Justus Eichstädt

The spatial emergence of laser-induced periodic surface structures under lateral displacement irradiation conditions

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University of Twente

Book Author:

Justus Eichstädt
Berliner Straße 60
14712 Rathenow

Graduation Committee:

Prof. dr. G.P.M.R. Dewulf	University of Twente, Chairman, secretary
Prof. dr. ir. A.J. Huis in 't Veld	University of Twente, Promotor
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Paranymphs:

Dr. Johann Zbigniew Pierre Skolski
Dr. Paul Christiaan Verburg

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THE SPATIAL EMERGENCE OF
LASER-INDUCED PERIODIC SURFACE STRUCTURES
UNDER LATERAL DISPLACEMENT IRRADIATION CONDITIONS

PROEFSCHRIFT

ter verkrijging van
de graad van doctor aan de Universiteit Twente,
op gezag van de rector magnificus,
prof. dr. H. Brinksma,
volgens besluit van het College voor Promoties
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Prof. dr. ir. A.J. Huis in 't Veld

Preface

With this book, I would like to present my contribution to science. The subject concerns the determination of laser irradiation parameters, an issue that puzzled me already for quite some time. Hence, the content of this book is determined by the experience that I have made with the analysis of laser experiments. It was necessary to understand the laser irradiation and the spatial emergence. So, I simply calculated the irradiation, as presented in Chapter 2 and 4, and started to think about the thresholds, as presented in Chapter 3. This work ended up in this scientific contribution.

In the following, I will briefly describe the framework in which this work has been carried out. The research results were compiled at the Chair of Applied Laser Technology within the Faculty of Engineering Technology of the University of Twente in Enschede - The Netherlands in the years 2009 to 2014. In the first three years of this period a European research project has been carried out. This project had the acronym NanoClean. The aim of NanoClean was the mass production of permanent self-cleaning plastic parts for the automotive sector under industrial manufacturing conditions. Therefore, a previously lab-scale demonstrated laser based manufacturing technology was implemented and evaluated for an industrial application. NanoClean was performed in a collaboration of seven project partners: Maier S. Coop. (Spain), Fundación Gaiker (Spain), Lightmotif B.V. (The Netherlands), Demcon A.M.B.V. (The Netherlands), Universiteit Twente (The Netherlands), Centro Ricerche Fiat S.C.P.A. (Italy) and BASF SE (Germany).

The achieved research results have been exploited in other research projects. One was actually the master thesis of Wendy Masman-Bakkers. The thesis had the title: Application of LIPSS - The production of homogeneous areas of laser-induced periodic surface structures (LIPSS) on two biodegradable polymers and the influence of LIPSS on the alignment and morphology of human mesenchymal stem cells. This research was a collaboration between the Chair of Applied Laser Technology, the Department of Tissue Regeneration and the Department of Biomechanical Engineering.

A third project was related to the study of the tribological properties of LIPSS for precision positioning applications. This research was carried out by the Chair of Applied Laser Technology. The friction measurements have been performed at the Laboratory for Surface Technology and Tribology by Justus Eichstädt. Furthermore, the results were applied in the national M2i research project: Texturing using ultra short laser pulses, abbreviated TULP, with the industrial partners ASML Holding N.V., Lightmotif B.V., FEI Netherlands B.V., Holst Centre - TNO, Philips Applied

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Technologies. As part of this, an application project was carried out, in which adhesion measurements have been performed at TNO. The writing of this thesis and a part of the analysis were finished in Rathenow.

Rathenow,
summer 2015,
Justus Eichstädt

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Chapter 1

Introduction

1.1 Motivation

This research was triggered by potential applications for different industrial markets, as for example automotive engineering, semiconductor equipment and biomedical engineering. Driven by customer needs, companies have an economical interest to improve existing products. These products can be complex technical systems, as passenger cars or precision machines. They consist of sub-systems and components, which can have multiple functions. If an improved function of a component has the potential to improve the overall product performance, it can justify a new component generation. To improve a function of an existing component, an adaptation design is required [1]. One approach to find a solution for a design is to identify the physical phenomenon, which determines this function [1]. With respect to the mentioned industrial markets, contact mechanical, tribological or biomechanical surface phenomena play an important role.

Surface phenomena are the subject of research in numerous fields of fundamental and applied science. One field where great progress has been achieved is contact mechanics. The contact at the solid-solid interface yields elastic and plastic [2–5] deformations, influenced by the surface roughness [6, 7] and forces [8–10]. Fuller & Tabor studied the influence of roughness on adhesion between elastic solids and found a higher sensitivity for materials with high elastic modulus, e.g. adhesion has a low value at $R_q = 1\text{ }\mu\text{m}$ for rubber and at $R_q = 5\text{ nm}$ for TiC [11]. Besides surfaces of technical products, also natural surfaces have been investigated. One example is the analysis of plants to understand the interaction of leaves with water [12, 13]. Plants have developed hydrophobic surface properties, to keep themselves clean and thus to receive more light. In general, wettability is determined by the chemo-physical [14] and topographical [15, 16] properties of the surface. In particular, hydrophobicity can be enhanced with structures having a micro- and nanometre scale [17].

The growing understanding of surface phenomena contributes to the improvement of surface functions of industrial components. This knowledge influences the design of the surface and the selection of materials. Generally, results of the design process

are summarised in the component documentation. This includes technical drawings, which contain information about geometries, dimensions, tolerances and materials of the components. Derived from drawings, the manufacturing preparation generates a process sheet, to produce the component [1]. The process sheet contains manufacturing technologies and their order in the manufacturing chain. To implement the improved design for an existing component, the chain needs to be adapted for an additional structuring process. This requires to select a manufacturing technology and adding additional manufacturing steps.

The specifications in drawings define requirements on manufacturing technologies. Criteria for the selection of a manufacturing technology are, for example, the machineable dimensions, shapes, materials, accuracies and the economic efficiency [1]. In the case of the above-mentioned phenomena and the resultant specifications, it is required to structure surfaces on a micrometre and even nanometre scale. Next to this main requirement, for the implementation of a manufacturing technology in an existing chain, also other component specifications, product functions and manufacturing steps need to be considered, to avoid a negative influence on the final product.

1.2 Field of research

Over time, a large variety of manufacturing technologies have been developed [18]. Surface structures can be created at different locations in the manufacturing chain and with different manufacturing technologies. The standard DIN 8580 classifies manufacturing methods generally, such as forming and separation [1]. The methods are divided into manufacturing groups, oriented on production steps from a blank to a product. The manufacturing methods can be associated with different manufacturing technologies. An example for the main group primary forming is micro moulding [19], for joining cold-welding [20] and for coating physical vapour deposition [21].

Laser technologies are applied for different manufacturing methods and hence can be found in different main groups, as joining and separation. On this general level, a nomenclature for using the laser beam as a tool for machining is laser beam machining [22], abbreviated LBM [23]. Major advantages compared to other manufacturing technologies are seen as forceless, contactless, localised heat input and precise machining [22, 24]. A recent state of the art description for laser beam machining can be found in reference [25].

To generate three dimensional structures, usually material is removed from a pre-treated surface. This is often a later step in the production of a product, where shape and dimensions are given. The group removal belongs to the main group separation and is described in more detail in the standard DIN 8590 [1, 23]. This group distinguishes thermal, chemical and electrochemical removal mechanisms. The thermal removal is, among others, associated with electro-discharge machining, abbreviated EDM. Laser beam removal falls, next to electron and ion beams, into the subgroup thermal removal by beams and is also used as nomenclature for structuring surfaces in literature [26].

In literature laser technologies are differentiated into a macro and micro regime.

This classification separates systems as well as processes. An example of a process related to micro machining is surface structuring [27] and an example of macro machining is macroscopic welding [28]. This classification has already been used in the year 1965 [29] and later on established on a broad basis [30]. Recently also laser nano machining [31] is added. An overview of laser technology for manufacturing can be found in [32, 23].

This research deals with manufacturing technologies, in particular laser technologies, to structure surfaces. With respect to the structure dimensions, laser surface structuring belongs to laser micro and nano machining. This research is required on a research level to investigate surface phenomena and on a production level to manufacture industrial components. Advantages of laser surface structuring are the possibilities to machine different materials, under environmental conditions and without tool wear [33]. Next to laser surface structuring, laser surface texturing [34] and laser surface engineering [35], can be found in literature as nomenclature.

1.3 System developments

1.3.1 Beam sources

After selecting a manufacturing technology, it is important to choose a system. The beam source is the key component within a laser system. With the theory about the stimulated emission of radiation [36] Einstein laid down the fundament for the development of a new kind of radiation source. Finally, it was Maiman who reported in August 1960 with a short note in Nature [37] and one year later in a more elaborated form [38, 39], about the technical realisation of light amplification by stimulated emission of radiation, abbreviated LASER. The three main elements of the first laser were a ruby crystal, as active material, with silver coated end faces, forming the resonator, and a flash lamp, to optically excite the population inversion [37].

Over the years, a number of different types of laser sources have been developed. They can be classified by different aspects: the energy levels, as three [37] and four [40], the active materials, as Nd:YAG [41] and Yb:YAG [42], the host material, as Nd:CAWO [43] and Nd:YAG [41], the material geometry, as rod [39], slab [44], disk [45] or fibre [46], the aggregate state, as solid [37], fluid [47] or gas [48–50], the pumping mechanism, as optically [37] and electrically [51], and the mode of operation, as continuous [48, 52] or pulsed [37]. A major part of the developed sources has been applied for machining. A selection of milestones, in the development of laser technologies for surface structuring, is presented in Table 1.1.

The radiation emitted by different gain media differs in central wavelength λ , spectral linewidth $\Delta\lambda$, wave form and power P . For laser surface structuring, laser sources were of interest, which have a dedicated laser wavelength or pulse duration τ . Both parameters influence the surface structuring process, e.g. localisation of energy and dimensions of structures. A reduced wavelength can either be directly achieved with the selection of a specific active material, as with Excimer lasers, or indirectly by a subsequent conversion of the optical frequency, as with the second harmonic

Table 1.1: Selection of milestones, in the development of laser technologies for surface structuring.

Year	Milestone	Ref.
1905	Light quantum hypothesis	[62]
1916	Theory about the stimulated emission	[36]
1928	Experimental evidence for the stimulated emission	[63]
1960	Technical realisation of the first laser source	[37–39]
1964	Demonstration of the Nd:YAG laser	[41]
1970	Demonstration of the Excimer laser	[64, 65]
1986	Demonstration of the Ti:S laser	[66]

generation of, for example, Nd:YAG lasers. The pulse duration can be reduced by different methods, as gain switching [53], quality switching [54], cavity dumping [55] and mode locking [56]. Over time, the pulse duration has been gradually decreased down to the picosecond [57, 58] and femtosecond [59–61] regime.

However, the reduction of τ is coupled with an increased intensity I , for otherwise constant parameters. Therefore, short pulses are commonly created at low pulse energies E_p and afterwards amplified for the purpose of material processing. Reports on pulse amplification can already be found in 1964 [67]. But this topic gained interest with the development of femtosecond pulses [68, 69], where a breakthrough is connected to the chirped pulse amplification [70], as applied for a regenerative Ti:S amplifier [71–73]. Nevertheless, sources with pulse durations in the picosecond and femtosecond regime were at that time complex systems, that is why their application in industry was very limited. Nowadays, compact and reliable sources, with a high average power P_a and pulse frequency f_r , have been developed [74, 75] and a growing acceptance by industry is observed.

1.3.2 System integration

Shortly after the invention of the laser, a potential was seen for machining materials. Although, inducing a damage on a solid surface basically needs not more than the laser radiation itself [76], one of the first experimental configurations consisted of a laser source and a focusing lens [77, 78]. Today, the technical system as a whole used for machining materials with laser radiation is referred to as laser system [79]. One of the first industrial laser system configurations consisted of a laser head on top of a light microscope [29]. Such a device was used for micro machining applications and allowed next to focusing a direct quality control of small features. Another step in the past, related to system integration, was the application of a motion system, to position the work piece relative to the beam [27, 24].

A general description of the technical construction of a recent laser system can be found in standard DIN EN ISO 11145:2001 [79]. According to this standard, a laser system consists of at least one laser arrangement and different systems for position-

ing, measurement and control. The laser arrangement contains the laser device and elements for beam guidance and beam forming. The laser device consists of a laser head unit and supply unit, which are often physically separated [22]. For laser surface structuring two methods have been developed, if the full aperture of a laser beam is focused it is referred to as direct-writing and if a part of the beam is blocked by an aperture as mask-projection. Whereas mask projection requires higher energy and is fast for fixed structure designs, direct writing allows a flexible structure design but requires in general longer machining time.

Actual trends related to laser systems for surface structuring focus on flexibility, precision and automation [80–83]. With respect to direct writing systems, flexibility is achieved with an increased level of system integration, meaning that one laser system can have multiple laser sources, frequency conversions, motion systems and optic systems. Precision is on the one hand achieved by granite gantry mechanics and combined linear stage, galvanometer and trepanning axis systems and on the other hand by short pulse and wavelength sources as well as beam expanded telecentric F-theta lens focusing. Automation is reached by an electronic and software architecture, which allows to control the entire laser system by one interface. Another aspect which is considered in the development of these systems is the manufacturing implementation, such as with direct CAD-CAM coupling and integrated quality control systems.

1.4 Process investigations

1.4.1 Observed phenomena

After the development of the laser, experiments were performed in which radiation has been focused into the bulk [84] or onto the surfaces [85] of materials, to study the laser-material interaction. In these studies transient and permanent laser induced phenomena have been observed. Transient phenomena are for example optical second harmonics [84], two-photon transitions [86], emission of charged particles [87, 77], a luminous plume [78, 88] and plasma absorption [77, 88]. Permanent phenomena, remaining after irradiation on the surface or in a subsurface region, are for example material ablation [89, 90], a heat affected zone [22], a shock affected zone [91], material redeposition [88, 90] and periodic surface structures [92].

Already in 1965 Vogel and Backlund suggested to apply microscopy techniques to study laser-induced surface modifications [93]. Within one irradiated location different types of irreversible surface modifications have been observed. Interestingly, the modifications were present in concentric zones, using a laser with a Gaussian shaped intensity distribution. The concentric arrangement indicated a dependence of the spatial emergence on the distribution of optical energy. After the first observations of laser-induced phenomena and the identification of potential laser applications, the development and application of laser sources were limited by the damage of optical components [94]. The surface of optical components was found to be particularly sensitive to destruction.

Figure 1.1: Classification of laser-induced surface modifications in a fluence modification diagram, based on reference [96, 97].

In this context, Liu suggested a method to determine the beam radius in the sample plane [95]. This method assumes that modifications emerge if the single pulse peak fluence ϕ_0 is above a defined threshold ϕ_{th} specific for each type of modification. Later on, the observed single pulse modifications were classified by the fluence thresholds above which they occurred and summarised in a fluence modification diagram [96, 97]. An example of such a diagram is shown in Figure 1.1. The modification with the highest threshold fluence is observed in the centre of the irradiated location, modifications with thresholds below are found to emerge in radially separated zones.

Furthermore, evidence has been found that single pulse irradiation, so called 1-on-1, and multi pulse irradiation, so called N_p -on-1, provide different damage thresholds [96]. These characteristics were found to be related to the incubation properties of the

material. For metals the damage threshold $\phi_{th}(N_p)$ for multi pulse irradiation has been found to decrease with increasing number of pulses N_p per location [96]. Musal suggested a thermo-mechanical mechanism and calculated a transient stress-strain response of single-crystal metal surfaces upon pulsed laser irradiation in analogy to mechanical-fatigue [98].

These thermo-mechanical considerations were extended and empirical relations were proposed for the first time [99, 100]. In order to investigate this cumulative laser interaction, Jee [96] analysed an accumulation curve, which revealed a specific empirical description. This relation was found to be valid for different materials, such as metals [101], semiconductors [102], polymers [103] and ceramics [104, 105]. Another form to describe incubation was presented by Rosenfeld et al. [106] and Ashkenasi et al. [107]. But this relation was deducted for transparent materials, involving a specific incubation mechanism, which might not be applicable to opaque materials.

1.4.2 Periodic surface structures

Laser-induced periodic surface structures, abbreviated LIPSS or frequently called ripples, are wavy surface alterations observed within the location irradiated by the laser beam. Contrary to multiple laser beam interference patterns [108], LIPSS can be produced with single beam illumination. LIPSS have been observed on different materials, e.g. metals [109–112], semiconductor [92, 76, 113–115], dielectrics [116–119] and polymers [120, 121, 103] and under different irradiation conditions. Usually, these structures have a unidirectional appearance if produced with linearly $\vec{p}$ or $\vec{s}$ polarised light. However, other polarisation states alter their orientation or appearance [122–126]. Because of their generally unidirectional appearance, LIPSS are usually described by their periodicity Λ and orientation Θ .

Different types of LIPSS have been observed. LIPSS with a spatial period in the order of the laser wavelength ($\Lambda \approx \lambda$) are usually referred to as low spatial frequency LIPSS (LSFL) and those with significant smaller period ($\Lambda \ll \lambda$) as high spatial frequency LIPSS (HSFL). Whereas LSFL have been studied since 1965 with continuous wave lasers [127] and different pulse durations [128, 129, 117], the HSFL type is more recently observed in the context of picosecond and femtosecond pulse irradiation [117, 102]. For LSFL, Λ has been found to vary with the wavelength [121, 92, 76], the angle of incidence θ [76, 114] and number of pulses [130]. If linearly polarised light is applied, the orientation of LSFL is usually perpendicular to the polarisation direction, contrary to HSFL which can also be parallel.

The first explanation, given by Birnbaum [92], is that LSFL originate from a diffraction effect of a focused laser beam. Emmony suggested that this ripple phenomenon is related to the interference of the laser beam with a surface scattered wave [76]. Hereafter, mathematical models have been developed to improve the explanation, Temple et al. [116], Guosheng [131], Emelyanov [132] and Sipe [133]. The model of Sipe et al. is often used for explaining the occurrence of LSFL and can be identified as the commonly accepted model. It calculates the inhomogeneous energy absorption based on an electromagnetic wave approach. The model considers optical material parameters, as the refractive index n and the extinction coefficient k , surface

roughness and laser parameters, λ , θ and $\vec{p}$ or $\vec{s}$, to calculate Λ and Θ of structures. This model was validated with structures created by nanosecond pulsed lasers and the analysis of diffraction patterns [114].

The HSFL type of structures cannot be explained by the existing theories and their origin is still under debate in literature. Proposed explanations follow either an electromagnetic approach [115, 134, 135] or self-organisation [118, 136]. Next to HSFL, two other phenomena have been observed in the context of femtosecond experiments, a decrease and increase of LSFL periodicity [130]. LSFL with an increased period, usually 2-3 times the wavelength [130], are less discussed in literature and also referred to as grooves [137]. For the decrease on silicon three different reasons have been proposed, the carrier density in the conduction band [138], grating-assisted surface plasmon laser coupling [139] and the variation of local surface angle with respect to the incident laser beam at the ablation crater [140].

1.4.3 Structuring of surface areas

The research about structuring of surface areas was often driven by applications. In order to structure areas lateral displacements are required, which apply dynamic irradiation conditions, abbreviated DIC. The contrary is referred to as static irradiation conditions, abbreviated SIC. Nowadays, surface structures, created with a major lateral displacement during irradiation, have been established in industrial manufacturing. One example is the structuring of ultra-high density magnetic disk drives [141–144]. Here rims of expanded material are manufactured. In this example, the lateral dimensions of the features are in the range of micrometres and the distance between the features is larger than the beam diameter.

Early proposals for applications using minor lateral displacements during irradiation were the micro machining of thin metal coatings [27] and integrated circuits [24]. In these examples, a line of material was removed, with a velocity v below a certain limit to create an overlap φ of subsequent pulses. In the nineties the ablation of complex geometries has gained more interest [145, 26]. Often, the relation between φ and geometrical parameters, as ablation depth d and surface roughness, has been investigated [146, 147]. In these studies it has been found experimentally, that a pulse overlap of 0.3-0.5 is suitable for the removal of a plane layer of a material. For a ceramic material and with a laser of $\lambda = 355$ nm and $\tau = 15$ ns, a surface roughness in the order of $R_a \leq 0.1$ μm has been achieved [146].

In 2000 the influence of the overlap on the ablation depth has been investigated theoretically by Hellrung [148]. The surface geometry has been obtained by a summation of single laser spots approximated by spherical segments. In that way Hellrung found a relation between overlap and the ablated depth per layer. Later on, a predictive model based on an approximation of ablated segments by the Gaussian function has been developed [149]. Davis, found an overlap of 0.75 as optimum to achieve a smooth surface finish. Examples of laser ablation implemented in manufacturing were the fabrication of stamping tools [81], engine blocks [150] and moulding tools [147].

In 1982, Fauchet and co-workers reported an experiment in which they extended the spatial emergence of LIPSS by lateral displacements [129, 131, 151]. They found,

that a certain pulse overlap between successive pulses is required, to extend structures in one direction. Recently, this topic gained interest, because potential prospects are seen in the field of surface functionalisation, such as tribology [152, 153], optics [154, 155], wettability [156, 157] and biomechanics [158]. In the frame of this research, also other type of LIPSS, as HSFL, have been applied and different irradiation conditions have been studied.

The values for the irradiation parameters were determined experimentally, usually direct under DIC, and often no specific approach was mentioned. Empirically, the laser power is selected such that a peak fluence around the ablation threshold and the displacement velocity such that a pulse overlap for a homogeneous structure is obtained [152, 153, 155, 159]. For irradiation conditions where the displacement applied was too small, an "over-hatch" effect has been observed [159], resulting in the disappearance of LIPSS. The influence of the number of irradiation repetitions N_r has not been investigated.

1.5 Scope of research

The feasibility to produce LIPSS and the potential to influence surface functionality have been shown in research. But so far, the implementation as an industrial manufacturing process is limited. Potential applications have a variety of structure dimensions, product geometries and engineering materials, requiring a particular type of LIPSS or lateral distribution. Whereas industrial laser sources and systems are available, knowledge about the application of this process is lacking.

The machining of LIPSS requires a set of irradiation parameters. Although the relation between the formation mechanism and parameters, such as λ and θ , have been studied intensively, the relation of parameters, such as P_a , v and N_r , to the spatial emergence was of less interest. However, the latter parameters are specific for a type of LIPSS, spatial distribution and laser-material combination. That is why different applications require different values, which need to be determined and if necessary optimised with respect to machining time, spatial distribution and spatial properties. Therefore, the aim of this research is to investigate the relation between certain irradiation parameters and the spatial emergence of LIPSS.

One approach of this research could be the purely theoretical simulation of the laser-material interaction process to obtain irradiation parameters. But, it is questionable if the results obtained in this way would support the industrial application. This would require the availability of accurate input data for engineering materials and a profound understanding of the formation of all types of LIPSS. The microscopic analysis of laser-induced surface modifications provides an experimental access to LIPSS and is usually available under industrial manufacturing conditions. This access and the knowledge about incubation form the basis for a phenomenological approach in this thesis.

The course of this thesis is divided in three main parts, theory, experiments and applications. The first part is divided in three chapters and presents theoretical considerations, which have been developed prior to the experiments. The second part

is dedicated to the experimental investigation of the spatial emergence of LIPSS. The third part, evaluates the applicability of the developed knowledge for different cases and presents application results. The thesis finishes with a summary, which contains the main conclusions and an outlook into further research directions.

Chapter 2

Irradiation model

2.1 Introduction

The knowledge about incubation has been obtained under SIC. The application of LIPSS requires additional conditions, such as 1d- and 2d-DIC. Without a detailed analysis of the differences between these irradiation conditions, the understanding of the spatial emergence of LIPSS remains difficult. Although, the irradiation of extended areas is similar for ablation and LIPSS, a comprehensive description of the irradiation process cannot be found in literature. Therefore, this chapter is dedicated to the theoretical analysis of the irradiation process. This analysis is performed in the framework of a model, in which irradiation parameters are described and irradiation conditions are investigated. The first section concerns the assumptions, on which the model is based. The second is dedicated to the numerical implementation of the irradiation model. The last describes the output of the model, using numerical example simulations.

2.2 Physical assumptions

2.2.1 Optical assumptions

For the preparation of this section, various sources, that provide general knowledge about optics and laser technology, were used [160–164]. The irradiation model is presented in a Cartesian coordinate system for a three-dimensional space. The axes are referred to as x -, y - and z -axis. Within this frame, locations are denoted by $\vec{\psi}$. The origin is located at $\vec{\psi} = [x = 0, y = 0, z = 0]^T$. The z -axis is defined as the optical axis and light is assumed to propagate in positive direction. The irradiation model analyses the irradiation of a certain x - y plane in the path of light propagation. This plane is referred to as the irradiation plane and, in this chapter, no machining related laser-material interaction is considered. In this chapter the plane at $z = 0$ is analysed. Since this model is supposed to analyse the irradiation which arrives in the

irradiation plane, where later the sample surface will be placed, the medium in which light propagates from the source to this plane is assumed to be air. The length of the path of light between the source and irradiation plane is assumed to be so small that air can be considered as a non-dissipative medium.

The propagation of light can be mathematically described by Maxwell's fundamental equations of electrodynamics, if light is considered as an electromagnetic field. According to the Maxwell theory for electricity and magnetism, the electromagnetic field consists of two mutually coupled fields, the electric field $\vec{E}$ and magnetic field $\vec{H}$. Under certain conditions, the propagation of this field can be described by a second-order linear partial differential equation, the wave equation, which shows that this field oscillates transversal to its direction of propagation. The energy flux density in propagation direction is given by the Poynting vector

$$\vec{S} = \vec{E} \times \vec{H}. \quad (2.1)$$

For the given paramagnetic medium, the magnetic part of the wave has a very small effect on the propagation of light and has a very small contribution to the transfer of energy. The electric field strength is assumed to be so small, that a non-linear response of the propagation medium air, such as an optical breakdown, does not occur. The optical frequencies at which the field oscillates are so high that optical detectors determine values, which are averaged over a few optical cycles T_c . These values correspond to the standardised irradiance

$$I = |\vec{S}|_{T_c}, \quad (2.2)$$

which is the time averaged absolute value of the Poynting vector. In optics this quantity is also referred to as the non-standard expression intensity, which is used in the following. Models, analysing the periodicity and orientation of LIPSS, consider the vector properties of such a field [133]. The irradiation model assumes that the periodicity and orientation of a certain type of LIPSS is predominantly determined by the wavelength and polarisation of a given laser system. Whereas, for otherwise suitable parameters, the ability to interact with a material is assumed to be determined by the intensity. The polarisation state of the field is assumed to be linear.

In order to determine the spatial intensity distribution in more detail, the wave equation can be solved. Analytical solutions of this equation are the plane and spherical wave, both are important theoretical extreme cases, with respect to angular and spatial confinement, but far from experimental reality. To come up with an appropriate solution for real laser beams, it is required to define simplifications and boundary conditions, as given in the form of the Helmholtz equation and its slowly varying envelope approximation, also referred to as the paraxial Helmholtz equation.

A solution of the paraxial Helmholtz equation is the Gaussian beam, which comes, with its propagation direction and transversal limitation, close to real laser beams. In this case the intensity distribution follows in transversal direction a circular symmetric Gaussian function. The peak intensity I_0 is located on the optical axis at $x = y = 0$. In axial direction the course of the beam is represented by a caustic with increasing beam width, in positive and negative direction, from the centre of the caustic.

The laser beam is assumed to be focused on the irradiation plane by an optical system, which is assumed to be located in the centre of the unfocused beam waist and the caustic is assumed to have a small divergence. The optical system is assumed to have a non-limiting and low numerical aperture, with a large focal length f_L , so that the wavefront normal vectors form small angles with the optical axis, as paraxial rays. The propagation characteristics and the field distribution of the Gaussian beam are maintained also after transformation through the optical system.

The angle of incidence is assumed to be normal $\theta = 0^\circ$, so that the focal plane is parallel to the z-axis. The centre of the focused beam waist is coincident with the origin and the distance ω from the optical axis in transversal direction, where the amplitude reaches a values of $1/e^2$, is defined as the focused beam radius. The specific beam radius in the focal plane at $z = 0$ is denoted with ω_0 .

The paraxial Helmholtz equation describes a stationary situation. With respect to the temporal characteristics, the laser is assumed to be pulsed with a Gaussian envelope. The intensity distribution in the plane of focus at $z = 0$, with a spatial and temporal Gaussian shaped pulse form, can then be written as

$$I(x, y, t) = I_0 \exp\left(-2\left(\frac{x^2 + y^2}{\omega_0^2}\right)\right) \exp\left(-\frac{t^2}{\tau^2}\right), \quad (2.3)$$

where τ is the $1/e$ pulse duration and t is time. The irradiation model has the aim to analyse different irradiation conditions and thus to support the understanding of the spatial emergence of LIPSS. In this research, the spatial emergence is analysed by microscopy after irradiation. Intra-pulse effects, with time scales below the pulse duration, are not analysed. Therefore, the stationary situation, in which an individual pulse has arrived instantaneously and has been fully transmitted to the irradiation plane is assumed. This irradiation situation can be described by the individual fluence ϕ , which can be determined by integrating the intensity over time

$$\phi(x, y) = \int_{-\infty}^{\infty} I(x, y, t) dt, \quad (2.4)$$

if a single pulse is considered. Inserting Equation 2.3 in Equation 2.4 gives

$$\phi(x, y) = I_0 \exp\left(-2\left(\frac{x^2 + y^2}{\omega_0^2}\right)\right) \int_{-\infty}^{\infty} \exp\left(-\frac{t^2}{\tau^2}\right) dt. \quad (2.5)$$

Changing the domain of integration and replacing the time derivative by $d\gamma = dt/\tau$ gives

$$\phi(x, y) = I_0 2 \exp\left(-2\left(\frac{x^2 + y^2}{\omega_0^2}\right)\right) \int_0^{\infty} \exp(-\gamma^2) \tau d\gamma. \quad (2.6)$$

To solve the integral a solution from [165] for the error function

$$\text{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x \exp^{-\gamma^2} d\gamma \quad (2.7)$$

is considered, for which the following applies $\lim_{x \rightarrow \infty} \text{erf}(x) = 1$, and gives

$$\phi(x, y) = I_0 \tau \sqrt{\pi} \exp \left(-2 \left(\frac{x^2 + y^2}{\omega_0^2} \right) \right). \quad (2.8)$$

Replacing the term before the natural exponential function by the individual peak fluence $\phi_0 = I_0 \tau \sqrt{\pi}$ gives

$$\phi(x, y) = \phi_0 \exp \left(-2 \left(\frac{x^2 + y^2}{\omega_0^2} \right) \right). \quad (2.9)$$

Whereas the fluence allows to distinguish different spatial observations, the total amount of energy within this space can be obtained by integrating the fluence over the transversal spatial dimensions x and y

$$E_p = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \phi(x, y) dx dy. \quad (2.10)$$

With the same approach as for the derivation of the fluence, E_p is given by

$$E_p = \phi_0 \omega_0^2 \pi \frac{1}{2}. \quad (2.11)$$

Usually, instead of an individual pulse, a certain number of pulses are released by the laser source. If the spatial emergence is analysed after a pulse train has been applied, the fluence has accumulated. The accumulated fluence Γ can be obtained by a summation of the individual fluence over n

$$\Gamma(x, y, n) = \sum_n \phi(x, y). \quad (2.12)$$

The pulses are applied at a given pulse frequency, for which the power is determined experimentally. In general, the power can be obtained by integrating the intensity over the transversal spatial dimensions x and y

$$P(t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} I(x, y, t) dx dy \quad (2.13)$$

and describes how much energy is delivered per time. The temporal course of the power can be described with two quantities, the peak power and the average power. If a single pulse is considered, the peak power $P_0 = I_0 \omega_0^2 \pi \frac{1}{2}$ can be obtained by substituting Equation 2.3 in Equation 2.13 and evaluating the integral at the peak for $t = 0$. The average power for the pulse period T_r can be obtained by

$$P_a = \frac{E_p}{T_r} = E_p f_r, \quad (2.14)$$

where $T_r = 1/f_r$. For pulses with a short duration, as in the picosecond regime, and a low repetition frequency, as in the kilohertz regime, the energy is confined in a short period of time and the peak and average power deviate significantly.

