to AISI 301, confirming the chemical composition specification from the supplier as listed in Table 1. A significantly different composition was found in the tip of the IIC (Table 5). The EDS analysis shows high aluminum and oxygen content, as well as a small amount of magnesium. Furthermore, iron and chromium are very low in weight percentage. This confirms that the dark spots in the tip of IIC consist of aluminum oxide (Al_2O_3), magnesium oxide (MgO) or a combination like spinel ($\text{MgO-Al}_2\text{O}_3$). These inclusions are a product of the steel making process and are often found as spherical particles of sizes ranging from approximately 2 to $8 \text{ }\mu\text{m}$ [35,36].

Fig. 8. Stop-motion SEM images showing the evolution of IICs (C0 and C1, C2, C3, C4) on ablated AISI 301 (laser fluence = 0.25 J/cm^2). Open arrows indicate inclusions being just emerged on the ablated surface.

Stop-motion SEM imaging was performed to investigate the evolution of a set of selected IICs, with the largest one C0 indicated by the solid arrow in Fig. 8. The figure is composed of multiple SEM micrographs of the same view field taken at the indicated depths of the hollow, D_p . This result shows that the size of the IICs increases at each ablation step as long as the inclusion in the summit remains. The moment when the inclusion of C0 is destroyed is clearly visible, at $D_p \sim 150 \mu\text{m}$. After that moment the cone starts to be flattened from the summit and ridge by normal ablation. Similar events occur for the other IIC's in the view. After the first ablation session, at the depth of $75 \mu\text{m}$, C1 has already been flattening. In contrast, C2 has just emerged. The entire process of this IIC, from emersion to flattening, is visible in the stop-motion SEM imaging. After two ablation sessions, at a depth of $125 \mu\text{m}$ the inclusion has been destroyed and flattening of the cone has started. C4 emerges at a depth of $150 \mu\text{m}$ and after ablating to a depth of $200 \mu\text{m}$, the IIC has grown in size and the inclusion remains, yet its size has decreased. The duration of this process from emersion until flattening is determined by the size of the inclusions, e.g. C2 vs. C3. The evolution of IICs over time confirms our hypothesis and is discussed in the next section.

4. Discussion

The hypothesis of IIC formation is proposed based on the fact that austenitic stainless steel contains ceramic inclusions that show different ablation behavior and ablation threshold required for efficient ablation. Different materials require different laser settings to achieve an efficient ablation process. Models have been created to describe and predict the ablation characteristics of certain materials in combination with certain laser settings [37–39]. An important parameter for describing ablation behavior is the ablation threshold, the minimal pulse energy that is required to initiate ablation. However, this parameter does not take into account other factors that influence ablation behavior like pulse duration or repetition rate. It is therefore an oversimplification of the process, but a useful starting point for understanding the ablation characteristics and is often used in research. An integral part of the IIC formation in alloys containing multiple phases is to have a higher ablation threshold than that for pure materials, as reported by multiple researchers [31,38,40–42]. Mannion et al., Jaeggi et al. and Le Harzic et al. [31,39,40] proposed a threshold fluence of $0.03\text{--}0.08 \text{ J/cm}^2$ for multiple pulses ablation of stainless steel with pulse durations from 0.5 to 10 ps. Single shot ablation requires 0.21 J/cm^2 as found by Mannion et al. [40].

Kim et al. [38] and Perrie et al. [41] investigated the ablation behavior of aluminum oxide and found an ablation threshold of $0.7\text{--}1.1 \text{ J/cm}^2$ after multiple pulses in the same spot with a pulse duration of 0.2 ps. Single shot ablation requires a fluence of over 3 J/cm^2 for sapphire (Al_2O_3) as reported by Ashkenasi et al. [43], which is close to the value of 3.27 J/cm^2 found by Wang et al. [44]. Notable is that these threshold values are significantly higher than those found for ablating (stainless) steels.

4.1. Ablation at sub-threshold fluence

The laser fluences, used in the low fluence regime ablation experiments where IIC were observed, are significantly lower than the threshold values of $0.7\text{--}3 \text{ J/cm}^2$ reported by the researchers above-mentioned, therefore no ablation of the oxide particles would be theoretically expected. However, the stop-motion SEM observation provides a clear proof that the inclusions in the summit of IICs are being ablated and decreasing in size. Furthermore, after multiple scans the inclusions are removed. This is a strong indication that, contrary to the expectation, ablation of the inclusions does occur, however at a much slower rate than the ablation in the surrounding

Fig. 9. SEM micrograph showing the detailed laser-induced periodic structures on the inclusion, cone body and bulk surface, and the difference in spatial frequency (laser fluence = 0.25 J/cm^2) with a double side arrow indicating the polarization of the incident laser beam.

bulk steel. As seen in Fig. 9, high magnification SEM observation shows an inclusion that is being flattened with typical LIPSS of relatively high spatial frequency in the summit of IIC. Both these findings indicate that the inclusion has been subjected to multiple scans of gentle ablation. Since the reflection of laser light on a surface is proportional to the incidence angle, laser ablation is more efficient in the center of spherical inclusions due to the fact that the incidence angle is closer to zero. A spherical particle is likely to be flattened by ablation of multiple scans.