2.2.2 Kinematic assumptions

So far a focused laser beam with a Gaussian fluence distribution around $x = 0$ and $y = 0$ has been defined and the x - y plane at $z = 0$ within the propagation direction has been analysed. Now, a limited area within this x - y plane is considered as irradiation plane. This area is assumed to have a flat and rectangular geometry, with edges parallel to the x - and y -axis. The centre of the area is located in the origin of the Cartesian frame. The relative position of this area with respect to the beam is assumed to be constant over the irradiation time. The edge lengths l_x in x and l_y in y direction are assumed to be larger than $2\omega_0$.

These irradiation requirements demand relative transversal movements, which are assumed to be translational motions. The direction of the translations is assumed to be parallel to the x - or y -axis. The beam profile, angle of incidence and irradiation plane are assumed not to change due to the displacement. The relative velocity, along the x direction is denoted with v_x and along y with v_y . In practice a motion system has a finite mass, that is why the movement is composed of an acceleration envelope at the beginning, a period of constant velocity in the middle and deceleration envelope at the end of the movement. For this analysis here, the movement is assumed to be uniform with constant velocity for both directions.

The path coordinates of irradiated locations are denoted with $\vec{\psi}_d$. For a single movement along x direction the irradiated locations can be described by the following equation

$$\vec{\psi}_d(t) = [x_1 + v_x t, y_1]^T. \quad (2.15)$$

In order to reach the first irradiated location and to return to the original position from the last location after the last pulse has been applied, two positioning movements are required. These movements are neglected with respect to the total irradiation time. The irradiation time starts at the first irradiation location with the application of the first laser pulse, including the pulse period before, and ends with the last laser pulse. The irradiation time depend on the pulse period and number of pulses.

In this analysis, multiple locations need to be irradiated on a line and a single movement is assumed, which passes over several locations. In this case the displacement time needs to be larger than the pulse period and multiple laser pulses are applied during a single movement. This causes the energy deposited on the surface to be distributed over an obround area. With respect to the considered laser systems and velocities in this research, the displacement during a pulse of e.g. 10 ps duration at a velocity of 1 m/s is 0.01 nm. This displacement is small, compared to a beam radius $\omega_0 > 10 \mu\text{m}$, and causes a negligible spread for $v \ll 2\omega_0/\tau$.

In order to irradiate locations in x and y direction, a sequence of two dimensional translations is assumed. The sequence is assumed to alternate between x and y translations, starting with the x . The direction of the motion parallel to the x axis is assumed to be positive and to reverse after each positive y increment, so that a meander routine is obtained. An example of such a sequence of translations, from the start position $\vec{\psi}_d = [x = -1, y = -1]^T$ to the end position $\vec{\psi}_d = [x = 1, y = 1]^T$, is shown in Figure 2.1 and coordinates along the translation path can be obtained from

Figure 2.1: Schematic drawing of an example for a meander movement path, illustrating the kinematics applied within the irradiation model.

the following equation

$$\vec{\psi}_d(t) = \begin{cases} [x_1 + v_x(t - T_1), y_1]^T, & T_1 \leq t \leq T_2 \\ [x_2 - v_x(t - T_3), y_2]^T, & T_3 \leq t \leq T_4 \\ [x_3 + v_x(t - T_5), y_3]^T, & T_5 \leq t \leq T_6, \end{cases} \quad (2.16)$$

where T_1 to T_6 denotes the boundaries for the irradiation periods. With respect to the selected temporal conditions, this sequence of translations can be analysed as a sequence of lateral displacements

$$\vec{\psi}_d(n) = \begin{cases} [x_1 + x_0(n - N_1), y_1]^T, & N_1 \leq n \leq N_2 \\ [x_2 - x_0(n - N_3), y_2]^T, & N_3 \leq n \leq N_4 \\ [x_3 + x_0(n - N_5), y_3]^T, & N_5 \leq n \leq N_6, \end{cases} \quad (2.17)$$

where n denotes the number of the pulse, x_0 the absolute displacement increment in x direction between two successive pulses and N_1 to N_6 the boundaries for the number of pulses. The movements along x direction are assumed to cover multiple irradiation locations and the movements along y direction one, which requires to move an increment y_0 .

If there is no displacement during the irradiation and a train of pulses is applied statically to a specific location, then the individual fluence can be obtained if the starting location x_s and y_s for the static irradiation are considered in the exponent of Equation 2.9, which yields

$$\phi(x, y) = \phi_0 \exp \left(-2 \left(\frac{(x - x_s)^2 + (y - y_s)^2}{\omega_0^2} \right) \right). \quad (2.18)$$

Inserting Equation 2.18 in 2.12 gives the Gaussian accumulated fluence of a displaced train of pulses

$$\Gamma(x, y) = N_p \phi_0 \exp \left(-2 \left(\frac{(x - x_s)^2 + (y - y_s)^2}{\omega_0^2} \right) \right). \quad (2.19)$$

In order to consider multiple displacements along a path, the sequence of displacements $\vec{\psi}_d(n) = (x_n, y_n)$ needs to be considered in the exponent

$$\phi(x, y, n) = \phi_0 \exp \left(-2 \left(\frac{(x - x_n)^2 + (y - y_n)^2}{\omega_0^2} \right) \right). \quad (2.20)$$

Under these conditions the individual fluence per location depends on the number of the pulse and the accumulated fluence is given by

$$\Gamma(x, y, n) = \sum_n \phi_0 \exp \left(-2 \left(\frac{(x - x_n)^2 + (y - y_n)^2}{\omega_0^2} \right) \right), \quad (2.21)$$

as a summation of the individual fluence over each pulse contribution and depends on the displacement path $\vec{\psi}_d(n)$.

The irradiation model was developed to support the understanding of the spatial emergence of LIPSS. In this context, the model is applied to analyse the differences between SIC and DIC. In Subsection 2.2.1, the individual and accumulated fluence has been introduced as physical quantity, to analyse the irradiation. If ϕ would have been independent from n , it would have been sufficient to analyse only the spatial distribution of $\phi(x, y)$ and $\Gamma(x, y)$. Due to the displacement of laser pulses, has, in addition, the fluence accumulation process to be considered.

2.3 Numerical implementation

2.3.1 Input data declaration

The aim of the numerical computations is to compute the individual and accumulated fluence as well as the fluence accumulation process. The basic structure of the program for the simulation of the irradiation process is described in this section. In Chapters 3 and 4, extensions of individual parts of the program are described.

The program has been divided in three parts. The first part concerns the declaration of the input data. The second part builds the main part of the program and contains the main computation operations. The program ends with a result part in which, based on the simulation data, values for characterisation parameters are determined.

The input data are related to computation options, physical parameters, derived quantities and computation variables. The computation options applies to the type of simulation, as on-line or off-line simulation, the type of kinematic, as dimension or path, the graphical visualisation, as visualisation speed or screen options and the analysis options, as analysis location, boundary conditions and type of result.

After this first part, variables for physical input parameters are declared. The irradiation model is supposed to simulate the irradiation applied by a direct-writing laser system. Thus, the physical input parameters of such a laser system are considered as input for the simulation. These parameters are referred to as system parameters and listed in Table 2.1 Class I on page 24.

From those physical input parameters, other quantities can be derived. The first set of derived quantities can be obtained by using the pulse frequency to calculate the pulse energy

$$E_p = P_a T_r = \frac{P_a}{f_r} \quad (2.22)$$

and the absolute displacement increment in x direction

$$x_0 = v_x T_r = \frac{v_x}{f_r}. \quad (2.23)$$

In the following the kinematic parameters are described for the x direction. The nomenclature in the y direction is the same. Another parameter is the focused beam radius, where either an input from a theoretical estimate or experimental determination can be used. The focused beam radius can be obtained from the following equation

$$\omega_0 = \frac{\lambda}{\pi} \frac{f_l}{\tilde{\omega}_0} M^2, \quad (2.24)$$

where the unfocused beam radius in the centre of the beam waist is denoted with $\tilde{\omega}_0$ and the parameter M^2 denotes the beam quality factor. Another set of quantities can be obtained if the energy and the displacement are normalised by the focused beam area or diameter. Rewriting Equation 2.11 for the peak fluence gives

$$\phi_0 = \frac{2E_p}{\omega_0^2 \pi}. \quad (2.25)$$

If the absolute displacement increment is normalised by the focused beam diameter, the relative displacement increment $\delta_x = x_0 / (2\omega_0)$ is obtained. In practice often the pulse overlap

$$\varphi_x = \frac{\Delta x}{2\omega_0} = 1 - \frac{x_0}{2\omega_0} \quad (2.26)$$

is used, which is the normalised geometrical intersection Δx of subsequent pulses. In the simulation program φ is calculated with Equation 2.26 based on the quantities

Figure 2.2: Schematic drawing illustrating the intersection of displaced laser pulses in x direction. The nomenclature in the y direction is the same.

derived before. In practice the pulse overlap can also be derived from v , ω_0 and f_r with equation $\varphi = 1 - v / (f_r 2\omega_0)$, if Equation 2.23 is substituted in 2.26. The derived quantities are summarised in Table 2.1 Class II on page 24.

For the computation two additional quantities are required, the number of displacements N_x and the first displaced position $\vec{\psi}_d(1)$. Whereas other input parameters can be rational numbers, N_x is a derived quantity and can just be a non-negative natural number $N_x \in \mathbb{N}_1$. This requires to round the derived values for N_x and yields odd and even integers. In order to centre the displacements around the origin, the values for N_x are determined for the half-space and rounded afterwards. The number of displacements follows from the definitions illustrated in Figure 2.2 and reads

$$N_x = \frac{l_x - 2\omega_0}{2x_0}. \quad (2.27)$$

Equations 2.23 and 2.27 can be used to determine the starting position for the first irradiated location, which is given by

$$\vec{\psi}_d(1) = [x_s, y_s]^T = [-x_0 N_x, -y_0 N_y]^T. \quad (2.28)$$

The absolute displacement in x direction is denoted with x_s . In these simulations, the first irradiation location is defined to be in negative x and y direction.

The declaration of input data ends with the declaration of computation variables. This can be for example a matrix for the space domain, a vector for the number of pulses or a set of graphics required for the on-line visualisation process. The declaration of these variables runs before the main part of the program starts, but in this text those variables are introduced when the associated function is explained.

2.3.2 Iterative main functions

This subsection describes the main computation operations for the simulation of the fluence accumulation process. The numerical implementation of the irradiation process uses an iterative control structure with a counter-controlled loop type. The iterations apply n as counter for the number of the pulse, with N_p as boundary. At certain values of n , kinematic events need to be executed, which are the selection of the translation axis, the selection of the translation direction and the extrapolation of the next event locations. The numerical implementation of the kinematic events, uses an iterative control structure with an event-controlled loop type.

Equation 2.17 gives an example of a position vector for a meander path. For such a path, generally four pulse boundaries need to be determined, the boundary N_a for the first pulse of the area, N_b for the last pulse of a line, N_c for the last pulse of the area and N_d for the total number of pulses. This set of boundaries is defined once outside the loop and given by

$$N_a = 1 \quad (2.29)$$

$$N_b = (2N_x + 1) \quad (2.30)$$

$$N_c = (2N_x + 1)(2N_y + 1) \quad (2.31)$$

$$N_d = ((2N_x + 1)(2N_y + 1))N_r = N_p. \quad (2.32)$$

The boundary N_b and N_c need to be updated within the loop, if one of the four boundaries is reached. Based on these definitions, the actual position is calculated by

$$\vec{\psi}_d(n) = [x_n, y_n]^T = [x_{n-1} + x_0 \varsigma_x(n) \vartheta(n), y_{n-1} + y_0 \varsigma_y(n)]^T, \quad (2.33)$$

where $\varsigma_x \in \{0, 1\}$ and $\varsigma_y \in \{0, 1\}$ are variables which switch the translation axes and $\vartheta \in \{-1, +1\}$ is a variable which switches the translation direction. These parameters depend on n . In case of a meander sequence, the direction of the x translation has to be switched after each line in x direction.

The functions, required to compute the fluence accumulation process are given by Equations 2.20 and 2.21, as continuous physical functions, and build the basis for the main numerical functions of the program. In order to evaluate both functions numerically two decisions are required, one is related to the space and the other to the pulse domain. In order to compare different irradiation conditions, it is required to calculate the spatial distribution of the final accumulated fluence. This can be obtained if the individual fluence is determined for each pulse and the complete space domain. This computation can also be used to extract the individual fluence for a specific location, which allows to analyse the fluence accumulation process. The individual

fluence for the complete space domain is not required for further analysis, because the peak fluence and individual fluence distribution are constant during irradiation.

For the simulations, a discrete space domain $[\mathbf{x}, \mathbf{y}]$, which goes from $-l_x/2$ to $+l_x/2$ in x direction with the spatial resolution q , has to be generated. The variables $\mathbf{x}$ and $\mathbf{y}$ represent matrices and each of them consist of a grid of data points with the indices i and j . The number of elements to sample the space domain is denoted by e and determined by l and q of the space domain. These parameters have to yield a satisfactory resolution of the fluence and displacements and an optimal computation time. The two main functions, placed within the loop, are the spatially discretised forms of Equation 2.20, which gives

$$\phi(\mathbf{x}, \mathbf{y}, n) = \phi_0 \exp \left(-2 \left(\frac{(\mathbf{x} - x_n)^2 + (\mathbf{y} - y_n)^2}{\omega_0^2} \right) \right), \quad (2.34)$$

and of Equation 2.21, which gives

$$\Gamma(\mathbf{x}, \mathbf{y}) = \sum_{n=1}^{N_p} \phi(\mathbf{x}, \mathbf{y}, n). \quad (2.35)$$

This yields two numerical matrices, which are updated during the computation process and contain the individual and accumulated fluence for all locations on the grid. In order to obtain the individual fluence at a specific location for each pulse contribution, the matrix $\phi(\mathbf{x}, \mathbf{y}, n)$ is used to store fluence values in a numerical vector $\vec{\phi}$ for each n . This specific location is referred to as analysis location and is denoted with $\vec{\psi}_a$. The fluence values are determined by identifying the element $[i, j]$ for the location $\vec{\psi}_a$ in the space domain and the corresponding value in the fluence domain. The matrix $[\mathbf{x}, \mathbf{y}]$ may not contains the exact location $\vec{\psi}_a$. Therefore, the next position is determined, which comes closest to $\vec{\psi}_a$. The spatial difference depends on the spatial resolution and yields a difference in fluence. The iterative main functions are summarised in Table 2.1 Class III on page 24.

Alternatively, the accumulated fluence can be obtained by a separate summation. Therefore, the variable n could be divided in a component for number of displacements in the x direction, another for the y direction and for the number of irradiation repetitions. By writing the equations for the individual and accumulated fluence in this way, a separation of the different kinematical contributions is obtained. However, this approach does not yield the individual fluence contribution of the n 's pulse of the irradiation process. Another alternative, to compute the accumulated fluence, is to calculate the individual fluence once, outside the loop, for a smaller space domain. This fluence values can be added, inside the loop, to the displaced location of a larger matrix. For a simulation with a large space domain, this solution could result in an optimised computation time. However, for the purpose of this research, the presented solution yields a sufficient computation time, to analyse the irradiation process.

2.3.3 Output characterisation

At the end of the numerical simulation a matrix $\phi(\mathbf{x}, \mathbf{y})$ for the individual fluence of the last pulse, a vector $\vec{\phi}(n)$ for the individual fluence at a specific analysis location for all pulse contributions and a matrix $\Gamma(\mathbf{x}, \mathbf{y})$ for the final accumulated fluence is obtained. The main simulation results, which need to be characterised, are contained in $\vec{\phi}(n)$ and $\Gamma(\mathbf{x}, \mathbf{y})$. These data represent the output of the model and contain information about the applied irradiation.

In order to analyse these data, irradiation output parameters need to be defined which characterise the simulation results. The characterisation depends on the aim of the model, which is on the one hand to describe the differences between different irradiation conditions, see Section 2.4 of this chapter, and on the other hand to determine the applied irradiation in experiments, see Chapter 5.

In this research the simulation results have been characterised by typical statistical quantities. These are for example the extreme values individual peak

$$\phi_p = \max \left\{ \vec{\phi}(n) \right\}, \quad (2.36)$$

valley

$$\phi_v = \min \left\{ \vec{\phi}(n) \right\} \quad (2.37)$$

and peak-to-valley difference fluence

$$\phi_d = \phi_p - \phi_v \quad (2.38)$$

and accumulated peak

$$\Gamma_p = \max \left\{ \Gamma(\mathbf{x}, \mathbf{y}) \right\}, \quad (2.39)$$

valley

$$\Gamma_v = \min \left\{ \Gamma(\mathbf{x}, \mathbf{y}) \right\} \quad (2.40)$$

and peak-to-valley difference fluence

$$\Gamma_d = \Gamma_p - \Gamma_v. \quad (2.41)$$

Next to these quantities, which represent information about singular values within the pulse or space domain, the average fluence is a quantity which gives information about the weighting of the fluence distribution. This parameter can be determined for the individual fluence vector

$$\phi_a = \frac{1}{N_p} \left(\sum_{n=1}^{N_p} \vec{\phi}(n) \right) \quad (2.42)$$

the individual fluence matrix

$$\phi_a(\mathbf{x}, \mathbf{y}) = \frac{1}{N_p} \left(\sum_{n=1}^{N_p} \phi(\mathbf{x}, \mathbf{y}, n) \right) \quad (2.43)$$

and the accumulated fluence matrix

$$\Gamma_a = \frac{1}{e^2} \left(\sum_j \sum_i \Gamma(\mathbf{x}, \mathbf{y}) \right). \quad (2.44)$$

If the irradiated area is large, compared to the focused beam diameter, the number of pulses with a small contribution is also large. Therefore, a fluence vector $\vec{\phi}(\hat{n})$ for an effective number of pulse contributions $\hat{n}$ can be defined for which $\forall \phi \in [\phi \geq \phi_b]$. The fluence boundary ϕ_b , limits the number of laser pulses which contribute to a location. Also the accumulated fluence values depend on the location where they are determined. In order to obtain fluence values $\Gamma(\hat{\mathbf{x}}, \hat{\mathbf{y}})$ from a specific analysis area, the space domain can be limited $\forall x \in [-x_b \dots x_b]$ and $\forall y \in [-y_b \dots y_b]$, where x_b and y_b are half sized space domain boundaries for the analysis field.

Furthermore, the dependence of certain characterisation parameters on the lateral displacements can be analysed relatively. This yields, for example, the relative individual peak fluence

$$\tilde{\phi}_p = \frac{\phi_p}{\phi_0} \quad (2.45)$$

and relative accumulated peak fluence

$$\tilde{\Gamma}_p = \frac{\Gamma_p}{\phi_0 N_r}. \quad (2.46)$$

Two further characterisation parameters, specific for the accumulated fluence, are the peak of the accumulated fluence

$$\Gamma_0 = \frac{\Gamma_p}{N_r} \quad (2.47)$$

and the accumulated centre fluence

$$\Gamma_c = \Gamma_v + \frac{\Gamma_d}{2}. \quad (2.48)$$

The peak of the accumulated fluence represents the amount of accumulated fluence per repetition. For a very large displacement, this quantity is equal to the peak fluence. The accumulated centre fluence is usually applied to characterise the location of the plateau part of the accumulated fluence.

Another characteristic quantity for the fluence accumulation process is the number of pulses applied for the irradiation. It is important to note, that next to Equation 2.32 the number of pulses can also be derived from equation

$$N_p = \left(\left(\frac{l}{2\omega_0(1-\varphi)} - \frac{1}{(1-\varphi)} \right) + 1 \right)^2 N_r, \quad (2.49)$$

for a quadratic area of edge length l and an, for both directions, equal pulse overlap φ . The number of pulses per location, depends on the edge length, pulse overlap and

Table 2.1: Classification of the irradiation parameters.

Class	Description	Variables
I	Input - System parameters	$P_a, f_r, v_x, v_y, \lambda, f_l, \tilde{\omega}_0, M^2, l_x, l_y, N_r$
II	Input - Derived quantities	$E_p, x_0, y_0, \omega_0, \phi_0, \varphi_x, \varphi_y, N_x, N_y, x_s, y_s, N_a, N_b, N_c, N_d$
III	Functions - Iterative equations	$N_b, N_c, x_n, y_n, \boldsymbol{\phi}(\mathbf{x}, \mathbf{y}, n), \boldsymbol{\Gamma}(\mathbf{x}, \mathbf{y}), \vec{\phi}(n)$
IV	Output - Characterisation	$\phi_p, \phi_v, \phi_d, \Gamma_p, \Gamma_v, \Gamma_d, \phi_a, \Gamma_a, \tilde{\phi}, \tilde{\Gamma}, \Gamma_0, \Gamma_c, N_p, \hat{N}_p$

beam radius. As already mentioned for the effective individual fluence, if the size of the irradiated area is large, the majority of pulses have a small contribution to a specific location. Therefore an effective pulse number

$$\hat{N}_p = \left(\frac{2}{(1 - \varphi)} + 1 \right)^2 N_r \quad (2.50)$$

can be defined based on a considered area of length $l = 6\omega_0$. This definition limits the maximum displacement to $x_0 = 2\omega_0$, which represents a fluence boundary of $\phi_b = \phi_0/e^8$. Equation 2.50 shows, that for an homogeneous irradiated area, the effective number of pulses per repetition depend only on the pulse overlap. The characterisation parameters are summarised in Table 2.1 Class IV.

The classification of irradiation parameters, presented in Table 2.1, follows the description presented in this section. The irradiation model needs laser system parameters as physical input. For the given optical and kinematical assumptions and an equal displacement increment in both directions, the input for the main functions is determined by the three parameters ϕ_0 , φ and N_r . Whereas ϕ_0 and φ are derived quantities, N_r is a laser system parameter. The numerical simulation of the fluence accumulation process with displaced laser pulses of Gaussian distribution, yields a numerical fluence vector $\vec{\phi}(n)$ for the pulse domain and a numerical fluence matrix $\boldsymbol{\Gamma}(\mathbf{x}, \mathbf{y})$ for the space domain as output. This data can be characterised by standard statistical quantities, referred to as irradiation output parameters.

2.4 Fluence simulation results

2.4.1 Accumulation process

Within the fluence accumulation process the lateral displacement of Gaussian laser pulses is transferred into an application of an individual and accumulated fluence to each location. Under certain conditions the fluence accumulation process results in

Figure 2.3: Simulation results #1 to #3 of a SIC, 1d-DIC and 2d-DIC. The grey scale represents the accumulated fluence in J/cm^2 .

characteristic individual and accumulated fluence simulation profiles. In the following, some profiles are shown, which are relevant in practice.

These simulations are performed on an area of length $l_x = l_y = 100 \mu\text{m}$ and with a spatial resolution of $q = 0.1 \mu\text{m}$. The focused beam radius is selected with $\omega_0 = 10 \mu\text{m}$. Other variables, as P_a , f_r and v , are selected such that a corresponding ϕ_0 and φ is obtained. The fluence boundary was $\phi_b = \phi_0/e^8$ and the space boundary was $x_b = y_b = 2\omega_0$. The individual fluence has been determined at two different analysis locations, in the origin at $\vec{\psi}_a(1) = [x = 0, y = 0, z = 0]^T$ and a distance away from the origin at $\vec{\psi}_a(2) = [x = \omega_0/3, y = \omega_0/3, z = 0]^T$. The sequence of translations followed a meander path.

Figures 2.3a to 2.3f show the simulation results #1 to #3, of a SIC, see Figures 2.3a and 2.3d, 1d-DIC, see Figures 2.3b and 2.3e, and 2d-DIC, see Figures 2.3c and 2.3f. The top Figures (a-c) show the individual fluence vector $\vec{\phi}(n)$ for the complete pulse domain and the bottom Figures (d-f) show the accumulated fluence matrix $\Gamma(\mathbf{x}, \mathbf{y})$ for the complete space domain. Quantitative results for a selection of the irradiation input and output parameters are summarised in Table 2.2 on page 30. The results can differ from values obtained in an analytical way, due to rounding and

discretisation during the numerical simulations. This table also contains the results of the following simulations.

Under SIC the accumulated fluence shows a Gaussian distribution and under DIC it differs from this. In the examples, the accumulated fluence was found to increase from SIC to 1d-DIC and further to 2d-DIC. On the side face of the irradiated area, the accumulated fluence drops according to the tails of the Gaussian distribution and under DIC the accumulated fluence decreases towards the outer side of the irradiated area, because there are more pulses contributing to the centre than to the outer regions.

Under SIC, the individual fluence was found to be constant and different, for both analysis locations. The analysis location $\vec{\psi}_a(1)$ is coincident with the centre of the Gaussian distribution and therefore yields values and an average fluence equal to ϕ_0 . The analysis location $\vec{\psi}_a(2)$ was displaced with respect to the centre of the Gauss and therefore yields values smaller than ϕ_0 . The number of pulses was equal to the number of irradiation repetitions.

Under DIC, the individual fluence has been found to vary for both analysis locations. Each subsequent pulse applied a different fluence and several extremes have been observed. The variation was different under 1d- and 2d-DIC. Of particular interest is a maximum turning point, abbreviated MTP. Whereas under 1d-DIC, one MTP has been observed for each repetition, under 2d-DIC there have been several. The MTP is related to the approach of laser pulses to the analysis location. Since under 2d-DIC the pulses approach the analysis location line by line, each MTP increases until the analysis location is reached and decreases afterwards. The number of MTPs depends on the number of lines and the whole sequence repeats with N_r . The location of a MTP, in terms of pulses, depends on the trajectory of the path and the analysis location.

The maximum of the MTPs is equal to ϕ_p , which depends on the peak fluence and on the relative position of the analysis location to the centre of a Gaussian pulse in the translation path. For location $\vec{\psi}_a(1)$ under DIC $\phi_p = \phi_0$, which is the same compared to SIC, but $\phi_a < \phi_0$, which is different compared to SIC. The average fluence is also smaller under DIC compared to SIC, because N_p was increased and different from N_r . If both analysis locations are compared with each other, differences can be ascertained. The difference of ϕ_p and ϕ_a between both locations is smaller under DIC compared to SIC.

The SIC yield different irradiation results in comparison to DIC. Whereas, under SIC the individual fluence is constant and the accumulated fluence distribution Gaussian, under DIC the individual fluence is varying and the accumulated fluence distribution non-Gaussian. Under SIC, the irradiation input parameters provide comprehensible information about the output parameters. That means, ϕ_0 represents ϕ_p and ϕ_a of the individual fluence in the centre of a pulse and N_r is equal to N_p . This also yields a simple and comprehensible relation between the individual and accumulated fluence. This is not the case under DIC, where the parameter combinations (ϕ_0, ϕ_p) and (N_r, N_p) partially lose their meaning for a comprehensible description of the energy input. Depending on the purpose, other characterisation parameters are required to describe the energy input.

Figure 2.4: Simulation results #4 to #6 of 2d-DIC with different pulse overlaps. The grey scale represents the accumulated fluence in J/cm².

Especially the individual fluence develops, with the introduction of lateral displacements, into a complex function, where further research is required to develop an adequate description. However, without determining the exact applied irradiation, a comparison of the obtained irradiation results, based on the irradiation input parameters, is difficult in practice. Another difference between SIC and DIC, is the difference, in terms of individual and accumulated fluence, between certain locations. Under

DIC the average individual and individual peak fluence provide smaller differences for different locations than under SIC. This can also be observed for the accumulated fluence, if the difference is compared for the SIC and 2d-DIC.

Figures 2.4a to 2.4i show the simulation results #4 to #6 of 2d-DIC with $\varphi = -0.75$, see Figures 2.4a, 2.4d and 2.4g, $\varphi = 0.40$, see Figures 2.4b, 2.4e and 2.4h, and with $\varphi = 0.63$, see Figures 2.4c, 2.4f and 2.4i. The top figures (a-c) show the individual fluence vector $\vec{\phi}(n)$ for the complete pulse domain, the middle figures (d-f) show the accumulated fluence matrix $\mathbf{\Gamma}(\mathbf{x}, \mathbf{y})$ for the complete space domain and the bottom figures (g-i) show the accumulated fluence vector $\vec{\Gamma}(x)$, which is a numerical cross-section of $\mathbf{\Gamma}(\mathbf{x}, \mathbf{y})$ at $y = 0$ along the x direction.

Example #4 shows a displacement, which results in a spatial separation and a displacement independent accumulated peak fluence. Example #5 shows a displacement, characteristic for a more complex variation and a minor increase of accumulated fluence. Example #6 shows a displacement, characteristic for a more constant accumulated fluence in the centre of the irradiated area and a major increase of accumulated fluence. In these three examples the accumulated peak and peak-to-valley difference fluence varies for different values of φ and the individual fluence varies too.

The number of MTPs increases with increasing pulse overlap, because more lines are approaching the analysis location. But, the effective individual fluence was constant, in Example #4, because only one pulse contributes effectively, and varied, in the Examples #5 and #6. Therefore, in Example #4 $\hat{\phi}_a = 1.0 \text{ J/cm}^2$ and decreases in the other examples. In Example #4 the average individual fluence was different for both analysis locations. In the other examples, the differences are smaller.

The simulation results #4 to #6 are representative examples of DIC. In this work, three different pulse overlap regimes are distinguished low, medium and high overlap DIC, abbreviated LODIC, MODIC and HODIC. With decreasing pulse overlap, the DIC change at some point in an effective SIC, for the individual as well as the accumulated fluence. The effective SIC regime approximately corresponds to the LODIC regime. In the effective SIC regime, ϕ_0 represents ϕ_p and ϕ_a of the individual fluence in the centre of a displaced pulse and N_r is equal to $\hat{N}_p$. With different pulse overlaps, the differences in individual and accumulated fluence for different locations vary. Based on the three example simulations, only the differences can be obtained. Therefore, additional simulations are required for a series of pulse overlap values, see Subsection 2.4.2.

Figures 2.5a to 2.5i show the simulation result #7 of a 2d-HODIC with $\varphi = 0.95$. The top figures (a-c) show the individual fluence distribution $\phi(\mathbf{x}, \mathbf{y})$ of the last displaced laser pulse, the accumulated fluence $\mathbf{\Gamma}(\mathbf{x}, \mathbf{y})$ for the complete space domain and the average individual fluence $\phi_a(\mathbf{x}, \mathbf{y})$ for the complete space domain. The middle figures (d-f) show the individual fluence vector $\vec{\phi}(n)$ for the complete pulse domain at $\vec{\psi}_a(1)$ in linear scale, at $\vec{\psi}_a(1)$ in semi-logarithmic scale and at $\vec{\psi}_a(2)$ in linear scale. The bottom figures (g-i) show the individual fluence vector $\vec{\phi}(\hat{n})$ for the effective pulse domain at $\vec{\psi}_a(1)$, the variation of the individual fluence $\Delta\vec{\phi}(n-1)$ for the complete pulse domain and the individual fluence vector $\vec{\phi}(\hat{n})$ for the effective pulse domain at $\vec{\psi}_a(2)$.

Figure 2.5: Simulation result #7 of a 2d-HODIC. The grey scale represents (a,c) the individual and (b) the accumulated fluence in J/cm^2 .

Figures 2.5a and 2.5b show the spatial differences between the individual and accumulated fluence distribution. For this relative high pulse overlap, the accumulated fluence per repetition is several orders of magnitude higher than the peak fluence. The spatial distribution of the average individual fluence has the same profile as the accumulated fluence. The individual fluence is characterised by a high number of pulse contributions and turning points. For both analysis locations, the individual fluence shows high symmetry, only shifted about a certain number of pulses due to

Table 2.2: Values for input and output parameters of simulations #1 to #7.

#	ϕ_0	φ	N_r	ϕ_a	$\hat{\phi}_a$	$\hat{\phi}_p$	$\hat{\phi}_v$	$\hat{\phi}_d$
-	J/cm ²	-	-	J/cm ²	J/cm ²	J/cm ²	J/cm ²	J/cm ²
1 $\vec{\psi}_a(1)$	1.000	-	5	1.000	1.000	1.000	1.000	0.000
2 $\vec{\psi}_a(1)$	1.000	0.75	5	0.147	0.313	1.000	0.000	1.000
3 $\vec{\psi}_a(1)$	1.000	0.75	5	0.022	0.134	1.000	0.000	1.000
4 $\vec{\psi}_a(1)$	1.000	-0.75	1	0.111	1.000	1.000	1.000	0.000
5 $\vec{\psi}_a(1)$	1.000	0.40	1	0.025	0.138	1.000	0.003	0.997
6 $\vec{\psi}_a(1)$	1.000	0.63	1	0.024	0.137	1.000	0.004	0.996
7 $\vec{\psi}_a(1)$	1.000	0.95	1	0.024	0.126	1.000	0.000	1.000
1 $\vec{\psi}_a(2)$	1.000	-	5	0.630	0.630	0.630	0.630	0.000
2 $\vec{\psi}_a(2)$	1.000	0.80	5	0.117	0.249	0.754	0.001	0.753
3 $\vec{\psi}_a(2)$	1.000	0.75	5	0.022	0.126	0.903	0.001	0.902
4 $\vec{\psi}_a(2)$	1.000	-0.75	1	0.070	0.630	0.630	0.630	0.000
5 $\vec{\psi}_a(2)$	1.000	0.40	1	0.022	0.133	0.630	0.002	0.628
6 $\vec{\psi}_a(2)$	1.000	0.63	1	0.024	0.119	0.630	0.001	0.629
7 $\vec{\psi}_a(2)$	1.000	0.95	1	0.024	0.124	0.994	0.000	0.993
#	$\hat{\Gamma}_a$	$\hat{\Gamma}_p$	$\hat{\Gamma}_v$	$\hat{\Gamma}_d$	$\hat{\Gamma}_c$	$\hat{\Gamma}_0$	N_p	$\hat{N}_p$
-	J/cm ²	J/cm ²	J/cm ²	J/cm ²	J/cm ²	J/cm ²	-	-
1 $\vec{\psi}_a(1)$	0.488	5.000	0.000	5.00	2.500	1.000	5	5
2 $\vec{\psi}_a(1)$	3.917	12.533	0.004	12.529	6.269	2.507	85	40
3 $\vec{\psi}_a(1)$	31.416	31.416	31.416	0.000	31.416	6.283	1445	235
4 $\vec{\psi}_a(1)$	0.098	1.000	0.000	1.000	0.500	1.000	9	1
5 $\vec{\psi}_a(1)$	1.079	1.237	0.954	0.284	1.095	1.237	49	9
6 $\vec{\psi}_a(1)$	2.868	2.870	2.867	0.003	2.869	2.870	121	21
7 $\vec{\psi}_a(1)$	157.08	157.08	157.07	0.006	157.08	157.08	6561	1251

the larger distance of $\vec{\psi}_a(2)$ from the starting location. If the individual fluence is plotted semi-logarithmic, the distribution of contributions over the complete fluence spectrum can be analysed. This figure shows that for pulses at a large distance, the fluence contribution is very small. That is why the effective individual fluence is introduced. However, this type of analysis also allows to identify the effect of different translation paths. For this particular pulse overlap, the effective individual fluence as well as the individual fluence variation contains a large number of contributions in the order of the peak fluence.

Compared to the previous examples, this example represents the case of a relatively high pulse overlap. If this example is compared to #6, it can be observed that ϕ_a does not decrease but $\hat{\Gamma}_p$ increase, so that this value is significantly larger than ϕ_0 and $\hat{\phi}_a$. That means the individual fluence is significant different from the

accumulated fluence. Without determining these irradiation output parameters, no quantitative information about the actually applied irradiation is available. Therefore, the theoretical determination of the applied irradiation is an important step for the experimental practice. This simulation confirms the results found in the previous simulations for different values of φ and shows how different DIC can be compared to SIC, in terms of individual fluence, accumulated fluence and number of pulses, dependent on the selected φ . However, for this high value of φ the differences, for the individual and accumulated fluence, between different locations decrease in the centre of the irradiated area, which is again different from static.

2.4.2 Displacement analysis

Based on the simulations, presented in the subsection before, the properties of the fluence accumulation process have been analysed. For this purpose, various characterisation parameters were used. Of particular interest was the variation of these parameters with respect to lateral displacements, which have been represented by the pulse overlap. After these simulations the dependence of the characterisation parameters on the overlap, also for different locations, remained unknown.

The aim of this subsection is to identify how specific output parameters depend on the pulse overlap. Therefore, the simulations are repeated for different values of the pulse overlap between $\varphi = -1$ to $\varphi = 0.9875$ in $\Delta\varphi = 0.0125$ steps and the selected output parameters are recorded. If not stated differently the first batch of simulations applies the same computation conditions as for example #7 in the subsection before.

Figures 2.6a and 2.6b show the dependence of the number of pulses for 1d-DIC and 2d-DIC on φ . The solid lines represent the data obtained from Equations 2.30 for 1d-DIC and 2.31 for 2d-DIC, the dashed lines from Equation 2.49 and the dash-dot lines from Equation 2.50. In the analytical cases, 1d-DIC and 2d-DIC differ by the square. The number of irradiation repetitions was 1.

With increasing φ , the number of contributing pulses increases non-linear and monotonically and converges to infinity if φ approaches 1. The number of pulses on an area is quadratic higher compared to a line. For an area of length $l_x = l_y = 6\omega_0$ and $\varphi = 0.0$, $\hat{N}_p = 3$ for a line and $\hat{N}_p = 9$ for an area. At high values of φ , for example $\varphi = 0.9$, small changes in φ cause a large changes in N_p and $\hat{N}_p$, which can be orders of magnitude different.

In practice and in the simulations, N_p and N_x can only be natural numbers. This results, under certain conditions, in a non-optimal filling of the irradiated area. Consequently, the length of the optically irradiated area can deviate from the geometrically defined. In the simulations it is required to round either N_p or N_x . If N_p would be rounded, N_x would include all positive integer values. In this case, the location of a displacement would vary according to odd and even contributions. For this reason, N_x is rounded in this computation and with an odd pulse number stable displacement and analysis locations are achieved.

The rounding does not affect the overlap between pulses. It adds or subtracts at the outer edge of the area pulses and therefore increases or decreases l_x . In Figure 2.6c the dependence of the length l_x on the φ is shown. This figure shows the variation

Figure 2.6: Simulation results for the dependence of N_p , $\hat{N}_p$, l_x , $\tilde{\Gamma}_p$, $\tilde{\Gamma}_v$ and $\tilde{\Gamma}_d$ on φ under 1d- and 2d-DIC.

of the length due to the rounding procedure. The variation is distinctive at low pulse overlaps and effects notably the length of small areas. This also adds or subtracts an additional fluence contribution. But this is, due to the large distance, usually neglectable in the centre.

Figures 2.7a to 2.7i show the dependence of ϕ_a , $\hat{\phi}_a$ and ϕ_p on φ for three different simulation batches. The solid lines represent the data obtained at location $\vec{\psi}_a(1)$ and the dashed lines at $\vec{\psi}_a(2)$. The top figures (a-c) show the first simulation batch for a fluence boundary $\phi_b = \phi_0/e^8$ at $\vec{\psi}_a(1) = [x=0, y=0]^T$ and $\vec{\psi}_a(2) = [x=\omega_0/3, y=\omega_0/3]^T$, the middle figures (d-f) the second batch for $\phi_b = \phi_0/e^{24}$ at the same locations and the bottom figures (g-i) the third batch with the same ϕ_b at $\vec{\psi}_a(1) = [x=0, y=0]^T$ and $\vec{\psi}_a(2) = [x=x_0/2, y=y_0/2]^T$. It should be noted, that in the last batch $\vec{\psi}_a(2)$ is different for each φ and represents the top right valley next to the origin.

The first batch shows, that with increasing φ , ϕ_a and $\hat{\phi}_a$ decrease at $\vec{\psi}_a(1)$ and $\vec{\psi}_a(2)$. At both analysis locations the values for ϕ_a are different for low pulse overlaps and the same values are approached for high pulse overlaps. The same is true for

Figure 2.7: Simulation results for the dependence of ϕ_a , $\hat{\phi}_a$ and ϕ_p on φ for three different simulation batches.

$\hat{\phi}_a$. With these results, these simulations confirm the observations from the previous subsection. Since for a certain geometrical area only an integer value of the number of pulses contributes, the course of the average fluence varies according to the pulse number contributions. The parameter $\phi_a < \hat{\phi}_a$, because $N_p > \hat{N}_p$. The parameter $\phi_p = \phi_0$ at $\vec{\psi}_a(1)$. At $\vec{\psi}_a(2)$ this parameter is determined for low pulse overlaps by

Table 2.3: Values for relative accumulation parameters under 2d-DIC.

φ	$\hat{\phi}_a(1)$	$\hat{\phi}_a(2)$	$\Delta\hat{\phi}_a$	$\tilde{\Gamma}_p$	$\tilde{\Gamma}_v$	$\tilde{\Gamma}_d$	$\tilde{N}_p$
-1.000	1.000	0.000	1.000	1.000	0.000	1.000	1
-0.500	0.200	0.000	0.200	1.000	0.000	1.000	5
± 0.000	0.111	0.006	0.105	1.001	0.073	0.928	9
+0.250	0.050	0.026	0.023	1.045	0.422	0.623	21
+0.500	0.044	0.048	-0.004	1.616	1.526	0.090	37
+0.600	0.040	0.041	-0.001	2.459	2.450	0.009	61
+0.700	0.043	0.039	0.004	4.363	4.363	0.000	101
+0.750	0.043	0.043	0.001	6.283	6.283	0.000	145
+0.800	0.041	0.041	0.000	9.818	9.818	0.000	241
+0.850	0.042	0.041	0.000	17.453	17.453	0.000	421
+0.900	0.041	0.042	-0.001	39.270	39.270	0.000	949
+0.925	0.041	0.042	0.000	69.813	69.813	0.000	1685
+0.950	0.042	0.042	0.000	157.080	157.080	0.000	3761
+0.975	0.042	0.042	0.000	628.319	628.319	0.000	15069

the relative position of the analysis location to the centre of the pulse, until at higher pulse overlaps larger contributions from other displacements reach this location.

If ϕ_b is varied, as in the second batch, ϕ_a and ϕ_p do not change, but $\hat{\phi}_a$ contains pulses with different fluence values. In the second batch ϕ_b has been decreased, so that pulses at a lower overlap and fluence contribute. The overall course of this two curves has hardly changed, but since more values have been considered, the curves converge to a smaller value. The fluence boundary determines at which overlap the average fluence starts to decrease and to which value the average fluence converges. If $\vec{\psi}_a(2)$ is coincident with the location of Γ_v , as in the third batch, the values ϕ_a , $\hat{\phi}_a$ and ϕ_p are zero at $\vec{\psi}_a(2)$ for low values of φ and increase with increasing φ .

The parameters ϕ_a , $\hat{\phi}_a$ and ϕ_p are different for the two analysis locations under LODIC and these differences vanish towards HODIC. This means under HODIC different locations receive a similar individual fluence. However, at $\vec{\psi}_a(1)$ the value for the average fluence of a certain location at low pulse overlaps is different compared to the value at high overlaps. This means, with increasing φ more pulses contribute with smaller fluences, which cause the average single pulse fluence to decrease. But the exact value to which the average fluence converges depends on the fluence boundary. Without any data about the laser-material interaction process, this boundary value cannot be determined.

Figures 2.6d and 2.6f show the dependence of $\tilde{\Gamma}_p$, $\tilde{\Gamma}_v$, $\tilde{\Gamma}_d$ on φ . The data for $\tilde{\Gamma}_p(\varphi)$ have been obtained from Equation 3.24 on page 56. The data for $\tilde{\Gamma}_v(\varphi)$ have been obtained from the same equation, but evaluated for the valley location. This equation delivers the same results as the numerical computation with a space domain boundary $x_b = x_0/2 = y_b = y_0/2$.

As can be observed, the peak and valley accumulation increases while the peak-to-valley difference decreases non-linearly with increasing pulse overlap. The course of $\tilde{\Gamma}_p$ is similar to N_p , but the quantities are different. The values for these parameters are different for 1d- and 2d-DIC. The parameter $\tilde{\Gamma}_p$ is smaller under 1d-DIC, because less pulses contribute. Next, a certain $\tilde{\Gamma}_d$ value is reached at smaller values of φ under 1d-DIC, because φ is defined along the axes. This is also the case for the cross-section under 2d-DIC.

Furthermore, these parameters depend on the analysis location and on the size of the irradiated area. If the analysis location would be constant, for example at $x_b = y_b = 2\omega_0$, $\tilde{\Gamma}_d$ would increase at high values of the pulse overlap. In this simulation, the analysis space domain was limited to a defined valley value and follows the valley position with increasing φ . As a result, the dependence decreases monotonically. This figure is also representative for irradiated areas, which are large compared to the beam radius. Table 2.3 shows a selection of values, which are frequently applied in practice, for the discussed parameters under 2d-DIC.

The accumulated peak-to-valley difference fluence decreases with increasing pulse overlap. That means, that the accumulated fluence for different locations gets similar, as observed for the individual fluence. However, at the same time the accumulated fluence increases with increasing pulse overlap. For high values of φ the accumulated peak fluence can be about orders of magnitude larger compared to low pulse overlaps and small variations cause large differences in accumulated fluence. Both effects, the increase of the accumulated fluence and the decrease of the individual fluence at $\vec{\psi}_a(1)$, take place with increasing φ . This information is important for the determination of irradiation parameters in practice.

Based on the data presented in Figure 2.6f a coarse separation of the different pulse overlap regimes can be made. The LODIC regime can be defined $\varphi < -0.50$, where the parameter $\tilde{\Gamma}_d$ converges to 1, the MODIC $-0.50 \leq \varphi \leq 0.50$, where $\tilde{\Gamma}_d$ takes values between 0.9 and 0.1 and the HODIC $0.50 < \varphi$, where $\tilde{\Gamma}_d$ converges to 0. However, the exact classification and separation of the different overlap regimes depends on the considered fluence and space domain boundaries.

2.5 Conclusions

In order to support the understanding of the spatial emergence of LIPSS, an irradiation model has been presented. This model has been applied to analyse the fluence accumulation process for different irradiation conditions.

Whereas, under SIC the individual fluence is constant and the accumulated fluence distribution is Gaussian, under DIC the individual fluence is varying and the accumulated fluence distribution is non-Gaussian. Under SIC, the irradiation input parameters provide comprehensible information about the output parameters, which is not the case under DIC, where the parameter combinations (ϕ_0, ϕ_p) and (N_r, N_p) partially lose their meaning for a comprehensible description of the energy input.

The DIC are characterised by three different pulse overlap regimes low, medium and high overlap DIC. With decreasing pulse overlap, the DIC change at some point

in an effective SIC, for the individual as well as the accumulated fluence. The effective SIC regime corresponds approximately to the LODIC regime. However, the exact classification and separation of the different overlap regimes depends on the considered fluence and space domain boundaries. With increasing pulse overlap the DIC get significantly different from SIC, the individual and accumulated fluence vary per location and approach similar values for different locations.

Chapter 3

Spatial emergence

3.1 Introduction

The objective of this thesis has been supplemented by two boundary conditions, one for the approach and one for the basis. The phenomenological approach, does not refer to a theoretical or experimental methodology. It means, rather than to differentiate individual laser-material interaction steps, to directly relate irradiation parameters and spatial emergence. So far, the spatial emergence of surface modifications has been described by ϕ_{th} and for specific materials with incubation properties a certain empirical relation between ϕ_{th} and N_p has been found. This knowledge, determined by microscopy of samples after static irradiation experiments, forms the basis for this research. Therefore, the aim of this thesis can be rendered more precisely, to investigate the phenomenological relation between certain irradiation parameters and the spatial emergence of LIPSS starting from the knowledge about incubation. In this theoretical chapter, this relation is investigated for static and dynamic irradiation conditions.

3.2 Static irradiation

3.2.1 Analysis conditions

Assumptions

This chapter applies the primary assumptions made in Chapter 2. But in this chapter, a sample is assumed to be placed in the optical path. The surface of the sample is coincident with the irradiation plane and this plane is referred to as sample plane. The sample geometry is assumed to be flat, with edge lengths larger than l_x in x and l_y in y direction. The centre of the area is assumed to be in the origin of the Cartesian frame. The surface of the sample is assumed to have a polished finish without any surface shape errors. The material is assumed to be homogeneous and of metal composition.

In this section SIC are investigated, which can be described by Equations 2.18 and 2.19. For this purpose, these equations can be rewritten as

$$\phi(r) = \phi_0 \exp\left(-2\left(\frac{r^2}{\omega_0^2}\right)\right) \quad (3.1)$$

for the individual fluence and as

$$\Gamma(r) = N_r \phi_0 \exp\left(-2\left(\frac{r^2}{\omega_0^2}\right)\right) \quad (3.2)$$

for the accumulated fluence, where r denotes the radial coordinate of a polar coordinate system, with $N_p \equiv N_r$ under SIC, N_p is substituted by N_r and the starting displacement is assumed to be $x_s = y_s = 0$.

In literature, often the influence of the irradiation input parameters ϕ_0 and N_r on the spatial emergence has been investigated. The spatial emergence has been described by the output parameters ϕ_{th} , as local individual fluence, and N_p . If all other parameters, as λ , τ , f_r , f_l , ω_0 etc., are constant and only these parameters are varied, an experimental matrix spanned between ϕ_0 and N_r and individual static irradiation experiments is obtained. Each individual experiment is performed on a different location. For each location only one input parameter ϕ_0 or N_r is varied. Figure 3.1 shows a schematic representation of a static irradiation experiment. The peak fluence can be varied by P_a , which has for otherwise constant parameters a linear relation, compare Equations 2.22 and 2.25. In the framework of the irradiation model N_p is considered as an output parameter. In order to achieve different SIC the input parameter N_r is varied.

The given laser-material combination, is assumed to yield an interaction which results after pulsed irradiation in different types of permanent modifications on the sample surface, including certain types of LIPSS. The time between pulses is assumed to be large, so that transient phenomena, as plasma emergence or heat accumulation, do not affect the beam propagation and material interaction of subsequent pulses. The irradiated locations are investigated by microscopy techniques, as suggested by Vogel [93]. The spatial emergence under static single and multi pulse irradiation conditions is assumed to show defined spatial transitions between different surface modifications, so that a similar result as shown in Figure 1.1 for single pulse modifications is obtained. The lateral dimensions of the surface modifications are quantified by the diameter D .

Descriptors

Vogel suggested, that the spatial emergence of surface modifications correlates to the energy distribution [93] and Liu applied this correlation to determine the focused beam radius in the sample plane [95]. Therefore, Liu assumed that the spatial emergence for single pulse irradiation can be described phenomenologically by a defined threshold value ϕ_{th} , which corresponds to the individual fluence at the location of the outer modification diameter. If the peak fluence exceeds the threshold value the modification is assumed to emerge on the surface. The fluence threshold values are

Figure 3.1: Schematic representation of a static irradiation experiment.

specific for the type of surface modification and laser-material combination. Later on, the fluence thresholds have been determined for different values of N_p [96, 97].

For multi pulse irradiation conditions where $\phi_0 < \phi_{th}(1)$, a modification emerges on the surface after a summation of individual material changes and that means after a certain number of pulses has been applied. The parameter N_p represents phenomenologically when a certain modification emerges. The spatial emergence of a certain modification can also be described by a threshold N_{th} . If the peak fluence is equal to the individual fluence threshold of a certain modification $\phi_0 = \phi_{th}(N_p)$, then the modification emerges on the surface after a specific value $N_r = N_{th}(\phi)$ has been exceeded. The dependency of the threshold N_{th} on the individual fluence, yields another way to describe the spatial emergence.

However, since N_p can only take integer values, also the spatial emergence can only be described by discrete values of $N_{th}(\phi)$. This is a general limitation of the parameter N_p for the description of the spatial emergence. The spatial emergence of surface modifications is usually continuous. Therefore, another way to describe the spatial emergence can be introduced if the accumulated fluence is determined for each irradiation experiment. The accumulated fluence can take other values than integers and can be assigned to different spatial observations. Beyond this, the accumulated fluence correlates to accumulated damage. The accumulated peak fluence can be defined in static as $\Gamma_p = N_r \phi_0$. Dependent on which irradiation input parameter is varied, a threshold value $\Gamma_{th}(N_p)$ for varying ϕ_0 or $\Gamma_{th}(\phi)$ for varying N_r can be determined.

The spatial emergence of laser-induced surface modifications, where LIPSS belong to, can be described by different parameters: ϕ_{th} , N_{th} and Γ_{th} . Dependent on which irradiation input parameter is varied, N_r or ϕ_0 , the spatial emergence can be analysed by two dependencies. However, modifications induced by multiple laser pulses, become visible on the surface as a result of an accumulated material change. Therefore, the following analysis is particularly concerned with the quantity Γ_{th} .

3.2.2 Incubation properties

In this subsection the multi pulse behaviour of the previously defined threshold parameters is investigated. Therefore, the material is assumed to show incubation in the static multi pulse irradiation experiment. If the spatial emergence of a certain surface modification is described by the individual fluence threshold, incubation means that this threshold depends on N_p . In particular, it has been found that $\phi_{th}(N_p)$ decreases non-linear with increasing N_p for a constant ϕ_0 per irradiation experiment. With respect to the assumed laser-material combination, the dependence of this decay is assumed to follow the empirical law

$$\phi_{th}(N_p) = \phi_{th}(1) N_p^{\xi-1} \quad (3.3)$$

presented by Jee [96]. Equation 3.3 relates the threshold $\phi_{th}(N_p)$ after N_p pulses to the threshold of a single pulse $\phi_{th}(1)$. Here $\xi \in \{0 : 1\}$ is a material dependent coefficient, referred to as the incubation factor. The dependence of ϕ_{th} on N_p is shown in Figure 3.2a for typical values of the assumed conditions. The curve starts with the initial value $\phi_{th}(1) = 0.15 \text{ J/cm}^2$ and decays between $N_p = 1$ and $N_p = 1000$ with $\xi = 0.86$. For $\xi = 1$ the threshold is independent of N_p and for $\xi < 1$ the threshold is dependent on N_p and decreases for smaller values of ξ .

Certain surface modifications have only been observed in a specific range of N_p . For example grooves are usually not observed for $N_p = 1$. In this case the value $\phi_{th}(1)$ is extrapolated although the phenomenon has not been observed at $N_p = 1$. Next to this, Equation 3.3 describes a non-physical behaviour for the boundary value $\phi_{th}(N_p) = 0$ if $N_p \rightarrow \infty$. This means that for very high values of N_p this equation may yield a deviation. However, this equation was found to be valid for a finite range of values for N_p .

The incubation effect can also be formulated in terms of accumulated fluence. Rewriting Equation 3.3 for the accumulated fluence gives

$$\Gamma_{th}(N_p) = \phi_{th}(N_p) N_p = \phi_{th}(1) N_p^{\xi-1} N_p = \Gamma_{th}(1) N_p^{\xi}, \quad (3.4)$$

which describes the non-linear increase of the threshold $\Gamma_{th}(N_p)$ for accumulated fluence with N_p . Taking into account that $\Gamma_{th}(1) = \phi_{th}(1)$, the curve for $\Gamma_{th}(N_p)$ can be plotted with the same data as for the curve of $\phi_{th}(N_p)$. The dependence of Γ_{th} on N_p is shown in Figure 3.2b. In comparison with the individual fluence threshold, which is reached at lower values with increasing N_p , the accumulated fluence threshold is reached at higher values. For $\xi = 1$ the threshold is again independent of N_p but also of the accumulated fluence and for $\xi < 1$ the dependence is given and the

Figure 3.2: Simulation results for the dependence of the (a) individual and (b) accumulated fluence threshold on the pulse number as well as the dependence of the (c) pulse number and (d) accumulated fluence threshold on the individual fluence.

accumulated fluence threshold is reached at smaller values for smaller values of ξ . To summarise, for increasing N_p more accumulation is required, but less for decreasing ξ . The same data, but in a double logarithmic representation, are used to determine ξ , which represents the slope in this graph.

Incubation formulates a relation between ϕ_{th} and N_p or Γ_{th} and N_p . In Equation 3.3, N_p is the independent variable representing the cause and $\phi_{th}(N_p)$ the depending variable representing the effect. However, in the experimental matrix for each

separate N_p -on-1 irradiation always one variable is changed and the other is constant. This allows to analyse the observations with exchanged variables. If the independent variable is changed from N_p to ϕ and the dependent from $\phi_{th}(N_p)$ to $N_{th}(\phi)$, based on Equation 3.3, the incubation dependence of the repetition threshold on the local individual fluence is obtained and given by equation

$$N_{th}(\phi) = \left(\frac{\phi}{\phi_{th}(1)} \right)^{\frac{1}{\xi-1}}. \quad (3.5)$$

The dependence of N_{th} on ϕ is shown in Figure 3.2c for the same values as assumed in Figure 3.2a. The curve decreases non-linear between $\phi = 0.05 \text{ J/cm}^2$ and $\phi = 0.15 \text{ J/cm}^2$ with $\xi = 0.86$. The Equations 3.3 and 3.5 and the Figures 3.2a and 3.2c describe the same observations. For $\xi = 1$ the threshold $N_{th}(\phi)$ is not defined and for $\xi < 1$ the threshold depends on the local individual fluence and is reached at smaller values for smaller values of ξ . This means for smaller values of ϕ more repetitions are required to achieve the modification and for smaller values of ξ less N_p .

In a similar manner as $\Gamma_{th}(N_p)$, the dependence of the accumulated fluence threshold on the local individual fluence can be obtained by

$$\Gamma_{th}(\phi) = \phi N_{th}(\phi) = \phi \left(\frac{\phi}{\Gamma_{th}(1)} \right)^{\frac{1}{\xi-1}}. \quad (3.6)$$

The dependence of Γ_{th} on ϕ is shown in Figure 3.2d for the same values as assumed in Figure 3.2a. The curve decreases non-linear between $\phi = 0.05 \text{ J/cm}^2$ and $\phi = 0.15 \text{ J/cm}^2$ with $\xi = 0.86$. The Equations 3.4 and 3.6 and Figures 3.2b and 3.2d reveal the same observations only for different dependent variables. For $\xi = 1$ the threshold is again not defined and for $\xi < 1$ the accumulated fluence threshold is dependent on ϕ and reached at smaller values for smaller values of ξ . For smaller values of ϕ more accumulation is required and for smaller values of ξ less.

The spatial emergence of LIPSS can be described by different parameters, as ϕ_{th} , N_{th} and Γ_{th} and, for materials which show incubation properties, the dependence of these parameters on the irradiation output parameters ϕ or N_p can be analysed. The analysis of the influence of incubation on the spatial emergence by using the accumulated fluence, allows to compare the effect of the variation of two different irradiation input parameters by one quantity.

For increasing N_p the individual fluence threshold is reached at lower values and the accumulated fluence threshold at higher, as indicated in Figures 3.2a and 3.2b. For smaller values of ξ both thresholds are reached at lower values. With increasing ϕ the required irradiation repetition and fluence accumulation, to induce a certain surface modification, decrease, as indicated in Figures 3.2c and 3.2d. For smaller values of ξ both thresholds are reached at lower values.

The parameter Γ_{th} describes when a certain phenomenon spatially emerges and, for materials which show incubation properties, this parameter is a function of the local individual fluence $\Gamma_{th}(\phi)$ and number of pulses $\Gamma_{th}(N_p)$. The dependencies are different, with increasing N_p the accumulated fluence threshold increases and with increasing ϕ the threshold decreases. The dependency of ϕ_{th} on N_p and N_{th} on ϕ , also indicate the inverse dependency of the irradiation input parameters ϕ_0 and N_r .

3.2.3 Domain requirements

In this subsection, the general knowledge about thresholds and their dependencies is applied to discuss properties of specific surface modifications. The fluence modification diagram, presented in Figure 1.1, summarises and classifies the experimentally observed single pulse surface modifications. The observations at multi pulse values N_p can also be summarised and classified in such a diagram. Therefore, Γ can be used instead of ϕ in a fluence modification diagram.