The LIPSS that appear on the flattened inclusion are of a higher spatial frequency than the LIPSS found on the bulk steel, see Fig. 9. Based on visual inspection, the wavelength of the LIPSS on the inclusion are of HSFL-type and that of the bulk are LSFL. The appearance of HSFL on the inclusion at these fluences is consistent with the results of ablating Al_2O_3 using an 800 nm USP with pulse width of 130 fs, where near-threshold ablation led to the formation of LSFL and sub-threshold irradiation resulted in HFSL [45]. LFSL on sapphire at near-threshold fluence was also documented by Ashkenasi et al. [43]. The exact periodicity of the HSFL ripples may be also affected by the amount of pulses, as there is a linear dependence of the wavelength of LIPSS on the amount of pulses [11,46].

4.2. Decreased ablation threshold

Even though the fluence applied in the experiments is significantly below the reported threshold values for ceramic materials the inclusions are observed to be ablated. There are a number of explanations for this behavior, amongst which the incubation effect, heating effect and increase in surface roughness are worthy for a discussion.

The incubation effect [47,48] describes a decrease of the ablation threshold as a function of the amount of incident laser pulses. A lower amount of energy is required for ablation due to heating of the material. The ablation threshold after N pulses is described by

$$F_{th}(N) \cong F_{th}(0) \cdot N^{S-1} \quad (1)$$

where S is the incubation coefficient generally in the order of $0.7\text{--}0.9$ for metals and ceramics. For the repetition rate and pulse duration used in this research the incubation coefficient was found to be 0.7 according to Kim et al. [38]. Only considering this effect will cause a decrease of the threshold from 1.5 to 0.202 J/cm^2 in approximately 800 shots. This decreased threshold already allows ablation at the applied fluence of 0.25 J/cm^2 . For comparison, the inclusion on top of the IIC shown in

Fig. 9 has received over 1000 shots before it was ablated, and this delayed ablation leads to the formation of IICs.

The threshold values reported in literature were obtained on clean and polished material. The LIPSS structure, shown in Fig. 9, is a so-called sub-wavelength structure that can cause a significant increase of laser beam absorption through the ‘moth-eye effect’ [7]. Obviously increased absorption allows ablation at lower fluence levels.

Based on the experimental observations, the expected sub-threshold HSFL formation and the incubation effect, it is proposed that the inclusions are capable of ablation, at a slow rate, eventually leading to the removal of the inclusion. After the initiation of ablation, the complete removal of the inclusion depends on its size. Higher fluences trigger a faster decrease of the threshold fluence by incubation and induce a larger ablation rate of the inclusions, resulting in the creation of less and smaller IIC, e.g. at a fluence of 0.5 J/cm^2 almost no cones are formed. At a fluence of 0.75 J/cm^2 , even the largest inclusions do not remain long enough to create a cone and subsequently no cones are found on the surface.

5. Conclusion

Inclusion-induced cones are observed on the ablated surface of stainless steels with an ultrashort pulsed laser in the *low* fluence regime. The cones are caused by the inclusions in the stainless steels having a higher ablation threshold than the surrounding bulk material. The inclusions may remain for multiple ablation scans and shade the bulk material underneath from ablation to form individual cones. The inclusions are finally eroded as multiple pulses have caused the ablation threshold to decrease through incubation effects consisting of heating and altered optical characteristics. The removal of the inclusions means the end of the shading and the cones are flattened by the laser pulses from that moment on. The height of the cones depends on the size of the inclusion and the applied fluence. Switching to *medium* fluence regime is an effective method to prevent IIC. This increased fluence exceeds the ablation threshold of the bulk stainless steel as well as the inclusions leaving a smooth surface with undetectable HAZ. Cone-like protrusions form in the *high* fluence regime, especially for the tested laser settings with long pulse duration. The processing window that allows a flat surface, without the two surface structures (cones and CLPs), to be ablated is the largest for the shortest pulse duration tested, and reduces with increasing the pulse duration. The optimum pulse duration is found to be 0.25–1 ps, whereby smooth ablation surface can be achieved with a wider range of fluences compared to 10 ps pulse duration.

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