In this case, the accumulated fluence threshold $\Gamma_{th(i)}(N_p)$ indicates a lower boundary to obtain the i 'th modification on the surface. However, $\Gamma_{th(i)}(N_p)$ of this modification also indicates an upper boundary for the modification below this accumulated fluence. In general, modifications, observed in a zone, do not only emerge on the surface if the accumulated fluence threshold $\Gamma_{th(i)}(N_p)$ has been exceeded, zonular modifications start to disappear if the next accumulated fluence value

$$\Gamma_{th(i+1)}(N_p) = \Gamma_{th(i)}(N_p) + \Delta\Gamma_{th(i)}(N_p) \quad (3.7)$$

has been exceeded. The spatial emergence of a zonular modification is actually not determined by one boundary $\Gamma_{th(i)}$ it also depends on a second boundary $\Gamma_{th(i+1)}$. That means, zonular modifications emerge only if the accumulated fluence deposited on the surface is within a specific range of accumulated fluence. In this thesis, this range of accumulated fluence will be referred to as accumulated fluence domain. The domain has a location, described by $\Gamma_{th(i)}$ and $\Gamma_{th(i+1)}$, and a size, described by $\Delta\Gamma_{th(i)}$.

LIPSS belong to the group of zonular surface modifications [96, 97, 130]. Assuming that LIPSS and the modifications above and below have been observed over a certain range of N_p and that the observation of these modifications can also be described by the incubation law, then, similar to Equation 3.4, also this upper domain boundary can be formulated in terms of accumulated fluence

$$\Gamma_{th(i)}(N_p) + \Delta\Gamma_{th(i)}(N_p) = (\Gamma_{th(i)}(1) + \Delta\Gamma_{th(i)}(1)) N_p^\xi, \quad (3.8)$$

where $\Gamma_{th(i)}(1) + \Delta\Gamma_{th(i)}(1)$ can also be an extrapolated value used to describe the emergence around the value N_p . Figure 3.3a shows the dependence of the domain location and size on N_p . The curve $\Gamma_{th(1)}(N_p)$ uses the same values as assumed in Figure 3.2a and $\Gamma_{th(2)}(N_p)$ starts with the initial value $\Gamma_{th(2)}(1) = 0.20 \text{ J/cm}^2$. With increasing N_p the location and size of the accumulated fluence domain increases.

If the range of N_p is sufficiently large, the boundaries of a zonular modification will be exceeded by a variation of N_r for a constant ϕ_0 . Basically, the domain can be approached by varying two different input parameters, variation of ϕ_0 which yields the domain size $\Delta\Gamma_{th(i)}(N_p)$ or N_r which yields $\Delta\Gamma_{th(i)}(\phi)$. The upper domain boundary can also be described by

$$\Gamma_{th(i)}(\phi) + \Delta\Gamma_{th(i)}(\phi) = \phi \left(\frac{\phi}{\Gamma_{th(i)}(1) + \Delta\Gamma_{th(i)}(1)} \right)^{\frac{1}{\xi-1}}. \quad (3.9)$$

Equations 3.8 and 3.9 are identical to Equations 3.4 and 3.6, but with different initial values. Figure 3.3b shows the curve $\Gamma_{th(1)}(\phi)$ together with $\Gamma_{th(2)}(\phi)$ for the same

Figure 3.3: Simulation results for the accumulated fluence domain dependent on (a) the number of pulses and (b) the individual fluence.

values as assumed in Figure 3.3a. With increasing ϕ the location and size of this accumulated fluence domain decreases.

In general, the values for the domain sizes $\Delta\Gamma_{th(i)}(N_p)$ and $\Delta\Gamma_{th(i)}(\phi)$ depend on the relative position of these curves and on the considered threshold value on the curve. In particular, the example presented in Figures 3.3a and 3.3b shows that for the same accumulated fluence threshold value, the accumulated fluence domain size is greater by varying N_p than ϕ . In fact, the accumulated fluence domain is determined by the incubation properties of the material and their location and size depends on the applied irradiation parameters.

In Subsection 3.3.2, SIC are proposed for the determination of values for the domain boundaries. Therefore, it is important to note, that the domain boundaries depend on the varied irradiation parameter. This correlation is discussed below and for that purpose example values are assumed for the equations shown above. If the irradiation input parameters are assumed to be $\phi_0 = \phi_{th(i)}(N_p) = 0.071 J/cm^2$ and $N_r = N_{th(i)}(\phi) = 200$ then the lower domain boundaries are obtained by

$$\Gamma_{th(i)}(N_{th(i)}) = \Gamma_{th(i)}(1)(N_{th(i)}(\phi))^\xi \quad (3.10)$$

and

$$\Gamma_{th(i)}(\phi_{th(i)}) = \phi_{th(i)}(N_p) \left(\frac{\phi_{th(i)}(N_p)}{\Gamma_{th(i)}(1)} \right)^{\frac{1}{\xi-1}} \quad (3.11)$$

and yield $\Gamma_{th(i)}(N_{th(i)}) = \Gamma_{th(i)}(\phi_{th(i)}) = 14.29 J/cm^2$. By increasing ϕ_0 , the upper pulse number dependent domain boundary is obtained by

$$\Gamma_{th(i)}(N_{th(i)}) + \Delta\Gamma_{th(i)}(N_{th(i)}) = (\Gamma_{th(i)}(1) + \Delta\Gamma_{th(i)}(1))(N_{th(i)}(\phi))^\xi \quad (3.12)$$

and yields 19.05 J/cm². By increasing N_r , the upper individual fluence dependent domain boundary is obtained by

$$\Gamma_{th(i)}(\phi_{th(i)}) + \Delta\Gamma_{th(i)}(\phi_{th(i)}) = \phi_{th(i)}(N_p) \left(\frac{\phi_{th(i)}(N_p)}{\Gamma_{th(i)}(1) + \Delta\Gamma_{th(i)}(1)} \right)^{\frac{1}{\xi-1}} \quad (3.13)$$

and yields 148.71 J/cm². These equations yield for the upper domain boundaries different values. This is because, the value $\Gamma_{th(i)}(N_{th(i)}) + \Delta\Gamma_{th(i)}(N_{th(i)})$ is actually determined for a different individual fluence as $\Gamma_{th(i)}(N_{th(i)})$ and $\Gamma_{th(i)}(\phi_{th(i)}) + \Delta\Gamma_{th(i)}(\phi_{th(i)})$ is determined for a different number of repetitions as $\Gamma_{th(i)}(\phi_{th(i)})$. If this is considered in the Equations 3.12 and 3.13, the same values can be obtained for the pulse number dependency

$$\begin{aligned} & \Gamma_{th(i)}(N_{th(i)} + \Delta N_{th(i)}) + \Delta\Gamma_{th(i)}(N_{th(i)} + \Delta N_{th(i)}) \\ &= (\Gamma_{th(i)}(1) + \Delta\Gamma_{th(i)}(1)) (N_{th(i)}(\phi) + \Delta N_{th(i)}(\phi))^{\xi} \end{aligned} \quad (3.14)$$

and individual fluence dependency

$$\begin{aligned} & \Gamma_{th(i)}(\phi_{th(i)} + \Delta\phi_{th(i)}) + \Delta\Gamma_{th(i)}(\phi_{th(i)} + \Delta\phi_{th(i)}) \\ &= (\phi_{th(i)}(N_p) + \Delta\phi_{th(i)}(N_p)) \left(\frac{\phi_{th(i)}(N_p) + \Delta\phi_{th(i)}(N_p)}{\Gamma_{th(i)}(1) + \Delta\Gamma_{th(i)}(1)} \right)^{\frac{1}{\xi-1}}. \end{aligned} \quad (3.15)$$

However, for the considered threshold $\Gamma_{th(i)}(N_{th(i)}) = \Gamma_{th(i)}(\phi_{th(i)})$ the resulting domain size for the pulse number dependence

$$\begin{aligned} \Delta\Gamma_{th(i)}(N_{th(i)}) &= \Gamma_{th(i)}(N_{th(i)}) + \Delta\Gamma_{th(i)}(N_{th(i)}) - \Gamma_{th(i)}(N_{th(i)}) \\ &= \Gamma_{th(i)}(\phi_{th(i)} + \Delta\phi_{th(i)}) + \Delta\Gamma_{th(i)}(\phi_{th(i)} + \Delta\phi_{th(i)}) \\ &\quad - \Gamma_{th(i)}(\phi_{th(i)}) \end{aligned} \quad (3.16)$$

is 4.76 J/cm² and for the individual fluence dependence

$$\begin{aligned} \Delta\Gamma_{th(i)}(\phi_{th(i)}) &= \Gamma_{th(i)}(\phi_{th(i)}) + \Delta\Gamma_{th(i)}(\phi_{th(i)}) - \Gamma_{th(i)}(\phi_{th(i)}) \\ &= \Gamma_{th(i)}(N_{th(i)} + \Delta N_{th(i)}) + \Delta\Gamma_{th(i)}(N_{th(i)} + \Delta N_{th(i)}) \\ &\quad - \Gamma_{th(i)}(N_{th(i)}) \end{aligned} \quad (3.17)$$

is 134.42 J/cm². The domain sizes are different dependent on which irradiation parameter is varied. In this example $\Delta\Gamma_{th(i)}(N_{th(i)}) < \Delta\Gamma_{th(i)}(\phi_{th(i)})$, because $\phi_{th(i)}(N_p)$ and $\phi_{th(i)}(N_p) + \Delta\phi_{th(i)}(N_p)$ have the same order of magnitude, whereas $N_{th(i)}(\phi)$ and $N_{th(i)}(\phi) + \Delta N_{th(i)}(\phi)$ are significantly different.

The domain sizes $\Delta\Gamma_{th(i)}(N_{th(i)})$ and $\Delta\Gamma_{th(i)}(N_{th(i)} + \Delta N_{th(i)})$ do not describe a different domain than $\Delta\Gamma_{th(i)}(\phi_{th(i)})$ and $\Delta\Gamma_{th(i)}(\phi_{th(i)} + \Delta\phi_{th(i)})$, only the size depends on the varied irradiation parameter and considered domain location. In the former case, the peak fluence is varied, while determining the lower and upper domain

boundary. In the latter case, these domain boundaries are determined which correspond to a certain peak fluence. Therefore, non-corresponding domain boundaries are determined, each of them with a different domain location and size.

In summary it can be said that the spatial emergence of LIPSS is determined by an accumulated fluence domain, whose location and size vary due to incubation with the applied irradiation parameters. In order to obtain a certain modification on the surface, this domain formulates requirements on the applied irradiation conditions.

3.2.4 Spatial characteristics

In the previous subsection, the spatial emergence has been discussed in the accumulated fluence domain. Therefore, the relations between the irradiation, threshold and material parameters have been investigated. In this subsection, this discussion is transferred into the space domain and the relations to spatial parameters are investigated. This particularly concerns the spatial emergences of the accumulated fluence domain under SIC.

The method of Liu [95] does not only yield the beam radius in the sample plane the method also yields a value for the individual fluence threshold. If the peak fluence exceeds this value, the modification spatially emerges on the surface and the diameter D increases. Liu deduced an analytical relation between the measured lateral dimension $D = 2r$ of the surface modification and the peak fluence. The same relation can also be derived for the accumulated fluence

$$D^2 = 2\omega_0^2 \ln \left(\frac{N_r \phi_0}{\Gamma_{th(i)}} \right). \quad (3.18)$$

With this equation, the diameter is a function of both irradiation input parameters and varies due to a different accumulated fluence. In this description, the threshold is constant, whereas, for materials which show incubation properties, the threshold is not constant and depends on the applied irradiation parameters.

In 2001 Bonse et al. considered incubation in the relation between the diameter of an ablation crater and the irradiation parameters ϕ_0 and N_p [102]. They derived a relation for the individual fluence and individual fluence threshold. A similar relation can also be derived for the accumulated fluence and accumulated fluence threshold of other phenomena

$$D(\phi_0, N_r) = \omega_0 \sqrt{2 \ln \left(\frac{N_r \phi_0}{\Gamma_{th(i)}(1) N_p^\xi} \right)}. \quad (3.19)$$

In this description, the diameter of the laser induced surface modification depends on the Gaussian distribution of the accumulated fluence and on the pulse number dependent accumulated fluence threshold, described by the irradiation input parameters ω_0 , ϕ_0 and N_r as well as the material parameter ξ and the laser-material interaction parameter $\Gamma_{th(i)}(1)$.

If, for multi pulse irradiation, ϕ_0 is varied, $\Gamma_{th(i)}(N_p)$ is constant and the accumulated fluence values for the Gaussian distribution change. Therefore, the accumulated

threshold is reached at a different spatial location, which corresponds to the Gaussian distribution. But, if N_r is varied, $\Gamma_{th(i)}(N_p)$ varies in addition to the change of the values for the Gaussian distribution. Therefore, the accumulated fluence threshold is reached at a different spatial location, which corresponds to the Gaussian distribution and the incubation law. If in particular N_r is increased, the diameter increases because the change of Γ_p is larger compared to the change of $\Gamma_{th(i)}(N_p)$.

A similar approach for the individual fluence dependent accumulated fluence threshold gives for the diameter

$$D(\phi_0, N_r) = \omega_0 \sqrt{2 \ln \left(\frac{N_r \phi_0}{\phi \left(\frac{\phi}{\Gamma_{th(i)}(1)} \right)^{\frac{1}{\xi-1}}} \right)}. \quad (3.20)$$

In order to calculate the diameter of the modification by this equation, the local individual fluence at the threshold diameter is considered. Equations 3.19 and 3.20 describe for corresponding input parameters the same output, but the threshold dependency is different for both equations. These equations yield the inner diameter of a zonular modification, if as threshold the upper domain boundary is applied.

If in Equation 3.20 N_r is varied, $\Gamma_{th(i)}(\phi)$ is constant and the accumulated fluence values for the Gaussian distribution change. Therefore, the accumulated threshold is reached at a different spatial location, which corresponds to the Gaussian distribution. But, if ϕ_0 is varied, $\Gamma_{th(i)}(\phi)$ varies in addition to the change of the values for the Gaussian distribution. Therefore, the accumulated fluence threshold is reached at a different spatial location, which corresponds to the Gaussian distribution and the incubation law. If in particular ϕ_0 is increased, the diameter increases because the threshold decreases and the accumulated fluence increases.

This is an interesting consideration, because if the accumulated fluence threshold depends on the individual fluence and the individual fluence varies in space, then also the accumulated fluence threshold varies in space. Whereas, for the pulse number dependence, the threshold is constant in space. If the complete individual fluence distribution according to Equation 3.1 is considered and in Equation 3.6 ϕ is substituted using Equation 3.1, then the spatial distribution of the accumulated fluence threshold

$$\Gamma_{th(i)}(r) = \phi_0 \exp \left(-2 \left(\frac{r^2}{\omega_0^2} \right) \right) \left(\frac{\phi_0 \exp \left(-2 \left(\frac{r^2}{\omega_0^2} \right) \right)}{\Gamma_{th(i)}(1)} \right)^{\frac{1}{\xi-1}}, \quad (3.21)$$

is obtained. In this case, the individual fluence distribution in space determines the spatial distribution of the accumulated fluence threshold. Because the individual fluence is Gaussian distributed in space, the accumulated fluence threshold decreases to the centre of the Gauss, while the accumulated fluence increases. The spatial emergence is determined by the intersection of the accumulated fluence threshold function with the accumulated fluence distribution. In case of the discussed zonular modifications, the spatial emergence is determined by the intersection of the accumulated fluence domain with the accumulated fluence distribution.

Figure 3.4: Simulation results for the spatial emergence under SIC, with (a) a constant threshold and (b) a varying threshold in space.

Figure 3.4 shows for the polar coordinate r the spatial distribution of the accumulated fluence and the accumulated fluence thresholds. The accumulated fluence distribution has been calculated with $\phi_0 = 0.15 \text{ J/cm}^2$, $\omega_0 = 10 \text{ } \mu\text{m}$ and $N_r = 200$. The accumulated fluence thresholds have been calculated with the same threshold and material parameters which have been used before.

In Figure 3.4a the accumulated fluence thresholds are constant in space, due to the constant pulse number, and in Figure 3.4b the thresholds vary in space, due to the varying individual fluence. The spatial emergence is represented by the projection of the intersections in the space domain. In this example, the surface modification emergences as an annular zone, with an inner radius of $4.8 \text{ } \mu\text{m}$ and outer of $6.1 \text{ } \mu\text{m}$. The apparent accumulated fluence domain is represented by the projection in the fluence domain. The lower boundary is reached at 14.29 J/cm^2 and the upper at 19.05 J/cm^2 . These values correspond to the accumulated fluence domain size $\Delta\Gamma_{th(i)}(N_p)$, which has been discussed in the subsection before.

In Subsection 3.2.3, the input parameters were varied to reach a certain threshold. If the threshold was reached, the surface modification emerged in the origin. In this subsection ϕ_0 and N_r have been further increased and the two different domain boundaries are reached at two different spatial locations in the tail of the Gauss. This is possible because the individual and accumulated fluence are varying per location.

Both the individual fluence and pulse number dependency yield the same result, which means both can be applied to describe the spatial emergence. But the assumption for the threshold dependency is different for both descriptions. In the one case, the individual fluence is assumed to vary the threshold, in the other the pulse num-

ber. Since the energy of a laser pulse changes the material, the individual fluence dependency should be the preferable description.

For the individual fluence dependence, the domain location and size vary per location. Therefore, the upper accumulated fluence domain boundary is reached at a different individual fluence than the lower. These boundaries do not correspond to the same individual fluence. As a consequence of this the fluence domain appears to be contracted and can be observed as an annular zone. In this case, incubation influences the spatial emergence, although the irradiation parameters are constant.

In conclusion: The spatial emergence of LIPSS is determined by the intersection of the applied accumulated fluence with the accumulated fluence domain boundaries. The location and size of the accumulated fluence domain vary due to incubation with the applied irradiation parameters. Both the individual fluence and pulse number dependency yield the same result for the spatial emergence. The annular zone usually observed under SIC, corresponds to the pulse number dependent fluence domain. This is important to note for the determination of irradiation parameters, but this also identifies limitations of the spatial emergence. In order to obtain LIPSS on the surface a minimum space is required and this space is determined by the spatial period Λ of the LIPSS type. If the accumulated fluence gradient is too strong or the fluence domain is too small, LIPSS do not emerge on the surface.

3.3 Dynamic irradiation

3.3.1 Lateral displacements

Introduction

This section applies the primary assumptions defined in Chapter 2 and the analysis conditions defined in Section 3.2. But contrary to the previous section, in this section DIC are assumed, which can be described by Equations 2.20 and 2.21. The aim of this subsection is to combine the knowledge about DIC, obtained in Chapter 2, and the spatial emergence under SIC, obtained in Section 3.2, in order to derive an understanding of the spatial emergence under lateral displacement irradiation conditions.

To repeat: The analysis of the individual and accumulated fluence in Chapter 2, revealed differences between SIC and DIC as well as LODIC and HODIC. Whereas, LODIC showed effectively the same properties as SIC, HODIC were found to be significantly different from SIC. The differences between LODIC and HODIC, were the effective individual fluence variation, different average individual fluence, different accumulated peak fluence and different spatial average individual fluence distribution and accumulated fluence distribution. The differences between LODIC and HODIC were found to vary with the pulse overlap.

For the analysis in this section, the understanding of the spatial emergence under SIC is applied. The spatial emergence can be described by an accumulated fluence domain, whose boundaries were found to vary due to incubation with the applied irradiation parameters. If the irradiation yields an accumulated fluence, within the accumulated fluence domain, LIPSS spatially emerge on the surface. In the following,

the difference between SIC and DIC for the accumulated fluence domain and consequently spatial emergence is discussed.

Incubation law

Based on the irradiation analysis, the LODIC were found, for certain individual fluence boundaries, to be effectively equal to SIC. In the following, a value for the individual fluence boundary is assumed, which limits the individual fluence to values that significantly vary thresholds. Under this condition, the spatial emergence at each displaced irradiation location under LODIC can be assumed to be equal to the spatial emergence under SIC. The spatial emergence under LODIC is assumed to yield individual circular LIPSS regions at each displaced irradiation location.

If the pulse overlap is increased, in order to obtain one homogeneous area of LIPSS, smaller fluences contribute to irradiated locations: compare Figures 2.4a and 2.4c, and the effective individual fluence is not constant anymore: compare Figure 2.7b. Under SIC, the accumulated fluence thresholds of different individual fluences were different, due to the incubation properties of the material, see Figure 3.2d. That means that, due to incubation, there is a difference between the application of different individual fluences.

If the different individual fluence values are subsequently applied to one spatial location, the accumulated fluence thresholds cannot be determined by the incubation law, compare Equations 3.3 to 3.6. These equations yield no quantitative information about the influence of different individual fluences per location on the accumulated fluence domain. This means, under HODIC, where individual fluence values contribute, which significantly vary the threshold, the incubation law cannot be applied to determine the domain location and size.

The incubation law presented by Jee [96] was developed in the eighties of the last century. Originally, the main purpose of the research was to improve the laser damage resistance of optical components and therefore to determine the threshold for the applicable laser power. There was no reason to deal with varying individual fluences, because the pulsed laser beam steered with a power, constant over a certain pulse period, to a constant location.

However, the analysis of DIC in this thesis, revealed the varying individual fluence per location. The influence, of a varying individual fluence on accumulated fluence domain boundaries, has not been considered before in literature. But, this effect is relevant for creating LIPSS areas and interesting for other applications, as for example the damage resistance of optical components. The movement of a beam reduces the average individual fluence and could therefore be a practical solution to increase the accumulated damage threshold of optical components.

Domain location

However, the knowledge about incubation gives qualitative information about the spatial emergence under HODIC. If the effective individual fluence is not constant, the domain location and size are determined by a sequence of different individual fluence values. The average individual fluence and individual peak fluence describe the distribution of the effective individual fluence under DIC. In the previous chapter

it was defined that in the origin of the Cartesian frame $\phi_p = \phi_0$. For the origin, ϕ_0 determines the domain location and size under LODIC.

With increasing φ and constant N_r smaller individual fluences contribute, which is why the effective average individual fluence decreases, compare Figure 2.7b. According to the knowledge about incubation in static, the application of pulses with smaller individual fluences requires more pulses, see Figure 3.2c, and accumulated fluence, see Figure 3.2d, to obtain the same phenomena on the surface. As a result the domain location will vary, compare Figure 3.3b. With increasing φ the domain location cannot decrease due to incubation, because no higher individual fluence values than ϕ_0 contribute. But, more pulses with smaller individual fluences must cause an increase of the domain location. This effect is in the following referred to as shift of the domain location.

But next to this shift, with increasing φ the accumulated peak fluence increases in the origin, as has been shown in Figure 2.6e. For otherwise constant irradiation and threshold parameters, this effect alone would lead to an accumulated fluence value above the upper domain boundary. But the decrease of the average individual fluence and the increase of the accumulated peak fluence occurs simultaneously with increasing φ . Therefore, the spatial emergence, with increasing φ , must depend on both effects and be determined by the ratio of domain location variation to accumulated fluence increase.

So far the effect of the varying individual fluence on the accumulated fluence domain was considered for the origin. For peripheral locations, which received under SIC or LODIC very small individual fluences, compared to the peak fluence, the average individual fluence is very small too, see Figures 2.4a and 2.4d. Therefore, the domain location must be higher, in terms of accumulated fluence, at these peripheral locations compared to the origin. If the pulse overlap increases, the accumulated fluence domain location decreases, while the applied accumulated fluence increases at these spatial locations.

For very high values of φ , the difference in individual and accumulated fluence between the origin and these peripheral locations vanishes, compare Figures 2.6 and 2.7. That means, although the individual fluence is not constant per location, the sequence is similar for different locations in the centre of the irradiated area. This causes the same domain location for different spatial locations. But next to the constant domain in space, under these conditions the accumulated fluence is constant in space too. The constant domain location and accumulated fluence cause the spatial emergence to be extended in this region, if the accumulated fluence requirements are satisfied.

This has further consequences, as the accumulated peak-to-valley difference fluence is very small under HODIC, see Figure 2.6f. Therefore, within the domain specific accumulated fluence values can be addressed individually. The addressing also includes the domain boundaries to other phenomena, above and below the LIPSS. But, under these conditions the formation and destruction of LIPSS can be expected progressively over a range of accumulated fluence and not at a specific value. The boundaries could appear as transition zones instead of sharp thresholds. If LIPSS emerge progressively, they may also have different spatial properties within the domain.

To conclude: Under DIC the incubation law was found to be invalid for a quantitative determination of the domain location. However, it can be applied to qualitatively discuss the spatial emergence under DIC. The spatial emergence with increasing pulse overlap, is determined by the ratio of domain location variation to accumulated fluence increase. Under these conditions, individual domain values can be addressed and the spatial emergence can be extended.

3.3.2 Parameter determination

Introduction

In Section 3.2 and Subsection 3.3.1 the spatial emergence of LIPSS has been investigated theoretically for SIC and DIC. This study has been performed in order to improve the experimental determination of irradiation parameters for extended surface areas covered with LIPSS. Therefore, the aim of this subsection is to derive a new approach for the parameter determination.

But before the irradiation parameters can be determined general input data for the structuring process are required, as, for example, structure periodicity, orientation and distribution. These input data are determined by the application. The structure periodicity determines the type of LIPSS, irradiation wavelength and accumulated fluence level, the structure orientation determines the polarisation and the structure distribution determines the spatial emergence irradiation parameters.

In Section 1.5 a purely theoretical approach for the determination of the spatial emergence irradiation parameters is mentioned, as alternative to the experimental. This approach is, on the one hand limited by the availability of accurate input data for engineering materials, which are required for the simulation of the laser-material interaction process. These input data are, for example, material data, which have been determined experimentally with specific analysis methods. On the other hand, this approach is limited by the current understanding of the LIPSS formation process. These issues may be solved in the future, but the aim of this research is to support the current application of LIPSS. That is why an experimental approach has been suggested.

Current approach

Currently, irradiation parameters are determined experimentally, usually direct under DIC, and no specific approach is mentioned in literature. For this purpose, dynamic irradiation experiments can be performed, similar to static. The input for DIC can be described by the three parameters ϕ_0 , φ and N_r . Within this discussion, φ for 2d-DIC is assumed to be equal for both directions. With respect to the static experimental matrix, compare Figure 3.1, the pulse overlap yields one additional degree of freedom for dynamic experiments. Figure 3.5 shows a schematic representation of a dynamic irradiation experiment, which is described in Chapter 5.

The input parameters are varied by the system parameters P_a , v and N_r . Under these conditions, the experimental matrix is spanned between the input parameters ϕ_0 , φ and N_r and yields a three dimensional space of DIC. Figure 3.5 is based on

Figure 3.5: Schematic representation of a dynamic irradiation experiment.

Figure 3.1 and the threshold results obtained under LODIC should correspond the results obtained under SIC. The determined irradiation parameters are specific for a type of LIPSS, spatial distribution and laser-material combination. That is why different applications require different values, which need to be determined and if necessary optimised with respect to machining time, spatial distribution and spatial properties. In the case of LIPSS, the effort to obtain these parameters is increased compared to ablated micro structures, because the dimensions of LIPSS are usually smaller and their emergence has a narrow domain.

In literature, where usually static experiments have been discussed, the spatial emergence has been described by the irradiation output parameters ϕ_{th} and N_p . Section 3.2, used the knowledge from literature as a starting point to continue the research and therefore the nomenclature has been further applied. However, the local individual fluence threshold ϕ_{th} and the edge length dependent parameter N_p are less suitable to describe the spatial emergence under DIC. Other output parameters,

such as ϕ_a , could be applied and also attributed to the emergence at a certain spatial location. Another possibility is to apply the input parameters ϕ_0 , φ and N_r .

In this case, the spatial emergence can be investigated for each parameter separately and for two dependencies respectively. If LIPSS spatially emerge, a threshold can be defined for each varied irradiation input parameter. However, also the accumulated peak fluence can be determined for each irradiation experiment and for each parameter two accumulated fluence thresholds can be determined. The thresholds determine a three dimensional accumulated fluence domain, which describes the spatial emergence under DIC. The window of irradiation parameters is obtained when $\Gamma(x, y)$ is limited to this accumulated fluence domain. The accumulated fluence allows to compare the results obtained under SIC and DIC by one quantity.

Effort comparison

Assuming the same beam conditions in a static and dynamic irradiation experiment, then the dynamic requires more experimental space and effort compared to the static. First of all, the irradiated area for each dynamic experiment is larger, due to the displacement. Secondly, if, for the irradiation parameters ϕ_0 and N_r the same boundaries and intervals as in the static experiment are applied, then additional space is required in dynamic for the parameter φ with a new boundary and interval.

Next to the experimental space, the handling of more irradiation parameters under DIC also requires more time for the dynamic experiments. On the one hand for preparation and execution and on the other hand for measurement and analysis of each irradiation experiment. Therefore, the static irradiation experiment yields, with a low effort, an overview about the available LIPSS structures. Beyond this, the static experiment allows in one experiment to quantify the beam radius in the sample plane, the domain boundaries and the incubation parameter.

Actually, in order to observe surface modifications, the interesting part of the irradiated area is the tail of the Gauss. The decreasing fluence and varying domain boundaries allow to observe multiple features in a small space. This can be observed in static and dynamic, but in static the apparent diameter can be used to measure the spatial emergence with the method of Liu [95]. In addition, the observation of surface modifications can be limited under HODIC, because the accumulation per repetition can be so high that a domain can be passed over.

But, the static experiment has also some disadvantages which should be considered. If the fluence gradient is too strong, a certain type of LIPSS will not appear on the surface, as discussed in Subsection 3.2.4. Furthermore, if the spot is small, then phenomena which appear statistically, as LIPSS initiated by surface defects or material inhomogeneities may not appear. The same holds for features which need some space to grow, as surface plasmon polariton initiated LIPSS. But, for most applications and the work in this thesis, regular types of LIPSS are relevant.

New approach

The determination of dynamic irradiation parameters could be improved by information about LIPSS types and their accumulated fluence domains from static experiments. Assuming a laser-material combination is given without further information

about LIPSS structures and domain boundaries, then a static irradiation experiment can be performed, which yields the available LIPSS structures and other types of surface modifications. Once a type of LIPSS is selected, values for the domain boundaries can be determined.

The domain boundaries can be derived from a single SEM image, when ϕ_0 and ω_0 are known. Another approach is to perform a D²-Experiment for a specific N_r . A third opportunity is to analyse the domain boundaries for different N_r , which gives the full domain and incubation factor. As a result of the static experiment a set of data $\Gamma_{th(i)}(N_p)$ and $\Delta\Gamma_{th(i)}(N_p)$ or $\Gamma_{th(i)}(\phi)$ and $\Delta\Gamma_{th(i)}(\phi)$ or $\Gamma_{th(i)}(1)$, $\Delta\Gamma_{th(i)}(1)$ or the incubation factor ξ is obtained, depending on the chosen static experiment. These data can be determined with the equipment, which is also applied for laser micro machining.

The static experiment represents a compromise between the above mentioned theoretical and the current experimental approach. This experiment yields quantitative data about the spatial emergence, which have been determined with a minimal effort. If the irradiation parameters are required for an area with an inhomogeneous distribution of LIPSS, the static information can be directly applied to derive irradiation parameters for LODIC. For HODIC this knowledge can be applied to narrow the range of irradiation parameters, because the domain boundaries enable an estimate of the required accumulated fluence.

3.3.3 Domain access and addressing

In the following two subsections, it is discussed how the domain boundaries can be applied to derive values for irradiation parameters. This subsection deals with the domain access through the accumulated peak fluence and the addressing of specific domain values. This is a preparation for Subsection 3.3.4, which deals with the derivation of approximated solutions for irradiation parameters. The starting point for the access to the domain is Equation 2.21. To solve this equation, simplifications are required. First of all, only the final accumulated fluence is required to obtain LIPSS on the surface. Furthermore, the distribution of the accumulated fluence on the different parts of the kinematic system is required, therefore n is divided in individual components. This transforms Equation 2.21 into

$$\Gamma(x, y) = N_r \sum_g \sum_k \phi_0 \exp \left(-2 \left(\frac{(x - x_k)^2 + (y - y_g)^2}{\omega_0^2} \right) \right), \quad (3.22)$$

where k and g denote integers for the number of displacements, the negative and positive boundaries of which can be calculated by Equation 2.27. The third summation for the number of repetitions is a multiplication because here the irradiation process is assumed to be constant for each repetition. Equation 3.22 cannot satisfy the domain boundaries at each location. But Equation 3.22 can be solved for Γ_p as a measure for the accumulated fluence. Next to this the displacement can be summarised by the

pulse overlap. These steps yield

$$\Gamma_p = N_r \sum_g \sum_k \phi_0 \exp \left(-2 \left(\frac{(-2\omega_0 (1 - \varphi) k)^2 + (-2\omega_0 (1 - \varphi) g)^2}{\omega_0^2} \right) \right). \quad (3.23)$$

The length of the area and the displacement intervals in x and y direction are equal. Together with the analysis location in the origin and the assumption regarding the number of displacements, a symmetrical kinematical system is obtained. Therefore, this equation can be further simplified, which gives

$$\Gamma_p = N_r \phi_0 \left(\sum_k \exp \left(-8 ((1 - \varphi) k)^2 \right) \right)^2 = N_r \phi_0 \tilde{\Gamma}_p (\varphi). \quad (3.24)$$

This equation describes the relation between the output parameter Γ_p and the three input parameters ϕ_0 , φ and N_r . Values for the relative accumulation $\tilde{\Gamma}_p (\varphi)$ can be obtained either by the simplified form presented in Equation 3.24 or by numerical simulations according to those presented in Chapter 2. The simplified form can be calculated with an effective pulse number, if for example an area of length $l = 6\omega_0$ considered. This is also discussed in Subsection 2.3.3 and simplifies the calculation.

The domain boundaries describe an amount of accumulated fluence too and can be applied to solve Equation 3.24 under certain conditions. If the domain boundaries would be addressed by Γ_p , the deposited accumulated fluence may not result in the desired spatial emergence. But, the domain consists of a range of accumulated fluence, where the j 'th accumulated fluence value $\Gamma_{th(i,j)}$ within the i 'th domain, can be addressed to satisfy the spatial emergence.

The basic idea behind this approach is presented in Figure 3.6. This diagram illustrates, the accumulated fluence deposition, denoted on the right side, and the boundaries of the surface modifications, denoted on the left side. The accumulated fluence thresholds of different surface modifications are indicated with $\Gamma_{th1-4} (\phi)$ as continuous lines. As example, the bandwidth of the domain is indicated with $\Delta \Gamma_{th(2)} (\phi)$ and the j 'th accumulated fluence value within this domain is indicated with $\Gamma_{2,j} (\phi)$ as dashed line, specifically for threshold number 2.

The window for irradiation parameters can be limited in three steps, first the selection of a type of LIPSS, second the selection of a domain location and third the selection of a domain value. A starting point is to address a central domain location and value. Further aspects related to the selection, are irradiation errors, input uncertainties, the machining time and accessibility. The latter means that, a lower domain location requires less accumulation and therefore less machining time, but it might not be accessible because an homogeneous area requires a minimum pulse overlap and therefore a certain accumulation.

The domain location and value can be determined either for the ϕ or N_p dependence. Values for ϕ and N_p are selected, based on the microscopic analysis of the static experiment. If a value for ϕ is selected, the determination of the domain boundaries

Figure 3.6: Concept of the domain access and addressing under DIC.

$\Gamma_{th(i)}(\phi)$ and $\Gamma_{th(i+1)}(\phi)$ yield an accumulated fluence value

$$\Gamma_{th(i,j)}(\phi) = \Gamma_{th(i)}(\phi) + \frac{\Delta\Gamma_{th(i)}(\phi)}{\rho}\sigma \quad (3.25)$$

and a value for the number of pulses

$$N_{th(i,j)}(\phi) = \frac{\Gamma_{th(i,j)}(\phi)}{\phi}. \quad (3.26)$$

The factors ρ and σ yield values of equal step size within the domain. For example, the centre of the domain is obtained if $\rho = 10$ and $\sigma = 5$. If a value for N_p is selected, the determination of the domain boundaries $\Gamma_{th(i)}(N_p)$ and $\Gamma_{th(i+1)}(N_p)$ yield an accumulated fluence value

$$\Gamma_{th(i,j)}(N_p) = \Gamma_{th(i)}(N_p) + \frac{\Delta\Gamma_{th(i)}(N_p)}{\rho}\sigma \quad (3.27)$$

and an individual fluence value

$$\phi_{th(i,j)}(N_p) = \frac{\Gamma_{th(i,j)}(N_p)}{N_p}. \quad (3.28)$$

The N_p dependent domain boundaries are easier to find and are more accurate, because they can be determined from a single microscopic image, without additional information about the incubation parameter and single pulse thresholds. Whereas for the ϕ dependent domain boundaries, the corresponding domain values need to be

determined experimentally, see the discussions in Subsections 3.2.3 and 3.2.4. Furthermore, the N_p domain boundaries, usually show sharper transitions.

A value for $\Gamma_{th(i,j)}$ can be determined without prior knowledge on the incubation factor for a certain combination of ϕ and N_p . If more information is supplied by the scope of the experiment, different values can be calculated. Each addressed value can be described by the ϕ

$$\Gamma_{th(i,j)}(\phi) = \phi \left(\frac{\phi}{\Gamma_{th(i)}(1) + \frac{\Delta\Gamma_{th(i)}(1)}{\rho}\sigma} \right)^{\frac{1}{\xi-1}} \quad (3.29)$$

or N_p dependence

$$\Gamma_{th(i,j)}(N_p) = \left(\Gamma_{th(i)}(1) + \frac{\Delta\Gamma_{th(i)}(1)}{\rho}\sigma \right) N_p^\xi. \quad (3.30)$$

However, also the corresponding pulse number or individual fluence dependent domain boundaries can be calculated, either with Equations 3.4 and 3.8 if $N_p = N_{th(i,j)}(\phi)$ or with Equations 3.6 and 3.9 if $\phi = \phi_{th(i,j)}(N_p)$. Next to this, the corresponding centre location can be calculated. But, similar to the different domain sizes, the N_p and ϕ dependent centre locations might differ for the same reason as discussed in Section 3.2 for the domain size. If one value is centred, e.g. the N_p dependent value, the other, in this case the ϕ dependent value, is decentred. Since the N_p dependent domain is usually smaller, small changes in ϕ are more sensitive and to centre this one might be preferable.

3.3.4 Derivation of approximations

The aim of this subsection is to derive analytical approximations for irradiation parameters to create extended areas of LIPSS. These approximations use static information about accumulated fluence domain boundaries as input in order to derive values for the laser system parameters P_a , v and N_r . The domain boundaries can be applied to determine different sets of irradiation parameters.

In this subsection it is assumed that two different areas are required, one with an inhomogeneous and another with a homogeneous distribution of LIPSS. For the inhomogeneous area, LIPSS emerge on individual circular areas at each displaced irradiation location. For the homogeneous area, LIPSS emerge on a rectangular area with a homogeneous distribution. The inhomogeneous area is the result of the LODIC condition, according to the irradiation shown in Figure 2.4d, and the homogeneous is obtained with HODIC, according to the irradiation shown in Figure 2.4f.

In Subsection 3.3.1 the validity of the incubation law has been discussed for DIC and was found to be valid for LODIC but invalid for HODIC related to the non-constant individual fluence. This means the static information can be applied to derive exact solutions for irradiation parameters which result in LODIC. For HODIC this information can only be applied to derive approximations, which narrow the range of irradiation parameters.

The exact boundary between these regimes depends on the contribution of a certain individual fluence to an accumulated fluence threshold and cannot be derived from the incubation law, presented in Subsection 3.2.2. Therefore, the solutions obtained for DIC are denoted as approximations, which give accurate values for LODIC and less accurate values for HODIC. In the following, the approximations for the inhomogeneous LIPSS area under LODIC are derived first and the domain boundaries are assumed to be constant for different values of φ in this regime.

In the previous subsections, it has been found that LIPSS spatially emerge, if the accumulated fluence is within a certain domain, whose location and size vary due to incubation with the applied irradiation parameters. This understanding determines how in the following the irradiation parameters are applied to control the spatial emergence. If the individual fluence dependence is followed, the peak fluence can be applied to reach a certain domain location, the number of repetitions to reach a certain domain value and the pulse overlap as a free parameter which controls the lateral distribution of the spatial emergences.

Based on this understanding, two conditions need to be formulated to obtain a specific spatial emergence. The first condition concerns the peak fluence, which has to yield a specific domain location

$$\phi_{th(i,j)}(N_p) \approx \phi \approx \phi_0, \quad (3.31)$$

and the second condition concerns the accumulated peak fluence, which has to yield the selected domain value

$$\Gamma_{th(i,j)}(N_p) \approx \Gamma_{th(i,j)}(\phi) \approx \Gamma_p. \quad (3.32)$$

The approximations can be derived mathematically for the ϕ and N_p dependence. Both dependencies provide useful information. Substituting Equation 3.24 and Condition 3.31 in Condition 3.32 and rewriting in a more general form using Equations 3.29 and 3.30 gives

$$\phi \left(\frac{\phi}{\Gamma_{th(i)}(1) + \frac{\Delta\Gamma_{th(i)}(1)}{\rho}\sigma} \right)^{\frac{1}{\xi-1}} \approx N_r \phi \tilde{\Gamma}_p(\varphi) \quad (3.33)$$

and

$$\left(\Gamma_{th(i)}(1) + \frac{\Delta\Gamma_{th(i)}(1)}{\rho}\sigma \right) N_p^\xi \approx N_r \left(\Gamma_{th(i)}(1) + \frac{\Delta\Gamma_{th(i)}(1)}{\rho}\sigma \right) N_p^{\xi-1} \tilde{\Gamma}_p(\varphi). \quad (3.34)$$

Substituting Equation 2.22 and Condition 3.31 in Equation 2.25 and solving this equation for the average power gives

$$P_a \approx \phi \frac{1}{2} \pi \omega_0^2 f_r \quad (3.35)$$

and

$$P_a \approx \left(\Gamma_{th(i)}(1) + \frac{\Delta\Gamma_{th(i)}(1)}{\rho}\sigma \right) N_p^{\xi-1} \frac{1}{2} \pi \omega_0^2 f_r. \quad (3.36)$$

In Approximation 3.35, the applied average power determines the domain location belonging to ϕ , which has been selected based on the static irradiation experiment. However, Approximation 3.36 gives the possibility to calculate, in addition to the experimentally determined values, other theoretical values. Furthermore, this approximation also identifies limitations, with respect to obtain a certain modification on the surface. Since the peak fluence has to fulfil $\phi_0 \leq \Gamma_{th(i)}(1) + \Delta\Gamma_{th(i)}(1)$, the selection of specific values for the average power is not applicable. Solving Approximations 3.33 and 3.34 for the number of repetitions N_r gives

$$N_r \approx \frac{N_{th(i,j)}(\phi)}{\tilde{\Gamma}_p(\varphi)} \approx \frac{\left(\frac{\phi}{\Gamma_{th(i)}(1) + \frac{\Delta\Gamma_{th(i)}(1)}{\rho}\sigma} \right)^{\frac{1}{\xi-1}}}{\left(\sum_k \exp\left(-8((1-\varphi)k)^2\right) \right)^2} \quad (3.37)$$

and

$$N_r \approx \frac{N_p}{\tilde{\Gamma}_p(\varphi)} \approx N_p \left(\sum_k \exp\left(-8((1-\varphi)k)^2\right) \right)^{-2}. \quad (3.38)$$

The quadratic term on the right side of Approximation 3.38 describes a function defined in an earlier paper as the relative accumulation factor Ψ [166]. This function is the inverse of $\tilde{\Gamma}_p$, shown in Figure 2.6e. Specific values, which are frequently applied in practice, have been presented in Table 2.3. If LIPSS have been observed at a certain N_p and the irradiation power is simply centred between the domain boundaries, a starting point for N_r is obtained with the reduction of N_p by the relative accumulation. The simple form, makes Approximation 3.38 an easy-to-use tool in practice. Whereas Approximation 3.37 allows to address different values within the domain.

Under LODIC, these approximations actually yield accurate values for displaced circular areas of LIPSS and the component $\tilde{\Gamma}_p$ is approximately 1 for different values of the pulse overlap. Approximations 3.36 and 3.37 describe a range of values, which can be applied to obtain LIPSS on the surface. The apparent diameter of an individual circular LIPSS area is determined by the beam radius, the addressed domain value and the pulse number dependent domain size.

The solution for an inhomogeneous area of LIPSS under LODIC yields a starting point to obtain a homogeneous area of LIPSS under HODIC. As discussed in Subsection 3.3.1, with increasing φ the individual fluence is not constant and fluences smaller than ϕ_0 also contribute. Therefore, the domain boundaries vary and the accumulated fluence increases. As a first approximation, the boundaries can be assumed to be constant and only the increasing $\tilde{\Gamma}_p$ can be considered. Using this assumption solutions are already given by Approximations 3.37 and 3.38.

Another approach is to evaluate each individual fluence contribution by its relative threshold contribution per pulse c_p . From the incubation law, the following relation can be derived

$$1 = \frac{\phi_{th(i,j)}(N_p) N_p}{\Gamma_{th(i,j)}(1) N_p^\xi} = \sum_{n=1}^{N_p} c_p. \quad (3.39)$$

Within a train of pulses, each individual laser pulse induces a certain physical change of the material according to the received fluence. The physical change of the material can be different, as for example stress, deformation or ablation. Since, the previous pulse has already modified the material, the interaction of the following pulse with the, by then, modified material might be different and yield a different individual material change. This might yield a non-linear laser-material interaction and material change process, over the complete pulse train. Whatever this non-linear relationship is, the spatial emergence of a certain type of surface modification requires a certain accumulated material change and the incubation law describes phenomenologically the non-linear variation of the accumulated fluence threshold.

The relative individual pulse contribution is by the incubation law related to a certain individual fluence. The relative pulse contribution is equal to the average relative individual material change and therefore represents information about the relative strength of the material change per pulse. For very high pulse numbers, the average relative individual material change might be close to the individual material change. Although the incubation law cannot be applied to determine the accumulated fluence thresholds for a sequence of pulses with different individual fluences, for high pulse numbers this law might yield a process and material related estimate for each individual contribution and thereby for the required repetitions. Under the assumption, that for high pulse numbers the relative pulse contribution yields an indication for the relative individual material change, the following approximation can be established

$$1 \approx \sum_{n=1}^{N_p} c_p(\phi) \approx \sum_{n=1}^{N_p} \left(\frac{\phi(n)}{\Gamma_{th(i,j)}(1)} \right)^{\frac{1}{1-\xi}}. \quad (3.40)$$

This approximation basically assumes a linear material change process and calculates for each individual fluence a relative contribution, whose summation yields 1 if the surface modification emerges. The relative contribution of a certain individual fluence depends on the single pulse threshold of a certain modification and the incubation parameter of the material. This can be applied to obtain an approximation for the weighting of each individual fluence contribution

$$N_r \approx \left(\sum_g \sum_k \left(\frac{\phi_0 \exp \left(-8 \left(((1-\varphi)k)^2 + ((1-\varphi)g)^2 \right) \right)}{\Gamma_{th(i,j)}(1)} \right)^{\frac{1}{1-\xi}} \right)^{-1}, \quad (3.41)$$

in a dynamic situation. Approximations 3.37 and 3.38 consider the influence of different power values due to incubation and the influence of fluence accumulation due to different pulse overlaps. The influence of the varying individual fluence is considered by accumulating each individual fluence with its Gaussian contribution to a peak fluence determined domain value. Whereas Equation 3.41 considered the varying individual fluence by accumulating each individual fluence with its Gaussian and incubation contribution to a contribution determined domain value.

So far a certain domain location and value has been addressed. The parameter which is left, is the pulse overlap, defined as a free parameter controlling the distribu-

tion of LIPSS on the surface, while the addressed domain value is kept constant by compensating with N_r . The selected value φ determines the displacement velocity by

$$v = 2\omega_0 (1 - \varphi) f_r. \quad (3.42)$$

However, if the beam conditions or the pulse frequency are varied, the pulse overlap varies too. Variations in φ , influence $\Gamma_p(\varphi)$, $\Gamma_d(\varphi)$ and the domain location and need to be compensated by N_r . The increased accumulated fluence could also be reduced by the peak fluence, but this would vary, due to incubation, the location of the domain at the same time. In order to merge the individual LIPSS regions, an increase of φ , from LODIC to HODIC, above a critical value φ_c is required. This value is reached, if Γ_v exceeds $\Gamma_{th}(\phi)$ at the valley location.

A simple way to obtain a value above φ_c is based on the irradiation analysis. If the pulse overlap is such that at the valley location next to the origin $\Gamma_v \approx \Gamma_p$, then a sufficient small accumulated peak-to-valley difference fluence value Γ_d is obtained. The pulse overlap analysis shows that already at $\varphi = 0.65$ the values for $\tilde{\Gamma}_d \approx 10^{-3}$. For pulse overlaps larger than this value, an approximately equal accumulated fluence is achieved in the centre of the irradiated area.

However, if a certain value is addressed within the domain, this value will have a certain distance, in terms of accumulated fluence, from the lower domain boundary. Therefore, the pulse overlap could be larger in practice. The parameter Γ_d could be compared to the domain sizes $(\Delta\Gamma_{th(i)}(N_p)/\rho)\sigma$ and $(\Delta\Gamma_{th(i)}(\phi)/\rho)\sigma$. But, these are static values and under HODIC the domain location shifts and the difference of the individual and accumulated fluence between different locations increases. Next to this, practical issues influence the selection of φ , as for example the required spread for a regular emergence and irradiation errors, see Chapter 4.

Discussion

In this chapter it is assumed, that the spatial emergence is analysed after the irradiation has been applied. Therefore, the spatial emergence is described by the fluence and not by the intensity. But, the intensity, and inherently the pulse duration, determines whether the material interacts with the radiation and a certain modification will be created on the surface. Furthermore, the time between pulses has been assumed to be large, so that transient phenomena, as plasma or heat accumulation, do not affect the beam propagation and material interaction of subsequent pulses. This allowed to work on a pulse-to-pulse basis and to apply the knowledge about incubation to study the spatial emergence mechanism. Nevertheless, this theory is limited with respect to the behaviour for other wavelengths, pulse durations or pulse frequencies, which can significantly vary the thresholds. A practical solution is to repeat static experiments for specific parameters, in order to determine morphologies and thresholds.

The predicted irradiation input parameters are approximations, based on the knowledge about incubation. But Jee's incubation law has been determined using a constant individual fluence. The effect of fluence variation on incubation has not been considered before in literature. The analysis of the fluence accumulation process shows that this point is relevant for surface structuring with overlapping pulses. Bearing in mind that Jee's incubation law may not be accurate at high values of N_p

and may not be valid in case of varying fluence, it has been used in this work to obtain approximations, which yield an indication for irradiation parameters under DIC. To find the range of validity and the boundaries of these approximations substantial experimental verification is required.

In the context of damage experiments, the meaning of incubation was seen in a varying laser damage threshold, caused by a varying number of applied laser pulses. For machining LIPSS areas, incubation has not been considered before. However, it has been shown that incubation plays an important role for the selection of irradiation parameters. The incubation law does not only formulate a relation between Γ and ϕ , it also relates the irradiation input parameters P_a and N_r . It is important to note, that these parameters are inversely related. Even if quantitative data for these parameters are missing, the approach gives important qualitative information. First, if the power is varied, the number of repetitions needs to be varied too, due to incubation. Second, it yields a direction, if the power is increased, the number of repetitions needs to be decreased, for the same reason. Furthermore, the approximations show the interrelation of irradiation and threshold parameters. This explains the difficulty to determine irradiation parameters under dynamic conditions.

The problem described in the scope of research for this thesis have been derived from the state of the art presented in Chapter 1. Later on, the research has been started and the theoretical part of this thesis has been developed. Parallel to the research for this thesis Li et al. worked on the same problem [167]. Hereinafter, this work is used for a comparison with the results which have been independently obtained in this thesis. The model of Li analyses the accumulation of fluence and points out the importance of accumulation of fluence for machining LIPSS areas. In particular, this model addresses the accumulated fluence distribution and the increasing accumulated peak fluence with increasing pulse overlap. In order to come up with a homogeneous area of LIPSS, it suggests to apply a fixed velocity, derived from the beam radius, pulse frequency and certain constant factors, which yields a uniform accumulated fluence distribution. Based on this velocity a relative peak accumulation is obtained and by comparing this value to the static accumulated fluence threshold a value for the peak fluence is obtained.

Thereby, the research results of Li confirm some of the results obtained in this thesis, such as that the selected velocity should yield a uniform accumulated fluence distribution, in order to obtain a homogeneous area of LIPSS, and that the accumulated peak fluence should be compared with static accumulated fluence thresholds. However, the irradiation analysis presented in reference [167] lacks on details regarding the irradiation parameters and accumulation process. Although Li notices a specific threshold for LIPSS and their disappearance at higher fluences, he does not come up with the accumulated fluence domain theory. Next to this, the influence of incubation on the accumulated fluence thresholds and irradiation input parameters is neglected. Furthermore, the peak fluence is not related to a threshold. Moreover, this approach does not explain the application of different irradiation repetitions.

As mentioned in the introduction, Huang et al. [159] suggested that increasing the overlap above the critical value for a homogeneous area of LIPSS may lead to the disappearance of LIPSS due to the observed "over-hatch" effect. This effect has also

been observed by Li, who suggested to apply a fixed critical velocity. Combining the theoretical results of Chapters 2 and 3, it is now possible to understand the "over-hatch" effect. This effect is caused by a larger increase of the applied accumulated fluence compared to the variation of the domain location, resulting in a value above the upper domain boundary. However, it seems possible to apply velocities below the mentioned critical value. If ϕ_0 is lowered, a higher accumulation is required. If ϕ_0 is even further decreased, different values of N_r can be applied. Next, the presented approximations also identify limitations for the selection of specific parameter combinations. Since ϕ_0 has to fulfil $\phi_0 \leq \Gamma_{th}(1) + \Delta\Gamma_{th(i)}(1)$, only certain combinations of ϕ_0 and $\tilde{\Gamma}_p$ are applicable. Moreover if a certain overlap is required, for a uniform area, the requirements for Γ_p and Γ_d may not be satisfiable at the same time.

This research focuses on a specific type of surface modification, namely LIPSS. However, also other types of surface modifications emerge in zones, whose thresholds are determined by incubation properties. The approach presented here might also be applicable to other surface modifications. With respect to the fluence modification diagram, ablation is usually observed as the upper modification in terms of fluence, emerging in the centre of an irradiated location. For ablation an upper boundary has not been described in literature, because ablation does not disappear on the surface for higher fluences. However, also the morphology of an ablation crater changes with increasing accumulated fluence, so that different regimes, which bear upper thresholds, can be identified. But if the crater morphology and ablation process vary strong with different pulse overlaps, other aspects, such as material recast and surface slopes, can influence the accumulated fluence thresholds. Therefore, the approach presented here might be limited to a certain ablation depth.

3.4 Conclusions

In this theoretical chapter, the phenomenological relation between certain irradiation parameters and the spatial emergence of LIPSS starting from the knowledge about incubation has been investigated for SIC and DIC. The spatial emergence of LIPSS is determined by the intersection of the applied accumulated fluence with the accumulated fluence domain boundaries. The location and size of the accumulated fluence domain vary, due to incubation, with the applied irradiation parameters. The annular zone usually observed under SIC, corresponds to the pulse number dependent fluence domain. Due to the varying individual fluence, the incubation law presented by Jee cannot be applied to obtain accurate quantitative information about the domain location under DIC. However, it can be applied to qualitatively discuss the spatial emergence under DIC. The spatial emergence with increasing pulse overlap, is determined by the ratio of domain location variation to accumulated fluence increase.

Based on the study of the spatial emergence of LIPSS under SIC and DIC, a new approach for the determination of irradiation parameters has been derived. The approach consists of two steps, first a static irradiation experiment, to determine the domain boundaries of a specific type of LIPSS, and second an estimate of irradiation parameters, using analytical approximations for DIC. The static experiment yields

information about the available LIPSS types and their domain boundaries. These are quantitative input data, which can be applied for the approximation of irradiation parameters under DIC. They can be determined with laser micro machining equipment and a minimal experimental effort. If the irradiation parameters are required for different types of LIPSS areas the static information can be directly applied to derive irradiation parameters for LODIC. For HODIC this knowledge can be applied to limit the range of irradiation parameters.

The experimental determination of irradiation parameters is influenced by the interrelations between input parameters and the non-linear relations between input-output parameters. Without the simulation of the irradiation process, no quantitative information about the actually applied irradiation is available. Therefore, numerical simulations are important for the experimental practice. Next, the simulations provide an understanding of the irradiation process. Together with the knowledge about incubation dependent accumulated fluence thresholds, this can be used to understand the spatial emergence of LIPSS under lateral displacement irradiation conditions.

This analysis yields interesting information for further research. The influence, of a varying individual fluence on accumulated fluence domain boundaries has not been considered before in literature. But, this effect is relevant for creating LIPSS areas and interesting for other applications, as for example the damage resistance of optical components. The movement of a beam reduces the average individual fluence and could therefore be a practical solution to increase the accumulated damage threshold of optical components.

Chapter 4

Irradiation errors

4.1 Introduction

So far the spatial emergence of LIPSS has been considered under optimal conditions. However, the implementation as an industrial manufacturing process can be affected by technical limitations, which can cause a deviation of the actually applied irradiation from the optimal. These deviations are referred to as irradiation errors. The aim of this chapter is to investigate the influence of technical limitations on the irradiation process. The mechanism and the effect of irradiation errors have not been addressed in literature. The analysis in this chapter is based on the irradiation model presented in Chapter 2. In this chapter, the irradiation model has been extended to include errors.

4.2 Model supplement

4.2.1 Physical assumptions

In this section, a set of fundamental assumptions for the irradiation process is described, which complement the assumptions of Section 2.2. Errors in the irradiation process can originate from various sources, as the environment, operator, laser system and their components. For this chapter, six technical errors have been selected, two beam, two position and two alignment errors. This selection is meant to be exemplary rather than to be complete. For the selection of these errors, their practical relevance and impact on the structuring process were considered.

In the primary assumptions for the irradiation model the spatial Gaussian distribution was assumed to be circular symmetric. In this chapter the distribution is assumed to have an elliptical deformation. This type of laser beam deviation can be caused by optical elements and alignment errors in the laser system. The transversal ellipticity is characterised by different beam radii. The short and the long axes of the ellipse are defined along the coordinate axes. The radii in x and y direction are

Figure 4.1: Schematic drawing illustrating the subaperture stitching kinematics, applied for the extended irradiation model.

denoted by ω_x and ω_y . Furthermore, the average power is assumed to vary over long and short time periods. The cause for this deviation can be found in electrical or ambient disturbances. The amplitudes of the relative pulse-to-pulse variations are denoted by $\tilde{P}_{e(1)}$ and $\tilde{P}_{e(2)}$.

In the irradiation model, irradiated locations were described by a sequence of lateral displacements. In this chapter, the addressed locations are additionally assumed to vary due to position errors. These errors can be caused by ambient disturbances, leading to oscillations of the mechanical systems, or by synchronisation issues, between the laser and the kinematic system. Two types of positioning errors are assumed, first a statistical variation of the irradiated locations and second a phase shift between irradiated lines. The amplitudes of the statistical variations are denoted by x_{e0} and y_{e0} . The phase shift is introduced for each new line in the x direction and their amplitude is denoted by $x_{\Phi 0}$.

Previously the irradiation plane was assumed to be located in the origin, in this chapter also other planes out of the focus are analysed. This is because for an irradiation experiment the sample surface needs to be aligned with respect to the focus location. As a result of this alignment, a certain axial error remains. The alignment

error is denoted by the axial offset z_0 . So far an irradiated area of edge lengths l_x and l_y was assumed, as depicted in Figure 2.1. In this chapter this length is assumed to be insufficient for covering larger areas, so that a second kinematic system with larger displacement capacities is required. Therefore, subapertures are displaced in order to mechanically increase the irradiated area to the lengths l'_x and l'_y . The assumptions for these kinematics are similar to the first. Figure 4.1 shows a schematic drawing for the subaperture stitching kinematics.

4.2.2 Numerical implementation

For the numerical simulations in this chapter, the computer program described in Section 2.3 has been extended. In the following changes to equations in the basic program are described. The direction dependent changes are described for the x direction only. The transversal deformation of the laser beam is described by two different ellipticity factors, ε_x for the x and ε_y for the y direction. These factors compress or stretch the focused beam along the coordinate axes x and y . This yields

$$\omega_{0x} = \omega_0 (1 + \varepsilon_x) \quad (4.1)$$

for the beam radius in the focal plane along the x direction. The beam propagation along z direction follows a caustic. Due to this caustic the beam radius is different for each location in z direction [161, 164], which can be described by

$$\omega_x(z) = \omega_{0x} \sqrt{1 + \left(\frac{z}{z_{Rx}}\right)^2}. \quad (4.2)$$

If the sample surface is placed out of the focal plane at a defocused location $z = z_0$ the beam radius varies. At a distance $z = z_{Rx}$ the beam radius is expanded by a factor $\sqrt{2}$. The parameter z_{Rx} is also referred to as Rayleigh length [161, 164] and given by

$$z_{Rx} = \frac{\pi \omega_{0x}^2}{\lambda}. \quad (4.3)$$

This alignment and beam error vary, for otherwise constant irradiation parameters, the peak fluence a priori. However, due to the linear relation between the average power and peak fluence, the latter is also varying during the irradiation process on a pulse-to-pulse basis. Within this analysis, two types of power variations have been assumed. A long term drift $P_{(1)}(n)$, whose relative variation is simulated by a sinus function

$$\tilde{P}_{(1)}(n) = \tilde{P}_{e(1)} \sin(2\pi f_e n + \pi f_e), \quad (4.4)$$

which follows a certain error frequency. The second type is a short term drift $P_{(2)}(n)$, which is simulated by a pseudorandom function $\Omega_p \in \{0 : 1\}$ creating relative variations

$$\tilde{P}_{(2)}(n) = \tilde{P}_{e(2)} (2\Omega_p(n) - 1), \quad (4.5)$$

which follow the laser pulse frequency. Both contributions vary the average power and yield the pulse number dependent power

$$P(n) = P_a \left(1 + \tilde{P}_{(1)}(n) + \tilde{P}_{(2)}(n) \right). \quad (4.6)$$

These alignment and beam errors, yield the z location and pulse number dependent peak fluence

$$\phi_0(z, n) = \frac{2P(n)}{\omega_x(z) \omega_y(z) \pi f_r}. \quad (4.7)$$

The different transversal elongations change, for constant absolute displacements, the overlap between pulses for each direction differently. In the x direction the overlap is given by

$$\varphi_x = 1 - v / (f_r 2\omega_{0x}). \quad (4.8)$$

The direction dependent overlap also has an, although very small, influence on the number of displacements

$$N_x = \frac{l_x - 2\omega_{0x}}{2x_0}. \quad (4.9)$$

Another set of changes in the program is related to the second kinematic system. In order to increase the irradiated surface area, sub-apertures are displaced and stitched together. During the stitching displacement, no irradiation takes place. The displacement has the aim to continue the irradiation applied within the subapertures. In order to continue the irradiation homogeneously, an overlap or gap of subapertures needs to be avoided. Therefore, the displacement of subapertures requires a displacement boundary. As a starting point, the stitching boundary is assumed to be $x'_0 = l_x$. In this case, the number of stitching displacements can be calculated per direction and half site by the following equation

$$N'_x = \frac{l'_x - l_x}{2x'_0} = \frac{1}{2} \left(\frac{l'_x}{l_x} - 1 \right) \quad (4.10)$$

and depends on the ratio between the length of the irradiated area l'_x and subaperture l_x . This also requires to adapt the starting location of the irradiation

$$\vec{\psi}_d(1) = [(-x_0 N_x) + (-x'_0 N'_x), (-y_0 N_y) + (-y'_0 N'_y)]^T, \quad (4.11)$$

to the new size of the irradiated area. If the area is extended, for otherwise constant parameters, the number of pulses increases and additional kinematic events need to be executed after a certain number of pulses. The pulse boundaries, introduced in Subsection 2.3.2, for the additional kinematic events are given by

$$N_a = 1 \quad (4.12)$$

$$N_b = (2N_x + 1) \quad (4.13)$$

$$N_c = (2N_x + 1)(2N_y + 1) \quad (4.14)$$

$$N_d = ((2N_x + 1)(2N_y + 1))(2N'_x + 1) \quad (4.15)$$

$$N_e = ((2N_x + 1)(2N_y + 1))(2N'_x + 1)(2N'_y + 1) \quad (4.16)$$

$$N_f = ((2N_x + 1)(2N_y + 1))(2N'_x + 1)(2N'_y + 1) N_r = N_p. \quad (4.17)$$

The pulse boundaries need to be updated outside and inside the main loop for the irradiation process. Finally, the two position errors need to be implemented. The statistical variations of the irradiation locations are numerically generated in a similar way as the statistical power error, using a pseudo random value generator $\Omega_d \in \{0 : 1\}$

$$x_e(n) = x_{e0}(2\Omega_d(n) - 1). \quad (4.18)$$

This is the same for the first and second kinematic systems, where the latter is denoted by $x'_e(n)$. The phase shift between irradiated lines is introduced by a sinusoidal expression

$$\tilde{x}_\Phi(n) = \sin\left(\frac{2\pi}{\Phi(n)}n\right), \quad (4.19)$$

whose pulse difference yields a relative displacement factor per line. The parameter $\Phi(n)$ denotes the line period. The absolute phase shift in x direction is obtained by

$$x_\Phi(n) = x_{\Phi 0}(\tilde{x}_\Phi(n) - \tilde{x}_\Phi(n-1)). \quad (4.20)$$

The basic calculation of the actual position $\vec{\psi}_d(n)$ is described in Subsection 2.3.2 and needs to be extended by the position errors and stitching displacement. The implementation must ensure that the statistical position errors do not accumulate, the phase shift is introduced only in the x direction and the stitching displacement includes the direction and axes switches and the statistical position errors. Based on these changes the actual position is then calculated by

$$\vec{\psi}_d(n) = [x_n, y_n]^T = \begin{bmatrix} \begin{pmatrix} x_{n-1} & +x_0\varsigma_x(n)\vartheta(n) \\ & -x_e(n-1) + x_e(n) \\ & +x_\Phi(n)\varsigma_y(n) \\ & +x'_0\varsigma'_x(n)\vartheta'(n) \\ & -x'_e(n) + x'_e(n) \end{pmatrix} \\ \begin{pmatrix} y_{n-1} & +y_0\varsigma_y(n) \\ & -y_e(n) + y_e(n) \\ & +y'_0\varsigma'_y(n) \\ & -y'_e(n) + y'_e(n) \end{pmatrix} \end{bmatrix}. \quad (4.21)$$

The last changes, within the main computation operations, are related to the two main equations of the computer program for the numerical simulations. These changes concern the enlarged spatial domain $[\mathbf{x}, \mathbf{y}]$, which goes from $-l'_x/2$ to $+l'_x/2$ in x direction, the axial dependence and pulse number dependence and the beam ellipticity. The updated main equations are given by

$$\phi(\mathbf{x}, \mathbf{y}, z, n) = \phi_0(z, n) \exp\left(-2\left(\frac{(\mathbf{x} - x_n)^2}{\omega_x^2(z)} + \frac{(\mathbf{y} - y_n)^2}{\omega_y^2(z)}\right)\right), \quad (4.22)$$

for the individual fluence, and by

$$\Gamma(\mathbf{x}, \mathbf{y}, z) = \sum_{n=1}^{N_p} \phi(\mathbf{x}, \mathbf{y}, z, n), \quad (4.23)$$

for the accumulated fluence. Finally, the deviation of accumulated fluence from optimal conditions needs to be quantified. This can be done by comparing numerically with analytically determined characterisation parameters. The analytical results are free from deviations and deviations are numerically added. Analytical equations are for example the quadratic term on the right side of Equation 3.38, which yields values for $\tilde{\Gamma}_p(\varphi)$. Therefore, first the numerical results are characterised by the output parameters defined in Subsection 2.3.3 and afterwards the analytical equations are executed for the same parameters.

The deviations can also be visualised, if the numerical data are recalculated using the analytical values as error free boundaries for the accumulated fluence domain. Two different boundaries have been applied. First, Γ_p and Γ_v are applied as boundaries for the normalisation of the upper and lower limit. This type of graph directly shows the spatial distributions of the deviations. Second, the domain boundaries can be defined as a percentage of Γ_c . This type of graph shows how the spatial emergence is influenced by the deviations. For both methods, areas of black and white colour indicate locations where the domain boundaries have been exceeded.

4.3 Fluence simulation results

4.3.1 Accumulation process errors

At first, the influence of different limitations on the irradiation process has been analysed on a small space domain for different pulse overlaps. The individual and accumulated fluence have been found to deviate from optimal conditions, due to the limitations. In the following, the influence of the limitations is shown with characteristic fluence profiles.

Figures 4.2 and 4.3 show simulation results #1 to #4 of a SIC and three 2d-DIC. The SIC simulation was performed with $P_a = 1.57$ mW at $f_r = 1$ kHz and $N_r = 200$. Figure 4.2a shows the power variation, Figure 4.2b shows the statistical variation of the x position, Figure 4.2c shows the individual fluence vector $\vec{\phi}(n)$ for the two analysis locations, as described in Chapter 2, Figure 4.2d shows the accumulated fluence matrix $\mathbf{\Gamma}(\mathbf{x}, \mathbf{y})$, Figures 4.2e shows the accumulated fluence matrix normalised by the boundaries Γ_p and Γ_v and Figure 4.2f by the boundaries $1.15\Gamma_c$ and $0.85\Gamma_c$.

The 2d-DIC simulations were performed with $P_a = 1.57$ mW at $f_r = 1$ kHz, $N_r = 1$ and $v = 35$ mm/s, see Figures 4.3a, 4.3d and 4.3g, $v = 18$ mm/s, see Figures 4.3b, 4.3e and 4.3h, and $v = 9$ mm/s, see Figures 4.3c, 4.3f and 4.3i. The top figures (a-c) show the accumulated fluence matrix $\mathbf{\Gamma}(\mathbf{x}, \mathbf{y})$ for the complete space domain, the middle figures (d-f) show the accumulated fluence matrix normalised by the boundaries Γ_p and Γ_v and the bottom figures (g-i) by the boundaries $1.15\Gamma_c$ and $0.85\Gamma_c$.

These simulations have been performed on an area of $l_x = l_y = 100$ μm and spatial resolution $q = 0.1$ μm . The beam radius was $\omega_0 = 10$ μm and the wavelength $\lambda = 1030$ nm. The beam profile was elliptical with $\varepsilon_x = 0.1$ and $\varepsilon_y = -0.1$. The average power was varied sinusoidally with $\tilde{P}_{e(1)} = \pm 0.04$ at $f_e = 0.01$ Hz and $\tilde{P}_{e(2)} = \pm 0.01$. Irradiated locations were varied statistically with $x_e = y_e = \pm 0.5$ μm and the

Figure 4.2: Simulation result #1 of a SIC with irradiation errors. The grey scale either represents the accumulated fluence in J/cm^2 or normalised by Γ_p and Γ_v in (e) and by $1.15\Gamma_c$ and $0.85\Gamma_c$ in (f).

phase shift amplitude was $x_{\Phi 0} = \pm 2.5 \mu m$ with a line period of $\Phi = 8$.

An elliptical beam profile causes the beam diameter to deviate in lateral directions, which influences the peak fluence and pulse overlap and therefore the individual and accumulated fluence. With respect to ϕ_0 , the particular direction of variation depends, on whether or not the ellipticity was previously considered, and if not, on the primary assumed beam diameter. With respect to φ , the overlap is different in x and y direction for equal lateral displacements in x and y direction. This causes, for LODIC and MODIC, the accumulated fluence to spread anisotropic over the area. In Figure 4.3e it can be seen that individual pulses are more connected in x direction than in y . If the ellipticity would be the only error, this effect would be constant for the whole irradiated area.

The variation of the power leads to a variation of the pulse energy and peak fluence following linearly the power error function, see Figures 4.2a and 4.2c. The variation in time is transformed into a spatial variation of the individual and accumulated fluence, where the temporal error frequency and the displacement velocity determine the localisation. Both, the long and short term drift, vary the peak or centre point of the accumulated fluence. In Figure 4.3i the long term drift can be observed as

Figure 4.3: Simulation results #2 to #4 of 2d-DIC with errors and different pulse overlaps. The grey scale either represents the accumulated fluence in J/cm^2 or normalised by Γ_p and Γ_v in (b,e,h) or by $1.15\Gamma_c$ and $0.85\Gamma_c$ in (c,f,i).

decreasing accumulated fluence in y direction.

Position errors vary the displacement between pulses. The effect of this variation depends on the pulse overlap and beam diameter. Under LODIC, the influence on neighbouring pulses is low, see Figure 4.3a. However, in this case pulses placed subsequently on one location do not coincide due to position errors. If pulses intersect significantly, as under HODIC, positioning errors lead to inhomogeneity, see Figure 4.3c. Although the errors do not modify the peak fluence, they can modify the indi-

vidual and accumulated fluence, because each displacement error, varies the individual fluence per location. Initial simulation results show that they determine the spread of accumulated fluence for high pulse overlaps.

A shift of irradiated lines, varies the start and end position of a line and the position of neighbouring pulses from subsequent lines, see Figures 4.3e and 4.3c. Whereas the variation of start and end positions increases with the shift, neighbouring pulses from subsequent lines get back in phase after a certain period. The shift varies the individual and accumulated fluence, due to the non-orthogonal contributions of subsequent pulses from different lines. The shift between lines can be different for each new line, e.g. increases until pulses are back in phase and then start again, dependent on the phase period.

Figures 4.4a to 4.4c show simulation result #5, which is a repetition of simulation #5 from Chapter 2 with $z_0 = 150 \mu\text{m}$. An offset from the centre of the beam waist in z direction, increases the beam diameter, which decreases the peak fluence and thereby the values within the individual fluence. Furthermore the accumulated fluence is lower for LODIC. The increased beam diameter also influences the overlap between subsequent pulses if the velocity is constant. This increases Γ_p and decreases Γ_d for MODIC. For HODIC these variations on the accumulated fluence have not been observed. If the axial offset would be the only error, its influence would be constant over the whole irradiated area.

The simulation with the second kinematic system and the assumed stitching boundary revealed under certain displacement conditions an inhomogeneous irradiated area. That means a stitching error is obtained, characterised by a variation of the accumulated fluence at the interface between stitched subapertures. Then further simulations were performed to identify the underlying mechanism. Figures 4.4d to 4.4f show simulation results #6 to #8 of stitched subapertures with different displacement boundaries. In Figure 4.4d the geometrical length was applied as the first stitching boundary and the stitching error was obtained.

Apparently, there are more factors influencing the stitching result then the so far considered length of the subaperture. In fact, if within the subaperture a certain pulse overlap is applied, then this pulse overlap also has to be considered as absolute value at the boundary between the subapertures. In Figure 4.4e, the length of the subapertures minus the absolute length of the pulse intersection was applied as the second stitching boundary. This boundary brought an improvement for certain pulse overlaps, but for other still an error has been found.

If the laser is switched on and off at the beginning and at the end of an irradiated line, only an integer value for the number of pulses can be applied. Because of that, the actual length of a subaperture can deviate from the defined length. Therefore, the actual length of a subaperture, based on the actually applied laser pulses, needs to be considered. In Figure 4.4f, the actual length of subapertures minus the absolute length of the pulse intersection was applied as the third stitching boundary and revealed for all investigated displacements an homogeneous irradiation.

Figures 4.4g to 4.4i show simulation result #9 for a larger space domain with irradiation errors. This simulation has been performed on an area of $l'_x = l'_y = 1100 \mu\text{m}$, with a subaperture length of $l_x = l_y = 200 \mu\text{m}$, a spatial resolution of $q = 0.5 \mu\text{m}$

Figure 4.4: Simulation results #5 to #9 of 2d-DIC with errors and different pulse overlaps. The grey scale either represents the accumulated fluence in J/cm^2 or normalised by Γ_p and Γ_v in (h) or by $1.15\Gamma_c$ and $0.85\Gamma_c$ in (i).

and a velocity of $v = 5 \text{ mm/s}$. The errors were the same as in the simulations #1 to #4 and in addition the subapertures locations were varied statistically with $x'_e = y'_e = \pm 1.0 \text{ } \mu\text{m}$. In this simulation, the spread of the accumulated fluence was found to be increased with $\Gamma_d = 0.66 \text{ J}/\text{cm}^2$ compared to Simulation #3 from Chapter 2 also with $\varphi = 0.75$ and $\Gamma_d = 0.00 \text{ J}/\text{cm}^2$. The average accumulated fluence was found to be decreased to $\Gamma_a = 6.04 \text{ J}/\text{cm}^2$ compared to a simulation without errors, where Γ_a

$= 6.28 \text{ J/cm}^2$ was obtained.

The diagonal in black colour in Figure 4.4h is produced by the long term drift of the average power. In Figure 4.4i it can be seen that the accumulated fluence exceeds the domain boundaries at the interface between the subapertures. This error is caused by the position errors of the stitching displacements. Another effect which can be observed is a spread of accumulated fluence within the subapertures, visible as noisy accumulated fluence. It cannot be distinguished from a single graph if this effect is, either caused by short term power variations or by position errors. But other simulations show that position errors have a large influence on the accumulated fluence spread especially for high pulse intersections.

4.3.2 Displacement sensitivity analysis

In the previous subsection, the influence of technical limitations on the irradiation process has been investigated. The individual simulations allowed to identify how the limitations influence the fluence accumulation process. For different pulse overlaps, the effect of the limitations on the fluence accumulation process was clearly different. In this subsection it is evaluated how sensitive certain irradiation output parameters react, to changes of irradiation input parameters, for different pulse overlaps. Therefore, the simulations presented in Subsection 2.4.2 are applied as basis and repeated for particular limitations. Figures 4.5a to 4.5i show the dependence of $\tilde{\Gamma}_p$, $\tilde{\Gamma}_v$, $\tilde{\Gamma}_d$ on φ for different technical limitations.

The Figures 4.5a to 4.5c show simulation results with an z -offset $z_0 = 150 \text{ } \mu\text{m}$. At low pulse overlaps the accumulated fluence is smaller because the increased beam diameter decreases ϕ_0 and consequently Γ_p . Hence, for low pulse overlaps, $\tilde{\Gamma}_d$ does not converge to 1. For higher pulse overlaps, this difference decreases and the peak and valley value approach the same value. In order to determine if the difference caused by the error is vanishing for higher pulse overlaps, the difference between the numerical and error free analytical parameter is calculated and given in Figure 4.5c. This figure shows that for low overlaps defocusing causes an accumulated fluence error, which disappears for high pulse overlaps. This is because the increased diameter causes a contribution of smaller fluence, which compensates the ϕ_0 reduction. Since the number of pulses is the same, the same result is obtained for ϕ_a . However, the individual fluence contains different values because ϕ_0 is decreased.

The Figures 4.5d to 4.5f show simulation results with an ellipticity $\varepsilon_y = 0.8$. For low pulse overlaps, the ellipticity causes an increase of peak fluence and a global anisotropic spread of accumulated fluence. In Figures 4.5d and 4.5e the accumulated fluence parameters are determined for the cross-sections along the x and y axes. Along the x direction, the accumulated valley fluence reaches higher values at lower pulse overlaps. The opposite is the case along the y direction. This result is confirmed by the data for $\tilde{\Gamma}_d$, see Figure 4.5e. However, similar to the result for the defocused beam, at high pulse overlaps the accumulated peak fluence with the error is the same as without and the anisotropic spread disappears, as indicated in Figure 4.5f.

In order to verify the three different stitching boundaries, the stitching simulations presented in Figures 4.4d to 4.4f have been repeated for different pulse overlaps. For

Figure 4.5: Simulation results for the dependence of $\tilde{\Gamma}_p$, $\tilde{\Gamma}_v$, $\tilde{\Gamma}_d$ on φ for different technical limitations, (a-c) defocusing, (d-f) ellipticity and (g-i) stitching.

the simulations the analysis area has been chosen larger than the size of a subaperture. In this way the values from the subaperture border are also considered. The obtained simulation results for the accumulated peak and valley fluence are shown in Figures 4.5g to 4.5i. These results confirm the observation from the previous subsection, that the stitching boundary depends on the actual length of subapertures and the abso-

lute length of the pulse intersection. For the first stitching boundary the accumulated fluence deviates from the optimum curve, shown in Figure 2.6e, for nearly all pulse overlaps. For the second stitching boundary the deviation is smaller compared to the first boundary. Only for the third stitching boundary the optimal curve of the accumulated fluence is obtained for all displacements.

Discussion

In this chapter an extension to the irradiation model has been presented, in order to analyse the influence of technical limitations on the irradiation process. The simulations show how the assumed variations influence the fluence accumulation process. The influence has been found in a deviation from optimal conditions, by a shift or spread of accumulated fluence and a variation of the individual fluence. The influence can be local or global, with respect to the irradiated area. One limitation can have multiple effects on the irradiation process. The mechanism underlying these effects, are related to variations of the peak fluence and pulse overlap.

The spatial emergence of LIPSS is related to the individual and accumulated fluence, as has been shown in Chapter 3. Therefore the deviations can easily influence the spatial emergence of LIPSS. On the one hand, an area homogeneously covered with LIPSS can be interrupted by the emergence of another morphology, if the accumulated fluence domain boundary is exceeded. On the other hand, the regularity of LIPSS can be affected, if the accumulated fluence spreads and addresses a range of values in the accumulated fluence domain of LIPSS. This effect could locally vary the height or splitting of LIPSS. However, detailed information about the spatial emergence of LIPSS under lateral displacement irradiation conditions with respect to their accumulated fluence domain boundaries and spatial properties are lacking in literature. But this information is important for the application of LIPSS in the field of surface functionalisation.

The analysis of deviations can be further improved if the artificial domain boundaries are replaced by experimentally determined domain boundary values for LIPSS. This would allow a comparison of the absolute error magnitude to the domain boundaries for LIPSS and to distinguish whether a certain error is critical or not. But also the effect of incubation should be considered.

Mostly, LIPSS are required to be uniformly distributed on a surface [159]. Therefore LIPSS areas are usually produced at high values of φ , see Figure 2.4f. If the applied irradiation deviates, see Figure 4.3c, the spatial emergence will also deviate, the irradiation parameters require further optimisation and this will increase the time required to empirically determine irradiation parameters. Whereas in practice, all effects contribute simultaneously, irradiation simulations offer the possibility to analyse the effect of a single limitation. Furthermore, in practice the required data for limitations are usually known from the specification of a laser system or are accessible by measurements qualifying a laser system. If simulations are performed with these data, realistic irradiation simulations can be obtained.

The simulations indicate that some limitations have less effect on the accumulated fluence at high intersections. In case of the z -offset the accumulated fluence deviates for low intersections but not for high. That means, a set of parameters would still

address the correct accumulated fluence value in the accumulated fluence domain for LIPSS. But a decreased individual fluence means that the location of the accumulated fluence domain could be different due to incubation. For the z -offset it seems important to determine the beam radius in the sample plane. This can be done by a D²-Experiment [95]. Such an experiment would also allow to determine the beam ellipticity. This information could be used to improve the accuracy of the peak fluence and to apply a different lateral displacement in x and y direction, which would reduce the spread of accumulated fluence present at low intersections. Also position errors seem to have an influence on the homogeneity of the irradiation. The effect of position errors could be reduced by increasing the beam diameter in focus, for example, with a beam expander.

4.4 Conclusions

The influence of technical limitations on the irradiation process for LIPSS was studied theoretically with irradiation simulations. Therefore the irradiation model has been extended by a set of assumptions for technical limitations. The influence of limitations has been found in a deviation of the irradiation from optimal conditions, by a shift or spread of accumulated fluence and a variation of individual fluence, related to variations of the peak fluence and pulse overlap. The analysis of the irradiation process by simulating the fluence accumulation process gives the possibility to perform realistic irradiation simulations and derive optimisation strategies for the determination of irradiation parameters. This is important for the design of a laser system and for the development of a robust process. The analysis of irradiation errors is required for the application of LIPSS for surface functionalisation. But the obtained results also identify interesting directions for further research. The relation between spatial properties of LIPSS on extended areas and accumulated fluence values addressed within an accumulated fluence domain is so far unclear and needs further attention.

Chapter 5

Irradiation experiments

5.1 Introduction

The aim of this thesis is to investigate the relation between irradiation parameters and the spatial emergence of LIPSS, as presented in Subsection 1.5. The theoretical results have been presented in Chapter 3. This chapter contains the experimental results of this investigation. But whereas Chapter 3 was based on the state of the art, this part complies with the theoretical considerations presented in Chapter 3. That means, the aim of this chapter is also to analyse whether the presented understanding of the spatial emergence is in agreement with the experimental observations and to verify whether the proposed approach for the determination of irradiation parameters is applicable. Therefore, a laser-material combination, relevant for the applications related to this thesis, has been selected. After a brief description of the applied setup and the followed methodology, the obtained results are presented in two parts. First a description of the results obtained in a static and second in a dynamic experiment.

5.2 Methodology

5.2.1 Laser system

This subsection describes the laser system used in the experiments presented in this chapter. As source for radiation a commercial laser system (Trumpf, TruMicro 5050) was used. This system has a compact structure and consists of a laser head and a supply unit. The supply unit accommodates electrical, optical, cooling, monitoring and control supplies and is connected to the laser head by cables and hoses. The laser head accommodates a mode-locked fibre oscillator seeding a regenerative Yb:YAG disk amplifier. The ring resonator of the amplifier is optically pumped in a multi-pass pump configuration by light with a wavelength of 940 nm. The laser diode array is located in the supply unit and connected by an optical fibre. The laser system delivers over an external electro-optical modulator and collimator laser pulses with

a fixed duration of 6.6 ps, central wavelength of 1030.2 nm and linear polarisation. Single pulses can have an energy up to 125 μJ and a period variable between 25 ms and 2.5 μs . This results in a repetition frequency between 40 Hz and 400 kHz and a maximum average power of about 50 W. The basic laser beam parameters are summarised in the top part of Table 5.1.

The fundamental wavelength of this laser system can be converted by external non-linear optical crystals to the second harmonic, which is $\lambda = 515$ nm, and third harmonic, which is $\lambda = 343$ nm. For each wavelength beam guidance and focusing elements are specific. For the experiments presented in this chapter the fundamental wavelength has been used. Beam guidance is done with highly reflective mirrors (Laser Components, HR1030+515/45 PP1025C). The mirrors consist of BK-7 substrates and dielectric coatings, which are designed for the specific operation wavelength and 45 degree angle of incidence. The energy of the laser beam can be attenuated externally, using a combination of a zero order half-wave plate (Thorlabs, WPH10M-1064) and a polarising beam splitting cube (Thorlabs, PBS 203). This allows to operate the system at high pump rates and to obtain pulses over a broad range with stable output conditions. All experiments are performed in direct writing configuration and the angle of incidence of laser light was perpendicular to the specimen surface. The beam was focused by a telecentric f-theta lens (Sill Optics, S4 LFT 0080/126) with an effective focal length of 79.9 mm.

Table 5.1: Basic laser beam parameters.

Parameters	Values
Wavelength	1030.2 nm
Pulse duration	6.6 ps
Polarisation state	linear
Single pulse energy	$\leq 125 \mu\text{J}$
Repetition frequency	$\leq 400 \text{ kHz}$
Power stability	± 1.0
Beam quality M^2	1.1
Beam circularity	94.3 %
Raw beam diameter ($1/e^2$)	4.3 mm
Focused beam radius ($1/e^2$)	13.1 μm

This laser system provides two positioning systems, a galvanometer-scanner system and a 3-axes linear translation system. The scanner system (Scanlab, IntelliScan 14) consists of two mirrors attached to two galvanometer scanners to allow for tiltable deflection. The mirrors and scanners are mounted into one scan head. The mirror coatings are designed for a wavelength of 1030 nm and have a reflectivity of more than 99.5 % per mirror over the full range of angles. The focusing lens is attached to the scan head. This scanner-lens combination results in a typical image field of 50 x 50 mm and a position repeatability of $< 1.8 \mu\text{m}$. The scan head is digitally controlled by a controller card (Scanlab, RTC4). The translation system consists of 3 mechanical

Figure 5.1: Design of the laser system used for experiments presented in this chapter. This graph uses the following abbreviations: IR infra-red, P 1-2 periscope 1-2, $\lambda/2$ half wave plate, PBS polarising beam splitter, BD beam dump, M1-M6 mirror 1-6 and X , Y and Z translation axes x , y and z .

bearing stages, two linear motor stages (Aerotech, ALS 20020) oriented horizontally as cross-pair for lateral beam displacements in x and y direction and a ball screw stage (Aerotech, ATS 150-250) oriented vertically for axial beam displacements in z direction. The repeatability of these axes is about $\pm 0.5 \mu\text{m}$. The translation stages are supported by digital servo amplifiers (Aerotech, Ndrive).

The scan head is mounted on the z -axis and aligned with the focusing lens downwards to the x - y stage. The scanner system modifies the beam position laterally in x and y direction before the focusing lens by deflection. The x - and y -axis modify the sample position laterally with respect to the beam focus position. The z -axis modifies the axial position of the scanner and therefore the focus position relative to the sample. If not stated differently, the beam manipulation, for the irradiation experiments in this chapter, was accomplished by the scanner system. The laser arrangement is mounted on a vibration isolated optical table and located in a temperature as well as humidity controlled clean room environment. All laser experiments were done in air, at atmospheric pressure and at room temperature $20 \pm 0.5^\circ\text{C}$ and relative humidity $40 \pm 10\%$. All supply units are stored separately in a temperature controlled room, to reduce the heat input into the cleanroom. The whole laser system is embedded in a laser safety system, including an enclosed cabinet.

The laser system is controlled by two computer systems with two separated control panels. One system is embedded in the laser supply and used to control the laser source. The other is stored in a separate cabinet, together with the positioning

supplies, and used to control the positioning and measurement systems. The panels use conventional computer equipment. The software for the laser source (Trumpf, TruControl 1000) allows control, monitoring and maintenance operations. Laser control can be done in two modes, manual and program mode. The same holds for the software (Aerotech, N-View) of the linear translation systems. For the experiments the stages were used in manual mode to position the sample visually. The scanner system was controlled by commercial available software (SCAPS, SamLight). For these experiments, programs have been developed in SamLight, in which a set of exposures have been defined. The laser source needs input of a trigger and a power level. To be able to use different inputs, an external electronic control unit (University of Twente, Triggerbox) has been made. The different inputs come from the second computer system, which holds the interface to the scanner and translation system. The trigger and power levels come from either the RTC4 card, the Ndrive system or a manual setting. Figure 5.1 shows a schematic drawing of the design of the laser system.

5.2.2 Laser irradiation

This and the following subsection describe the method followed in these experiments. The method can be divided in four steps: 1. Preparation, 2. Irradiation, 3. Imaging and 4. Analysis. Until the final experiment has been performed, this procedure has been repeated several times. Two types of experiments have been performed, first a sequence of locations has been irradiated statically by a single laser pulse or multiple laser pulses and second a sequence of locations has been irradiated dynamically by multiple displaced laser pulses. For each location a set of irradiation parameters have been varied, in static $[P_a, N_p]$ and in dynamic $[P_a, v, N_r]$.

At first the SamLight programs have been prepared. The programs contain the irradiated locations, irradiation size, scanner settings and a part of the irradiation parameters, for static N_p and for dynamic v and N_r . For these experiments the average power has been varied manually at the triggerbox to measure the power in the time between exposures. The irradiation locations and sizes were selected as such to optimise the influence on neighbouring locations, the irradiation time for the experiments and number of experiments per specimen. The scanner settings, as field of view, laser on/off delays, jump/mark delays, influence the accuracy of the displacements, as for example displacement distance, irradiated field size, acceleration/deceleration envelopes and displacement shifts between lines. These settings have been optimised experimentally, to ensure that the laser system accurately implements the irradiation.

After this optimisation, statistical variations have been observed. The absolute deviation of irradiation positions, determined in 2-on-1 experiments, has been found to be 0.6 μm in average and 1.6 μm maximum. This yields a deviation of the average fluence of 0.3 % in average and maximum 1.4 %, determined in irradiation simulations. The interval and boundaries for the static irradiation parameters have been determined experimentally as well. For the dynamic experiment the interval and boundaries for P_a and N_r have been chosen based on the static experiment and v has been selected with respect to effects of the pulse overlap.

Before irradiation the beam quality and profile were determined once: see Figure

Figure 5.2: Measurement results for the beam profile (left) and caustic (right).

5.2. The beam profile was measured with a camera based system (Thorlabs, BC106-VIS) before the focusing lens. This measurement yields a beam circularity of 94.3 % and a raw beam diameter of 4.3 mm. The beam quality was determined with a camera based beam diagnostic system (Primes, MicroSpotMonitor). This measurement yields a focused beam radius of $\omega_0 = 13.1 \mu\text{m}$ and a beam quality of $M^2 = 1.1$. The theoretical estimation yields a beam radius of $\omega_0 = 13.4 \mu\text{m}$. The average power of the laser was measured before each experiment with a power meter (Coherent, Field Max II-TO) equipped with a thermophile sensor (Coherent, PM 10). The convective air cooled measurement head is suitable for the power range 5 mW to 10 W and provides a noise equivalent power of 0.2 mW. The measurement head was positioned in a defocused plane behind the exit of the galvanometer scanner system. A measurement over 5 hours revealed a long term power variation of less than $\pm 1.0 \%$ determined in a range between 0.2 W and 10 W. The measured laser beam parameters are summarised in the bottom part of Table 5.1.

The overall aim of this research was to support the application of LIPSS. Therefore, the presented theory was evaluated with respect to an application. The leading application for this research was the NanoClean project, see Section 6.2. Because of that, a stainless mould tool steel was selected as material for the specimens. The brand name of the steel is Mirrax ESR and this steel is produced by the company Uddeholm. Mirrax is a modified AISI 420 steel with a chemical composition: 0.25 % C, 0.35 % Si, 0.55 % Mn, 13.3 % Cr, 0.35 % Mo, 1.35 % Ni, 0.35 % V and +N [168]. The material was ordered in form of a plate, which has been cut in cylindrical parts of 10 mm height and 30 mm diameter by wire electrical discharge machining. The size of the samples has been selected to match the capabilities of measurement devices. Afterwards each sample has been glued (ATA Hydraulik GmbH, Opal 450) and polished (ATA Hydraulik GmbH, Saphir 360 E, Rubin 520) to improve the sur-

face quality of the front face. The polishing process followed a standard routine for materials analysis with conventional utilities. The polished surface has a roughness R_a below 10 nm in a measurement window of 69.1 μm by 92.2 μm . If not stated differently, samples are handled with gloves. Cleaning of the specimen was done before and after laser irradiation. First an ultra-sonic-bath filled with Isopropanol was used. Secondly, the sample surface was cleaned manually following a standard routine for cleaning optics. The data for the irradiation parameters and further experimental information have been recorded in electronic datasheets (Microsoft, Excel 2010).

5.2.3 Specimen analysis

Inspection of the specimen surface after irradiation was done by optical microscopy (Leitz, DMRX), abbreviated OM, confocal laser scanning microscopy (Keyence, VK 9710K), abbreviated CLSM, and scanning electron microscopy (JEOL, JSM 6400), abbreviated SEM. The OM was used for an initial inspection in the laser laboratory after irradiation. It yields information about the quality of the machining and cleaning procedure. SEM was used for the analysis of the surface modifications. Next to the qualitative analysis of the obtained surface structures, it was also used for the quantitative analysis of their lateral dimensions. CLSM was used for the detailed analysis of the depth of ablation craters. The illumination in the CLSM is accomplished by a laser of 408 nm wavelength. A microscope objective with a magnification of 150x and numerical aperture of 0.95 was used.

Imaging was followed by analysis, which was divided in three steps. First the qualitative analysis of the surface modifications. Second the analysis of the spatial properties of the obtained structures. Third the location of the parameters range for the spatial emergence. The surface modification analysis has been done on the basis of SEM images. Next the spatial properties, such as periodicity, orientation and regularity, of LIPSS were determined. The periodicity was measured with two methods. First, the distances between individual periods have been measured manually in SEM images. This has been repeated at least ten times at different locations. Values obtained by this method were denoted by Λ_{MA} . Second, Fourier transform of the SEM images were generated. Values obtained by this method were denoted by Λ_{FT} .

Finally, the results are encoded by different colours and assigned to the parameters in the electronic datasheets. In particular cases, the method of Liu [95], see Chapters 1 and 3 and Equation 3.18, has been applied, to specify the range of spatial emergence in more detail. Therefore, the outer and inner diameters of the surface modification have been measured. This was done with a commercial software (NIH, ImageJ 1.46r), where polylines were applied to encircle the outer borders of the structures. The information about the power and diameter measurements were used to determine the beam diameter in the sample plane and the fluence thresholds, also with commercial software (MathWorks, Matlab 2011). The beam diameter was determined in the 1-on-1 experiment and the obtained result was applied to calculate the peak fluence and accumulated peak fluence in the datasheets. The multi pulse experiment supported another parameter, the incubation factor.

Basically, the dynamic experiment was analysed by the same procedure, except

for one additional step. In static, the datasheet initially contains the laser system parameters. If the parameter ω_0 is determined, for each power value the parameter ϕ_0 is assigned. Based on this, the output parameter Γ_p can be calculated by a multiplication of ϕ_0 and N_p for each irradiation trial. In dynamic, an additional computation step was required. As can be seen in Equation 3.24, to determine the parameter Γ_p in dynamic, the parameter $\tilde{\Gamma}_p$ is required. This parameter has been calculated with the computer program for the irradiation model described in Section 2.3 and Subsection 4.2.2.

In the dynamic experiment, a few values for P_a and a broad range of values for φ and N_r have been applied. Therefore, in the datasheets basically 2d φ - N_r matrices were obtained, each corresponding to a different power value. In order to verify the approximations for the number of irradiation repetitions, the domain boundaries have been determined experimentally for each value of φ . These boundaries have been applied for a comparison with the numerical simulations.

5.3 Static experiment

5.3.1 Surface modifications

The following section describes the observations, which have been made in the microscopic analysis of the statically irradiated surface. The static experiment was repeated four times in order to optimise the interval and boundaries of ϕ_0 and N_p empirically. During the optimisation attention was paid to the spatial emergence of LIPSS. Each trial has been analysed partially by microscopy. The observed surface modifications and their parameter ranges have been found to be reproducible over these trials. The last of the four trials has been analysed in more detail and the obtained results are presented in this section. This particular subsection, contains the description of the observed surface modifications, with the primary focus on LIPSS.

After the irradiation, a film of ablated and redeposited material was observed on the surface. By cleaning, a major part of this material has been removed. In general, the cleaning procedure used, did not damage the surface structures. However, after cleaning some contamination still remained on the surface. The contamination consists of particles collected around or within the structures and thin films appearing dark in the SEM images. The SEM images have been recorded over several weeks in a laboratory without clean-room equipment. Therefore, the samples were cleaned several times manually with an IPA solution before the individual measurements. Over time, the sizes of the dark areas increased. Since, one cause was seen in the IPA solution, this was not applied anymore.

For this laser-material combination different surface modifications have been observed. They are mentioned and enumerated in the order in which they were observed with increasing pulse number. The enumeration uses the abbreviation SM for surface modification and a roman number as index. In the following text and figures, the spatial emergence of these surface modifications is described and illustrated for the investigated parameter range. The figures show the development of these modifications at various stages, even at initial stages where the structures are hardly visible.

Figure 5.3: SEM images of the steel sample surface after irradiation with varying ϕ_0 and N_p under SIC. These images show the surface modifications SM-I and SM-II (top) and SM-III and SM-IV (bottom). The inserts are doubled in size.

Figure 5.3 top left and right show SEM images of the steel sample surface exposed to $N_p = 1$. Two different modifications have been observed. One appears in the SEM as a sharply defined circular surface region surrounded by a rim. This one was denoted by SM-I, see Figure 5.3 top left. The other is denoted by SM-II and hardly visible in Figure 5.3 top left due to the initial emergence stage. In Figure 5.3 top right the SM-II appears as a molten surface and contains bubbles and exploded craters. Only for $N_p = 1$, no periodic surface structures have been observed. The LIPSS observed for $N_p > 1$ showed a unidirectional appearance.

In the range $1 < N_p < 10$ two types of LIPSS have been observed. Figure 5.3 bottom left and right show the emergence of these structures in an initial stage. One type forms spatially in the same region as SM-I and has a rather small period $\Lambda < \lambda$ and an orientation $\Theta = \parallel$ to the polarisation. This modification was denoted by SM-III. The other type forms spatially in the same region as SM-II and has a period $\Lambda \sim \lambda$ and an orientation $\Theta = \perp$, denoted by SM-IV. With increasing N_p they become clearly developed. Figure 5.4 shows SEM images of these structures at different values for ϕ_0 and N_p .

Figure 5.4: SEM images of the steel sample surface after irradiation with varying ϕ_0 and N_p under SIC. These images show the surface modifications SM-III and SM-IV at different formation stages. The inserts are doubled in size.

In the range $10 < N_p < 100$ the SM-III disappear and the appearance of SM-IV varies. The variation is characterised by different changes. In the range $1 < N_p < 10$ the formation of the SM-IV was characterised by bubbles forming on top of the rims, see Figure 5.4 bottom left. In the range $10 < N_p < 100$ the SM-IV lose the bubbles on top of their elevations and the rims appear smoother, see Figure 5.4 bottom right. Another change is that they start to split and merge for a specific range of fluence. The splitting is characterised by a smaller periodicity and same orientation, see insert of Figure 5.4 bottom right. This variation of the SM-IV is seen as a new modification denoted by SM-V.

In the range $100 < N_p < 2000$ three other modifications have been observed. One was the formation of hole structures, denoted by SM-VI. In Figure 5.5 top right and bottom left SEM images of these structures are shown. The structures can be observed in the centre of the irradiated area. This phenomenon was difficult to identify, due to the small apparent spot size. However, also this phenomenon seems to be periodic and to have a specific orientation, which was found to be $\Lambda > \lambda$ and $\Theta = \parallel$. Towards higher fluences and pulse numbers, the holes merge to grooves. They clearly

Figure 5.5: SEM images of the steel sample surface after irradiation with varying ϕ_0 and N_p under SIC. These images show the surface modifications SM-IV, SM-V, SM-VI and SM-VIII.

build an upper border, in terms of fluence, which separates the smaller perpendicular structures, compare Figure 5.5 bottom left.

The hole structure seems to be accompanied by the formation of an ablation crater. Ablation is denoted by the surface modification number SM-VII. The last type of surface modification which has been observed are again periodic structures and have been denoted by SM-VIII. In Figure 5.5 bottom right a SEM image of this structure is shown. Interestingly, the elevations of these LIPSS appear as if they are untreated and no material ablation has occurred. The previously described structures seem to form beneath. This structure was characterised by $\Lambda \sim \lambda$ and $\Theta = \parallel$.

Figure 5.5 top left shows a SEM image where the SM-V can be observed in a zone at the right edge of the irradiated area. They can be identified by their straight and long propagation. At the outer boundary, the structures are partly covered by particles. Inwardly, a more irregular structure with larger periodicity emerges. This variation is seen as the back merged SM-IV. They form an upper boundary for the SM-V, which was more difficult to define than the lower. The insert of Figure 5.5 top left shows the formation of the splitting phenomenon at an initial state.

To summarise, eight different surface modifications have been observed in the static irradiation experiment. Five of them show spatial periodic and unidirectional characteristics. Two of the periodic phenomena show a periodicity in the order of the irradiation wavelength. Two of them have a periodicity significantly smaller and one larger. Two phenomena show an orientation perpendicular and three parallel to the polarisation direction. This analysis was performed with a SEM. Other microscope techniques may reveal additional surface modifications.

5.3.2 Spatial characteristics

This subsection is dedicated to the analysis of the spatial characteristics of the observed LIPSS. Therefore, the lateral sizes of the structures in the SEM images are analysed and data from CLSM measurements are added. For $N_p = 1$ no periodic structures have been observed. However, two different modifications have been observed in two different fluence regimes. The CLSM analysis reveals for the slightly elevated rim around the circular region of SM-I a step of about 16 nm height from the peak of the rim to the inner area. For SM-II, the peak to valley height between the zone which appears molten and untreated surface region varies up to $d = 100$ nm.

The periodicity of SM-III has been determined at $\phi_0 = 1.21$ J/cm² and $N_p = 4$ and revealed an average of $\Lambda_{MA} = 372$ nm. With this value, the SM-III was the structure with the smallest periodicity, in this experiment. At $\phi_0 = 1.21$ J/cm² and $N_p = 3$ the periodicity has been found to be even smaller, with $\Lambda_{MA} = 281$ nm. In the SEM image, Figure 5.4 top right, it can be seen that the periodicity varies cross the irradiated area. Differences between $\Lambda_{MA} = 200$ nm at the edge and $\Lambda_{MA} = 500$ nm in the centre have been observed. This was the largest spread of periodicity in this experiment. The Fourier transform of this SEM image is shown in Figure 5.6b and confirms this observation with a characteristic ellipse shape for the frequency distribution.

The periodicity of SM-IV has been determined at $\phi_0 = 1.21$ J/cm² and $N_p = 6$ and revealed an average of $\Lambda_{MA} = 906$ nm. The spread of periodicity was smaller compared to SM-III. The Fourier transform of the SEM image $\phi_0 = 2.98$ J/cm² and $N_p = 2$ is shown in Figure 5.6a and revealed $\Lambda_{FT} = 955$ nm. The frequency distribution shows a characteristic sickle shape. The comparison of measurements at different values for N_p revealed that the period gradually decreases with increasing N_p , from $\Lambda_{FT} = 955$ nm at $N_p = 2$ down to $\Lambda_{FT} = 587$ nm at $N_p = 2000$, compare Figure 5.7.

The modification SM-V was characterised by a very regular, straight and fine shape. The periodicity of these LIPSS has been determined at $\phi_0 = 0.34$ J/cm² and $N_p = 100$ and revealed an average of $\Lambda_{MA} = 420$ nm. Figure 5.6c shows the Fourier transform for the SEM image at $\phi_0 = 0.71$ J/cm² and $N_p = 40$. This image shows the frequencies of SM-IV and SM-V. In this image the period of SM-IV was $\Lambda_{FT} = 884$ nm and of SM-V was $\Lambda_{FT} = 444$ nm, which is half of the period of SM-IV. Both phenomena have a similar frequency distribution. Although they were not possible to observe in the SEM image, in the frequency domain of the Fourier transform the SM-V structures were clearly identified.

Figure 5.6: Fourier transforms of SEM images after exposure to 5.6a $\phi_0 = 2.98 \text{ J/cm}^2$ and $N_p = 2$, 5.6b $\phi_0 = 1.21 \text{ J/cm}^2$ and $N_p = 4$, 5.6c $\phi_0 = 0.71 \text{ J/cm}^2$ and $N_p = 40$ and 5.6d $\phi_0 = 0.08 \text{ J/cm}^2$ and $N_p = 2000$. The images use a linear grey scale. The frequency domain was normalised by the norm of the wave vector, $2\pi/\lambda$. A high pass filter was applied, since these frequencies are mainly related to the ablation crater.

The periodicity of SM-VI has been determined at $\phi_0 = 0.34 \text{ J/cm}^2$ and $N_p = 200$ and revealed an average of $\Lambda_{MA} = 3152 \text{ nm}$. Below this pulse number, the SM-VI structure forms less periodic. In the SEM images, compare Figure 5.5 top right and bottom left, only between 1 to 3 periods can be observed. This was related to the small ratio of modification diameter to structure period. Under these conditions it

Table 5.2: Measurement results for the spatial sizes.

ϕ_0 J/cm ²	N_p -	Γ_p J/cm ²	Numeration -	Λ_{MA} nm	Λ_{FT} nm	Θ -	d μm
2.98	2	5.96	SM-IV	-	955	⊥	0.2
1.21	3	3.64	SM-III	281	295		0.1
1.21	3	3.64	SM-IV	-	936	⊥	0.1
1.21	4	4.85	SM-III	372	496		0.2
1.21	4	4.85	SM-IV	-	871	⊥	0.2
1.21	6	7.28	SM-III	-	483		0.3
1.21	6	7.28	SM-IV	906	878	⊥	0.3
1.21	15	18.21	SM-IV	-	886	⊥	-
1.21	15	18.21	SM-V	-	442	⊥	-
0.71	40	28.38	SM-IV	881	884	⊥	0.9
0.71	40	28.38	SM-V	-	444	⊥	0.9
0.71	100	70.95	SM-IV	-	800	⊥	2.1
0.71	100	70.95	SM-V	420	421	⊥	2.1
0.34	100	33.75	SM-IV	-	758	⊥	1.3
0.34	100	33.75	SM-V	-	433	⊥	1.3
0.71	200	141.90	SM-IV	-	749	⊥	4.2
0.71	200	141.90	SM-V	-	417	⊥	4.2
0.71	200	141.90	SM-VI	3152	-		4.2
0.71	300	212.84	SM-VI	3092	-		7.0
0.08	1000	84.86	SM-IV	-	626	⊥	-
0.08	1000	84.86	SM-VIII	-	996		-
0.08	2000	169.72	SM-IV	-	587	⊥	0.0
0.08	2000	169.72	SM-VIII	1003	963		0.0

was not possible to obtain useful information from the Fourier transform of the SEM images for SM-VI.

The CLSM analysis revealed an ablation crater for all modifications except the last one. This confirms that the SM-VIII form close to the untreated surface. Another structure which also forms very close to the untreated surface was SM-V. With the transition back to SM-IV, the ablation depth increases. With increasing accumulated fluence the hole phenomena forms, which was accompanied by a removal of material in the micrometre range, indicated as d in the last column of Table 5.2.

The periodicity of SM-VIII has been determined at $\phi_0 = 0.08$ J/cm² and $N_p = 2000$ and revealed an average of $\Lambda_{MA} = 1003$ nm. Figure 5.6d shows the Fourier transform of the corresponding SEM image. The periodicity of SM-VIII was $\Lambda_{FT} = 963$ nm. Next to this it shows the large frequency spread of the perpendicular features.

In literature it is commonly accepted to distinguish LSFL and HSFL, which are either parallel or perpendicular to the polarisation direction [169]. With the period-

Figure 5.7: Dependence of the SM-IV spatial period on the number of laser pulses.

icity smaller than half of the irradiation wavelength and an orientation parallel, the observed SM-III can be attributed to HSFL type of LIPSS. In different Fourier transforms, this type shows a characteristic ellipse shape distribution. This is in agreement with the information from the SEM images, where the periodicity has been found to vary. This points to a fluence dependency of the periodicity. Such a dependence has been described in literature for LIPSS [170, 171]. But, the results in literature have been obtained with a different laser-material combination.

With the periodicity equal to the laser wavelength and an orientation perpendicular to the polarisation, the observed SM-IV can be attributed to LSFL type of LIPSS. For femtosecond pulse irradiation of silicon surfaces a decrease of the spatial period of LSFL with increasing pulse number has been observed [130]. This property has also been observed for the SM-IV, compare Figure 5.7, which decrease in the range from $N_p = 2$ to $N_p = 2000$ down to 57 % of the irradiation wavelength.

With the periodicity smaller than half of the irradiation wavelength and an orientation perpendicular, the observed SM-V can be attributed to HSFL. But interestingly, these LIPSS show different properties compared to the SM-III HSFL. Their orientation is perpendicular and their periodicity has a distribution similar to the SM-IV LSFL. However, using the periodicity as criterion they belong to HSFL. The SM-V structures have also been observed in literature and attributed to HSFL [172].

For femtosecond pulse irradiation of silicon surfaces, it is reported that for high number of laser pulses per area, $N_p > 100$, periodic groove structures can occur [130]. The SM-VI is characterised by the formation of holes, which form, with increasing ϕ_0 and N_p , grooves. The grooves have a period of 3 times the irradiation wavelength and an orientation parallel to the polarisation direction. Therefore, this structure can be attributed to LSFL, as described in reference [130] for a different laser-material combination.

With the periodicity about the wavelength, the observed SM-VIII could be at-

Table 5.3: Observed and classified LIPSS.

Numeration	Periodicity	Orientation	Classification
SM-III	$< \lambda$	$\parallel$	HSFL
SM-IV	$\sim \lambda$	$\perp$	LSFL
SM-V	$< \lambda$	$\perp$	HSFL
SM-VI	$> \lambda$	$\parallel$	LSFL
SM-VIII	$\sim \lambda$	$\parallel$	LSFL

tributed to LSFL, but remarkably enough the orientation of this LIPSS was found to be parallel instead of perpendicular to the polarisation direction. Although the observation of parallel LSFL is rare, the SM-VIII are classified as those because their periodicity is even closer to the irradiation wavelength than the SM-IV. The observed structures and their classification are summarised in Table 5.3.

5.3.3 Irradiation parameters

For the static experiment the peak fluence was varied in 54 steps between $\phi_0 = 0.02$ J/cm² and $\phi_0 = 5.90$ J/cm² and the number of pulses covered the range $N_p = 1$ and $N_p = 2000$ in 36 steps. The observed surface modifications have been encoded by different colours and their observation has been denoted in the electronic datasheet. Figure 5.8 shows a cut-out of the datasheet. The assignment of observations to irradiation parameters yields an overview about the spatial emergence of LIPSS. Settings with no colour indicate that these images have not been archived for later analysis. The colour grey denotes archived but not encoded images.

The presented range of fluence has been adjusted during 4 trials to observe LIPSS. For increasing fluence, LIPSS have been observed shortly after the material starts to modify and until strong ablation starts to occur. The latter case was characterised by the formation of an ablation crater for high values of N_p , where LIPSS form at the walls of the crater. With respect to the pulse number, the first initiation of periodic surface structures have been found at $N_p = 2$ and for all other investigated values, LIPSS have been observed.

The SM-I and SM-II appear in different fluence regions, SM-I between $\phi_0 = 0.30$ J/cm² and $\phi_0 = 1.48$ J/cm², encoded by light blue, and SM-II between $\phi_0 = 1.73$ J/cm² and $\phi_0 = 5.90$ J/cm², encoded by dark blue. SM-III, as the first LIPSS type, emerge in a fluence range between $\phi_0 = 0.30$ J/cm² to $\phi_0 = 1.35$ J/cm², encoded by light orange, and SM-IV, the second LIPSS type, between $\phi_0 = 1.42$ J/cm² to $\phi_0 = 2.98$ J/cm², encoded by light green. In comparison with the modifications found at $N_p = 1$, the parallel structures SM-III form in the fluence region of SM-I and the perpendicular structures SM-IV in the fluence region of SM-II.

Between $N_p = 2$ and $N_p = 5$ the range of fluence can be clearly separated for both structures. Between $N_p = 6$ and $N_p = 10$ the perpendicular structures propagate spatially into the region of the parallel. This means they form at the same range of

Figure 5.9: (a) Dependency of the squared outer diameter of SM-I on the peak fluence for $N_p = 1$ (Semi-logarithmic). (b) Dependency of the accumulated fluence thresholds on the number of pulses (Double-logarithmic).

fluence, which makes a clear distinction difficult. The modification SM-III is clearly developed at $N_p = 4$ and SM-IV at $N_p = 20$ and SM-III vanishes between $N_p = 10$ and $N_p = 20$ and SM-IV at $N_p > 2000$. Actually the disappearance of SM-IV has not been observed in this experiment.

At $N_p = 20$ the formation of SM-IV undergoes a morphological disturbance between $\phi_0 = 0.21 \text{ J}/\text{cm}^2$ and $\phi_0 = 0.23 \text{ J}/\text{cm}^2$. The modification SM-V, encoded by dark green, emerges with increasing N_p within this fluence range. The splitting phenomenon is fully developed at $N_p = 100$ and has been observed until $N_p = 800$. However, in the range $N_p > 500$, a reliable distinction of SM-IV and SM-V was difficult by SEM. Overall, the SM-V showed a narrow window for ϕ_0 , compared to the LIPSS types SM-III and SM-IV, and have been observed only for a few values of ϕ_0 .

The hole phenomenon, SM-VI, emerge at $N_p = 100$ between $\phi_0 = 0.49 \text{ J}/\text{cm}^2$ and $\phi_0 = 0.94 \text{ J}/\text{cm}^2$, encoded by the colour brown. The holes have been observed between $N_p = 100$ and $N_p = 2000$. The observation of the holes was found to be influenced by the quality of the SEM observations and their stochastic emergence. The formation of ablation craters, SM-VII, has not been encoded by a colour or assigned to the irradiation parameters in the datasheet.

The structure SM-VIII has been observed at the lowest fluence, $\phi_0 = 0.06 \text{ J}/\text{cm}^2$ and $\phi_0 = 0.08 \text{ J}/\text{cm}^2$, and highest pulse numbers, $N_p = 800$ and $N_p = 2000$. This modification has been encoded by dark orange. This phenomenon just appeared within the window of applied experimental parameters.

The datasheet denotes the experimentally applied values, which have been varied

Table 5.4: Selection of boundary values for the spatial emergence of SM-V denoted by different threshold parameters. The data are extracted from the datasheet, see Figure 5.8.

$\phi_{th}(N_p)$ J/cm ²	$N_{th}(\phi)$ -	$\Gamma_{th}(N_p, \phi)$ J/cm ²	$\Delta\Gamma_{th}(N_p)$ J/cm ²	$\Delta\Gamma_{th}(\phi)$ J/cm ²
0.21	20	4.20	0.47	4.20
0.19	30	5.57	1.45	5.57
0.16	40	6.43	1.97	9.65
0.14	50	6.81	2.47	34.04
0.11	100	11.05	5.03	77.36

stepwise. Therefore the observation of the spatial emergences follows the stepwise variation of the accumulated fluence. The step size can be large, especially for high values of N_p . In order to specify the boundaries for the spatial emergence in more detail either more experimental values could be applied or the boundaries could be extrapolated. The latter can be done with the method of Liu [95].

In order to determine the single pulse modification threshold, this method has been applied for SM-I. Figure 5.9a shows the quantitative analysis of the diameters dependent on the peak fluence. The experimental data follow in this semi-logarithmic representation a straight line. The slope of the line yields the beam radius in the sample plane with $\omega_0 = 12.7 \mu\text{m}$. The extrapolation yields a single pulse fluence threshold of $\phi_{th}(1) = 0.27 \text{ J/cm}^2$. This value was found to be in agreement with the observation shown in Figure 5.8.

Within the range of investigated values for N_p , the material has been found to modify at lower fluences with increasing pulse number. In Figure 5.8 this can be observed for all modifications. This observation points to the incubation properties of the material. In order to quantify this property the incubation parameter has been determined, by analysing the outer modification diameters of SM-III. Figure 5.9b shows the dependency of the accumulated fluence threshold on the pulse number. This analysis also yields a straight line, whose slope yields the incubation parameter $\xi = 0.82$, see Equation 3.4.

In order to investigate the spatial emergence of LIPSS in static, but also for a comparison with dynamic, a particular type of LIPSS has been selected. The SM-V is a type of modification, which belongs to the group of LIPSS, their range of spatial emergence is enclosed by the investigated parameter range, they form close to the untreated surface and emerge at moderate values of ϕ_0 and N_p , so that a comparison between static and dynamic is possible. A selection of values from the datasheet is presented in Table 5.4. The data of different threshold parameters for SM-V revealed, that ϕ_{th} decreased with increasing N_p , N_{th} increased with decreasing ϕ and Γ_{th} and $\Delta\Gamma_{th}$ increased with increasing N_p and decreasing ϕ . Furthermore, the domain size was larger in terms of accumulated fluence for the same accumulated fluence threshold value, when N_p was varied instead of ϕ .

Table 5.5: Comparison of observed (upper part) with extrapolated (lower part) domain boundary values.

N_p -	$\Gamma_{th(i)}(N_p)$ J/cm ²	$\Gamma_{th(i+1)}(N_p)$ J/cm ²	$\Delta\Gamma_{th(i)}(N_p)$ J/cm ²
100	11.05	16.08	5.03
200	22.10	27.23	5.13
300	33.16	40.85	7.70
400	44.21	54.47	10.26
100	8.97	17.24	8.26
200	17.69	31.54	13.85
300	23.99	42.93	18.94
400	32.82	57.30	24.48

Also the fluence range of LIPSS can be specified in more detail by extrapolation. This has been done exemplarily for the values $N_p = 100$, $N_p = 200$, $N_p = 300$ and $N_p = 400$, see Table 5.5. This range is interesting, because at those pulse numbers the SM-V were found to be fully developed and the accumulated fluence was found to be sufficiently large to apply these data for an approximation of irradiation parameters under HODIC. The extrapolation extends the experimental data and confirms the observed range of fluence denoted in the datasheet. Since in the datasheet the actual observation was denoted, the extrapolated values yield a larger accumulated fluence range. For individual cases the extrapolated value can be below the value for the next closest experimental step. This is because the spatial emergence of LIPSS requires a minimum lateral space.

Discussion

The determination of the spatial emergence was slightly influenced by the type of SEM and cleaning procedure. With respect to the available resolution, it was hardly possible to observe SM-III. That means, it was not possible to observe structures with a periodicity below 100 nm. Next, the achieved cleaning result, influenced the determination of the spatial emergence boundaries. In individual cases the exact identification of a modification was more difficult, because the structures were partly covered by impurities, compare the edge of the irradiated area in Figure 5.5. However, these issues have a minor influence on the overall results for the range of parameters, due to the extensive scope of the experiment.

Furthermore, the determination of the boundaries was influenced by the transition between LIPSS morphologies. Whereas, the outer boundary of SM-I was found to be very sharp and hence was defined very precise, the upper boundary of the SM-V was more fuzzy and its determination consequently has a larger uncertainty. The latter required a detailed study of the SEM images. Nevertheless, the quality of the obtained results was sufficient to determine unambiguously the different types of LIPSS and their range of spatial emergence.

A characteristic parameter to describe the laser-material interaction is the single pulse fluence modification threshold. In this study the surface modifications were analysed by a SEM and the fluence value where the material starts to modify was found to be $\phi_{th}(1) = 0.27 \text{ J/cm}^2$, as indicated in Figure 5.9a. This may not be the lowest modification threshold of this specific laser-material combination. Because, other analysis methods, such as OM or AFM, could reveal further surface modifications. In literature for laser irradiation with $\lambda = 775 \text{ nm}$ and $\tau = 150 \text{ fs}$ of stainless steel an ablation threshold value of $\phi_{th}(1) = 0.21 \text{ J/cm}^2$ has been found [173]. This analysis was also done with a SEM, but of different type. That the value from literature is slightly smaller, can be due to the smaller wavelength and pulse duration, which cause a higher absorption and increased intensity. But, also the type of SEM can lead to different observations.

In this study, a significant incubation has been observed. The incubation parameter has been determined for the modification SM-I and SM-III until $N_p = 10$, because the modification SM-III has not been found at $N_p = 1$ but between $N_p = 2$ and $N_p = 10$ and forms exactly within the fluence boundaries of SM-I. In literature publications can be found, such as [97], which determine this parameter also for higher values of N_p . This has not been done in this study because a significant variation of the type of surface modifications until $N_p = 100$ has been noticed. In literature it cannot be found how the incubation parameter is influenced by the determination based on different types of modifications. However, in this study a sufficient number of N_p was selected and a sufficient extrapolation quality was obtained, see Figure 5.9b. The latter also indicates that the selected incubation model is suitable to describe the incubation behaviour of the selected laser-material combination. In this thesis a value of 0.82 has been found for the incubation parameter. In literature for laser irradiation with $\lambda = 775 \text{ nm}$ and $\tau = 150 \text{ fs}$ of stainless steel a value of $\xi = 0.86$ has been found [173]. The difference for the incubation parameter can be due to the different type of steel, different pulse number range and uncertainties in the determination procedure.

In conclusion: The accumulated fluence was found to be a suitable quantity to describe the spatial emergence of surface modifications. Together with an information about the individual fluence, such as the peak fluence, a good comparison of different static irradiation results is possible. The incubation properties of this material can be described by the selected incubation model. The spatial emergence was indeed determined by an accumulated fluence domain, whose location and size is influenced by the incubation properties of the material and varies with the irradiation parameters. The variation of the threshold parameters were found to be in agreement with the theoretical description. The size of the domain was found to be dependent on the irradiation input parameter that was varied, in this particular case larger for the variation of the pulse number. The static irradiation experiment revealed an overview about the available LIPSS and allowed to determine their domain boundaries with a minimal experimental effort. Overall, the results obtained in the static irradiation experiment confirm the description of the spatial emergence of LIPSS under SIC presented in Chapter 3. However, this experiment could be improved by a high resolution SEM and the outcome is influenced by the cleaning procedure. Further study to obtain experimental values for SM-V and SM-VIII would be interesting.

5.4 Dynamic experiment

5.4.1 Surface modifications

The following section describes the observations, which have been made in the microscopic analysis of the dynamically irradiated surface. The dynamic experiment was repeated several times in order to optimise the scanner parameters and irradiation time, see Subsection 5.2.2. Each trial has been analysed partially by microscopy. The last of the four trials has been analysed in more detail and the obtained results are presented in this section. This particular subsection, focuses on the description of the morphological changes due to the introduction of lateral displacements.

With the dynamic irradiation, the exposed area has been increased. This caused more material to be ablated, which increased the contamination of the structured surfaces compared to the static experiment. By cleaning, a major part of this material has been removed. However, after cleaning some contamination still remained on the surface. The contamination consists of particles collected around or within the structures and thin films appearing dark in the SEM images. In general, the cleaning procedure used after laser irradiation, did not damage the surface structures. But, in particular cases scratches on the structures have been observed.

Figure 5.10 shows SEM images of the steel sample surface after irradiation with $\phi_0 = 0.15 \text{ J/cm}^2$, $N_r = 200$ and varying displacement between $\varphi = -0.97$ and $\varphi = 0.71$. These images yield an overview about the characteristics of this experiment. As discussed in Chapter 2, the size of the irradiated area and number of pulses vary dependent on φ . For each value of φ , the scanner parameters have been optimised to obtain an accurate displacement. For these values of φ , which have been specifically selected to obtain this overview, no uniform surface was obtained.

Figure 5.11 shows SEM images after irradiation with two different peak fluences, $\phi_0 = 0.29 \text{ J/cm}^2$ and $\phi_0 = 0.39 \text{ J/cm}^2$, three different overlaps, $\varphi = 0.02$, $\varphi = 0.51$ and $\varphi = 0.80$, and $N_r = 20$. These images show the influence of varying lateral displacements on the spatial emergence of LIPSS. The lateral displacement influences the distribution of the spatial emergence. For this examples, three different surface morphologies have been obtained, a separated, as shown in Figure 5.11 second row left for $\varphi = 0.02$, an interconnected, as shown in Figure 5.11 second row middle for $\varphi = 0.51$, and a connected, as shown in Figure 5.11 second row right for $\varphi = 0.80$. The spatial emergence corresponds approximately to the irradiation profiles that were shown in Figures 2.4d to 2.4f.

In the dynamic experiment, the surface modifications, described in the section before, were recovered and no new types were found. In Figure 5.11 second row left, the initiation of SM-IV and SM-V can be observed. The morphologies produced at the displaced irradiated locations are spatially separated and the area in between appears to be untreated. This inhomogeneous distribution is characterised by the distance of displaced locations, the diameter of the spatial emergence and the size of the irradiated area.

In Figure 5.11 second row middle, the formation of SM-V was clearly found by an advanced splitting. Along the displacement directions, the spatial emergence extends

Figure 5.10: SEM images of the steel sample surface after irradiation with varying pulse overlap under DIC.

and morphologies start to connect. But in diagonal direction, less treated surface areas can be observed. For this pulse overlap an inhomogeneous and complex surface morphology was obtained. Interestingly, the orientation of LIPSS was found to be influenced by adjacent laser spots, where LIPSS seem to prefer to connect peaks and valleys.

In Figure 5.11 second row right, a homogeneous area was obtained, where no interruption due the displacements can be observed anymore. The structure type was difficult to define based on the visual impression from the microscopy images. But it seems as if the structure type has varied and the irregularity points to the back

Figure 5.11: SEM images of the steel sample surface after irradiation with varying pulse overlap under DIC. The inserts are doubled in size.

merged SM-IV type.

For $\phi_0 = 0.39 \text{ J/cm}^2$ and $\phi = 0.02$, an increased apparent diameter was obtained. This reduces the untreated space in between the irradiated locations. Although for $\phi = 0.02$ the formation of SM-IV was observed and nearly no SM-V initiation, for $\phi = 0.51$ the SM-V surround the peaks with a clear splitting. For $\phi = 0.80$ the back merged SM-IV type emerge und the SM-VI start to initiate. The variations due to the varying displacement, are in agreement with the observations, which have been made at lower fluence.

Whereas previously examples were presented for three different pulse overlaps,

Figure 5.12: SEM images of the steel sample surface after irradiation with varying pulse overlap under DIC. The inserts are doubled in size.

which yield a first overview of the spatial emergence, in the following an example is given which shows the transition from interconnected to connected in more detail. Figure 5.12 shows SEM images after irradiation with $\phi_0 = 0.79 \text{ J/cm}^2$, $N_r = 1$ and varying displacement between $\varphi = 0.66$ and $\varphi = 0.80$. Because the peak fluence was increased and the number of repetitions decreased, different surface modifications have been observed. Whereas Figure 5.12 top left shows the SM-III, the image Figure 5.12 bottom right shows the SM-IV. Again, the variation of the displacement was accompanied by a variation of the surface modification and distribution at the same time. In this example, the transition between interconnected and connected was between $\varphi = 0.71$ and $\varphi = 0.75$.

In the previous images, after increasing the pulse overlap one type of surface modification with an homogeneous distribution was obtained. These images were recorded in the centre of the irradiated area. Figure 5.13 shows SEM images after irradiation with $\phi_0 = 0.79 \text{ J/cm}^2$, $\varphi = 0.80$ and varying N_r . At the edge the accumulated fluence drops and different surface modifications were found, similar as in static. But here, their spatial emergence was found to be different on the edges of the irradiated area. At the edge parallel to their orientation, the LIPSS were found to propagate over a

Figure 5.13: SEM images of the steel sample surface after irradiation with varying N_r under DIC.

long distance compared to their period and the transition between different modifications were found perpendicular to their orientation. At the edge perpendicular to their orientation new periods were found to appear very regular and the transition between different modifications were found parallel to their orientation. These characteristics depend on the edge orientation and polarisation direction.

In the case of separated irradiated locations the spatial emergence and the morphological changes of the surface were found to be similar to static, as described in the section before. In the following a few examples are presented, for the interconnected morphologies. Figure 5.14 shows SEM images for varying number of repetitions at a pulse overlap of 0.51. With increasing N_r , the sequence of surface modifications was traversed as known from static at a similar peak fluence, starting with SM-III, over SM-IV to SM-VI. Except in Figures 5.14 top left, the images usually show multiple surface modifications at once. Often the orientation of LIPSS was found to be influenced by the next displaced location. As has been found in the previous section, the hole phenomenon was accompanied by material ablation in the micrometre range. Whereas under SIC an ablation crater has been formed, in the interconnected dynamic case a complex ablation pattern has been formed.

Figure 5.14: SEM images of the steel sample surface after irradiation with varying N_r under DIC. The inserts are doubled in size.

As the pulse overlap was even further decreased than this 0.51, e.g. down to 0.02, still structures were found to be interconnected for this particular peak fluence. Figure 5.15 top left shows a SEM image corresponding to this irradiation situation. But in contrast to the above mentioned interconnected case, where the surface modifications interconnects in the centre of a displaced location, here the outer surface modifications were interconnected due to the increased apparent diameter. In this case the

Figure 5.15: SEM images of the steel sample surface after irradiation with different parameters under DIC.

accumulated peak fluence addresses one morphology in the centre of the displaced locations and in between, the accumulated fluence was sufficient for the modification that follows below this fluence. Another example for such a case is shown in Figure 5.15 top right for the SM-IV and SM-VIII.

Again, for both examples the orientation of LIPSS were found to be influenced by the location of the connection. In some SEM images the orientation of the SM-IV has

Figure 5.16: SEM images of the steel sample surface after irradiation with varying peak fluence under DIC. The inserts are doubled in size.

been found to be influenced by the direction of scratches. One example is shown in Figure 5.15 second row left. Another influence on the orientation has been observed for SM-VI. For certain pulse overlaps the holes were aligned along the displacement direction. Figure 5.15 second row right, third row left and right shows examples. The displacement influences the periodicity and orientation of the holes. With increasing pulse overlap, the periodicity has been found to decrease. For lower pulse overlaps the orientations seems to be parallel to the displacement direction. If a homogeneous structure is obtained, the orientation was found to be aligned with the polarisation direction. One reason for this effect can be that the accumulated fluence domain of the holes was reached for lower pulse overlaps at the peak locations, so in the valleys of the ablation patterns.

In this section the observations have been described, which have been made in the microscopic analysis of the dynamically irradiated surface. Under DIC, the same surface modifications were found as under SIC. The lateral displacement extends the spatial emergence of LIPSS, if the accumulation of fluence is sufficient. This influences the distribution of the spatial emergence and creates separated, interconnected, connected and combined surface morphologies. The study of these surface morphologies

Figure 5.17: Fourier transform of the SEM images Figure 5.16 after irradiation with varying peak fluence. The images use a linear grey scale. The frequency domain was normalised by the norm of the wave vector, $2\pi/\lambda$. A high pass filter was applied, since these frequencies are mainly related to the ablation crater.

Figure 5.18: SEM images of the steel sample surface after irradiation with varying number of irradiation repetitions under DIC. The inserts are doubled in size.

is important, because they bear potential for applications. The variation of the displacement or overlap was accompanied by a variation of the surface modification and distribution at the same time. These effects are related to the variation of ϕ_a , Γ_p and Γ_d , as discussed in Subsections 2.4.2 and 3.3.1. Without determining the applied irradiation and the sequence of surface modifications, the experimental determination of irradiation parameters remains difficult. The transition between interconnected and connected was found to be between $\varphi = 0.71$ and $\varphi = 0.75$. This confirms that a homogeneous area of LIPSS can be obtained, if Γ_d is sufficiently small, as discussed in Subsections 2.4.2 and 3.3.4.

Figure 5.19: Fourier transform of SEM images Figure 5.18 after irradiation with varying number of irradiation repetitions. The images use a linear grey scale. The frequency domain was normalised by the norm of the wave vector, $2\pi/\lambda$. A high pass filter was applied, since these frequencies are mainly related to the ablation crater.

5.4.2 Spatial characteristics

This subsection is dedicated to the analysis of the spatial characteristics of the obtained LIPSS and their transitions with varying irradiation parameters. Therefore, specific SEM images have been selected and the Fourier transform of these images have been determined. This analysis was performed for a set of irradiation parameters, so that the transition between different LIPSS types was obtained.

Figure 5.16 shows SEM images of the steel sample surface after irradiation with

Figure 5.20: SEM images of the steel sample surface after irradiation with varying number of irradiation repetitions under DIC. The inserts are doubled in size.

$\varphi = 0.80$, $N_r = 20$ and varying peak fluence between $\phi_0 = 0.10$ J/cm² and $\phi_0 = 0.59$ J/cm². Figure 5.17 shows the corresponding Fourier transforms of these SEM images. This sequence shows the transition between SM-IV, SM-V and SM-VI. The transitions occur gradually over a range of conditions. The transitions are characterised by the observation of frequency components from multiple modifications or a spreading of frequencies. The FT analysis confirms the observations by microscopy and shows clearly which modifications are dominant. The periodicity of SM-IV was 797 nm at $\phi_0 = 0.12$ J/cm², of SM-V was 406 nm at $\phi_0 = 0.17$ J/cm² and of SM-IV was 756 nm at $\phi_0 = 0.59$ J/cm². The hole phenomenon could not be detected in the FT.

Figure 5.21: Fourier transform of SEM images in Figure 5.20 after irradiation with varying number of irradiation repetitions. The images use a linear grey scale. The frequency domain was normalised by the norm of the wave vector, $2\pi/\lambda$. A high pass filter was applied, since these frequencies are mainly related to the ablation crater.

Figure 5.18 shows SEM images of the sample surface after irradiation with $\phi_0 = 0.17 \text{ J/cm}^2$, $\varphi = 0.80$ and varying number of repetitions between $N_r = 6$ and $N_r = 250$. Figure 5.19 shows the corresponding Fourier transforms of these SEM images. This sequence shows the transitions between SM-IV, SM-V and SM-IV. The transitions were similar to those by changing the peak fluence. The periodicity of SM-IV was 908 nm at $N_r = 8$ and of SM-V was 406 nm at $N_r = 20$. For larger values of N_r , a large spread of spatial frequencies was obtained. The hole phenomenon could not be detected in the FT. Between $N_r = 8$ and $N_r = 15$ the periodicity of the SM-IV

Figure 5.22: SEM images of the steel sample surface after irradiation with varying number of irradiation repetitions under DIC.

was found to decrease from 908 nm to 794 nm. Between $N_r = 10$ and $N_r = 20$ the periodicity of the SM-V was found to decrease from 459 nm to 406 nm.

Figure 5.20 shows SEM images of the sample surface after irradiation with $\phi_0 = 0.79 \text{ J/cm}^2$, $\varphi = 0.80$ and varying number of repetitions between $N_r = 1$ and $N_r = 100$. Figure 5.21 shows the corresponding Fourier transforms of these SEM images. This sequence shows the transitions between SM-IV, SM-V and SM-VI. The transitions were different compared to the previous, as a result of a lower peak fluence. The sequence starts with the presence of SM-IV and SM-V, changes to SM-IV and ends at SM-VI. The spread of frequencies for the transition between SM-V and SM-VI was less pronounced at this fluence. The SM-IV were present for all values of N_r , but during variation of N_r they undergo morphological changes. Whereas at the beginning they form directly on the surface, with increasing N_r they form first in between the holes and later on the elevations of the grooves.

For this analysis, first the SEM images have been evaluated and second the FT images. In the first step the SM-V have not been identified. Their signature was not recognised until the evaluation of the FT images. At the repeated analysis of the SEM images, these structures were identified as dark areas on the SM-IV elevations.

Figure 5.23: Fourier transform of SEM images Figure 5.22, Figure 5.14 top right, Figure 5.12 top left, Figure 5.12 top right and Figure 5.12 bottom left. The images use a linear grey scale. The frequency domain was normalised by the norm of the wave vector, $2\pi/\lambda$. A high pass filter was applied, since these frequencies are mainly related to the ablation crater.

The SM-VI type form in two different stages, for lower values of N_r as randomly distributed holes and for higher values as periodic grooves. The latter state of the SM-VI type were clearly detected in the FT images. The periodicity of SM-IV was 848 nm at $N_r = 1$, of SM-V was 453 nm at $N_r = 1$ and of SM-VI was 3262 nm at $N_r = 80$. Between $N_r = 1$ and $N_r = 4$ the periodicity of SM-IV was found to decrease from 848 nm to 790 nm and of SM-V from 453 nm to 396 nm.

Figure 5.22 shows SEM images of the sample surface after irradiation with $\phi_0 = 0.10 \text{ J/cm}^2$, $\varphi = 0.80$ and varying number of repetitions between $N_r = 80$ and $N_r = 200$. Figures 5.23a to 5.23d show the corresponding Fourier transforms of these SEM images. The space and frequency domain show the formation of SM-IV and SM-VIII with varying N_r . With increasing N_r , these structures clearly developed, which was characterised by a reduced frequency spread in the Fourier domain. The periodicity of SM-IV was 511 nm at $N_r = 200$ and of SM-VIII was 976 nm at $N_r = 200$.

Figures 5.23e to 5.23h show the Fourier transform of the SEM images in Figure 5.14 top right, Figure 5.12 top left, Figure 5.12 top right and Figure 5.12 bottom left. The FT image 5.23e shows the frequency components of the three structures SM-III, SM-IV and SM-V at once. In this image a high pass filter was applied, because the

SM-III components were difficult to observe, due to the strong intensity of the SM-IV. The other three FT images show the formation of the SM-III with increasing φ . First only the structure SM-III has been observed, which develops with increasing N_r into SM-IV. The FT image for the SEM image at $\phi_0 = 0.79 \text{ J/cm}^2$, $\varphi = 0.80$ and $N_r = 1$, which follows with increasing φ , is shown in Figure 5.21a. Both modifications, SM-IV and SM-V, were found to be present.

In this subsection the spatial characteristics of LIPSS, obtained under HODIC, were analysed. The analysis has been performed for all observed LIPSS types and the results for the spatial periods were in agreement with the result obtained under SIC. It was also possible to obtain the transitions from one to another LIPSS type. Whereas under SIC, different structure types were usually present at once and a sharp transition in form of a threshold has been observed, under HODIC the sequence of morphologies is traversed gradually but with a smooth transition. The FT results of the LIPSS obtained under HODIC were found to be more clear than under SIC. The holes structures have not been observed under SIC but under HODIC. They seem to be stochastically distributed until they transform to grooves.

Within this experiment homogeneous areas of LIPSS have been obtained and these surfaces are of particular interest for applications. Because, in order to achieve a certain physical effect, often a large area of a particular structure with a specific spatial property, is required. Furthermore, the obtained structures, can be analysed over an extended area, which yields more information. However, a small change in the irradiation parameters can change the spatial characteristics.

The aim of this experiment was to investigate the spatial emergence of LIPSS. Therefore, a broad range of parameters have been screened. This offered the possibility to analyse the variation of the spatial characteristics together with the transitions of surface modifications. For further studies more values within the accumulated fluence domain of LIPSS would be interesting, in order to analyse the variation of their spatial properties in more detail. The Fourier analysis was found to be a good way to yield quantitative information about the spatial properties of LIPSS, such as periodicity and regularity. However, in this study a parameter for the spread of spatial frequencies was missing. Such a parameter, in combination with the suggested study of the intra domain properties, could yield the optimal irradiation conditions for specific LIPSS types.

5.4.3 Irradiation parameters

For the dynamic experiment the peak fluence was varied between $\phi_0 = 0.10 \text{ J/cm}^2$ and $\phi_0 = 1.58 \text{ J/cm}^2$ in 12 steps, the pulse overlap was varied between $\varphi = -0.97$ and $\varphi = 0.99$ in 25 steps and the number of repetitions covered the range $N_r = 1$ and $N_r = 2000$ in 24 steps. The values were selected such that a screening of the spatial emergence of LIPSS was obtained. The analysis of the irradiation parameters has been focused on the SM-V structure.

The observed surface modifications have been encoded by different colours and their observation has been denoted in the electronic datasheet for the dynamic experiment. Settings with no colour indicate that these images have not been archived for

later analysis. The colour grey denotes archived but not encoded images. Figures 5.24 and 5.25 show different cut-outs of the datasheet. Each individual cut-out presents a part of the irradiation parameters for a specific peak fluence. For these figures, those peak fluence values have been selected which contain information about the SM-V structure. The cut-outs show the applied pulse overlap on the abscissa (horizontal) and the number of irradiation repetitions on the ordinate (vertical).

The computer program for the irradiation model, see Section 2.3, has been used to obtain the relative accumulated peak fluence $\tilde{\Gamma}_p$, denoted in the table by G rel. These values have been multiplied with the peak fluence and yield the peak of the accumulated fluence Γ_0 , denoted in the table by G abs. The accumulated peak fluence Γ_p was obtained by a multiplication with N_r and is denoted within the matrix.

The assignment of observations to irradiation parameters yields an overview of the spatial emergence of LIPSS. With increasing peak fluence and number of irradiation repetitions different surface modifications have been observed. In the dynamic experiment the values for the peak fluence and number of irradiation repetitions were slightly different compared to static. However, under LODIC for comparable peak fluence values the same type and sequence of surface modifications as in static have been observed. This means at $\phi_0 = 0.10 \text{ J/cm}^2$ SM-VIII and SM-IV, at $\phi_0 = 0.17 \text{ J/cm}^2$ SM-IV, SM-V, SM-IV and SM-VI and at $\phi_0 = 0.79 \text{ J/cm}^2$ SM-I, SM-III, SM-IV and SM-VI. Furthermore, the surface modifications form in a similar range of N_r .

With increasing pulse overlap four different phenomena have been observed. First the repetition threshold, at which LIPSS spatially emerge, have been found to decrease, second the range of repetitions has been found to decrease, third the domain location has been found at an increased accumulated fluence and fourth the size of the accumulated fluence domain has been found to increase. These variations were found to be similar for the different peak fluence values and for different surface modifications. Only for $\phi_0 = 0.10 \text{ J/cm}^2$ the SM-VIII showed a stronger decrease in repetitions from $\varphi = 0.02$ to $\varphi = 0.51$. But, at $\varphi = 0.02$ between $N_r = 300$ and $N_r = 450$ the surface was covered by contaminations. For these values, an unambiguous identification of the structure was not possible.

In order to underline these observations some boundary values have been extracted from the datasheet of the static and dynamic irradiation experiment and brought together in Table 5.6. This table shows the agreement between SIC and LODIC domain boundaries, if N_{th} is compared for different ϕ_0 , the decrease of the repetition threshold N_{th} from LODIC to HODIC and the shift of the domain location, characterised by Γ_{th} , from LODIC to HODIC. Another variation, which can be observed in Figures 5.24 and 5.25 if the different cut-outs are compared, is that with increasing peak fluence, not only the different surface modifications emerge, also the number of repetition and accumulated fluence domain location, e.g. of SM-V, decrease for all pulse overlap values.

Afterwards, the experimental values for the lower and upper boundary of the irradiation repetitions have been compared to values obtained from Approximations 3.38 and 3.41. Figures 5.26a to 5.26c show the dependence of the number of irradiation repetitions on the pulse overlap for three different peak fluences, Figure 5.26a $\phi_0 = 0.15 \text{ J/cm}^2$, Figure 5.26b $\phi_0 = 0.17 \text{ J/cm}^2$ and Figure 5.26c $\phi_0 = 0.20 \text{ J/cm}^2$. The

OL	-0.97	-0.48	0.02	0.11	0.21	0.31	0.41	0.51	0.56	0.61	0.66	0.70	0.75	0.80	0.85	0.90	0.91	0.92	0.93	0.94	0.95	0.96	0.97	0.98	0.99
G rel	1.00	1.00	1.00	1.01	1.03	1.09	1.26	1.66	2.02	2.54	3.31	4.50	6.49	10.13	18.02	40.54	50.05	63.34	82.73	112.60	162.15	253.35	450.41	1013.40	4053.70
Nr/G abs	0.10	0.10	0.10	0.10	0.10	0.11	0.13	0.17	0.21	0.26	0.34	0.46	0.66	1.03	1.84	4.14	5.11	6.46	8.44	11.49	16.54	25.85	45.96	103.40	413.60
1	0.10	0.10	0.10	0.10	0.10	0.11	0.13	0.17	0.21	0.26	0.34	0.46	0.66	1.03	1.84	4.14	5.11	6.46	8.44	11.49	16.54	25.85	45.96	103.40	413.60
2	0.20	0.20	0.20	0.21	0.21	0.22	0.26	0.34	0.41	0.52	0.68	0.92	1.32	2.07	3.68	8.27	10.21	12.92	16.88	22.98	33.09	51.70	91.91	206.80	
4	0.41	0.41	0.41	0.41	0.42	0.45	0.51	0.68	0.82	1.04	1.35	1.84	2.65	4.14	7.35	16.54	20.42	25.85	33.76	45.96	66.18	103.40	183.82	413.59	
6	0.61	0.61	0.61	0.62	0.63	0.67	0.77	1.02	1.23	1.55	2.03	2.76	3.97	6.20	11.03	24.82	30.64	38.77	50.64	68.93	99.26	155.10	275.73		
8	0.82	0.82	0.82	0.82	0.84	0.89	1.03	1.36	1.65	2.07	2.70	3.68	5.29	8.27	14.71	33.09	40.85	51.70	67.53	91.91	132.35	206.80			
10	1.02	1.02	1.02	1.03	1.05	1.11	1.29	1.70	2.06	2.59	3.38	4.60	6.62	10.34	18.38	41.36	51.06	64.62	84.41	114.89	165.44	258.50			
15	1.53	1.53	1.53	1.54	1.57	1.67	1.93	2.54	3.09	3.88	5.07	6.89	9.51	15.51	27.57	62.04	76.59	96.94	126.61	172.33	248.16				
20	2.04	2.04	2.04	2.06	2.10	2.23	2.57	3.39	4.12	5.18	6.75	9.19	13.24	20.68	36.76	82.72	102.12	129.25	168.82	229.78					
40	4.08	4.08	4.09	4.11	4.20	4.46	5.15	6.78	8.23	10.35	13.51	18.38	26.47	41.36	73.53	165.44	204.25	258.50	337.63						
60	6.12	6.12	6.13	6.17	6.29	6.68	7.72	10.17	12.35	15.53	20.26	27.57	39.71	62.04	110.29	248.16	306.37								
80	8.16	8.16	8.18	8.22	8.39	8.91	10.29	13.56	16.46	20.71	27.01	36.76	52.94	82.72	147.06										
100	10.20	10.20	10.22	10.28	10.49	11.14	12.86	16.95	20.58	25.89	33.77	45.96	66.18	103.40	183.82										
150	15.30	15.30	15.33	15.42	15.74	16.71	19.30	25.43	30.86	38.83	50.65	68.93	99.26	155.10	275.73										
200	20.41	20.41	20.44	20.56	20.98	22.28	25.73	33.90	41.15	51.77	67.53	91.91	132.35	206.80											
250	25.51	25.51	25.55	25.70	26.23	27.85	32.16	42.38	51.44	64.72	84.42	114.89	165.44	258.50											
300	30.61	30.61	30.66	30.84	31.47	33.42	38.59	50.86	61.73	77.66	101.30	137.87	198.53												
350	35.71	35.71	35.77	35.98	36.72	38.99	45.02	59.33	72.02	90.60	118.19	160.85	231.62												
400	40.81	40.81	40.88	41.12	41.96	44.56	51.46	67.81	82.31	103.55	137.07	183.82													
450	45.91	45.91	45.99	46.26	47.21	50.12	57.99	76.29	92.59	116.49	151.95	206.80													
500	51.02	51.02	51.10	51.40	52.45	55.69	64.32	84.76	102.88	129.43	168.84	229.78													
600	61.22	61.22	61.32	61.68	62.95	66.83	77.18	101.71	123.46	155.32	202.60														
800	81.63	81.63	81.76	82.24	83.93	89.11	102.91	135.62	164.61	207.09	270.14														
1000	102.03	102.03	102.20	102.50	104.91	111.39	128.64	169.52	205.77	258.86															
2000	204.06	204.06	204.41	205.59	209.82	222.78	257.28	339.05	411.53																

(a)

OL	-0.97	-0.48	0.02	0.11	0.21	0.31	0.41	0.51	0.56	0.61	0.66	0.70	0.75	0.80	0.85	0.90	0.91	0.92	0.93	0.94	0.95	0.96	0.97	0.98	0.99
G rel	1.00	1.00	1.00	1.01	1.03	1.09	1.26	1.66	2.02	2.54	3.31	4.50	6.49	10.13	18.02	40.54	50.05	63.34	82.73	112.60	162.15	253.35	450.41	1013.40	4053.70
Nr/G abs	0.12	0.12	0.12	0.12	0.13	0.13	0.16	0.20	0.25	0.31	0.41	0.56	0.80	1.25	2.22	5.00	6.17	7.81	10.20	13.89	20.00	31.25	55.56	125.00	500.00
1	0.12	0.12	0.12	0.12	0.13	0.13	0.16	0.20	0.25	0.31	0.41	0.56	0.80	1.25	2.22	5.00	6.17	7.81	10.20	13.89	20.00	31.25	55.56	125.00	500.00
2	0.25	0.25	0.25	0.25	0.25	0.27	0.31	0.41	0.50	0.63	0.82	1.11	1.60	2.50	4.44	10.00	12.35	15.62	20.41	27.78	40.00	62.50	111.11	250.00	
4	0.49	0.49	0.49	0.50	0.51	0.54	0.62	0.82	1.00	1.25	1.63	2.22	3.20	5.00	8.89	20.00	24.69	31.25	40.82	55.56	80.00	125.00	222.22	499.99	
6	0.74	0.74	0.74	0.75	0.76	0.81	0.93	1.23	1.49	1.88	2.45	3.33	4.80	7.50	13.33	30.00	37.04	46.87	61.22	83.33	120.00	187.50	333.33		
8	0.99	0.99	0.99	0.99	1.01	1.08	1.24	1.64	1.99	2.50	3.27	4.44	6.40	10.00	17.78	40.00	49.38	62.50	81.63	111.11	160.00	250.00			
10	1.23	1.23	1.24	1.24	1.27	1.35	1.56	2.05	2.49	3.13	4.08	5.56	8.00	12.50	22.22	50.00	61.73	78.12	102.04	138.89	200.00	312.50			
15	1.85	1.85	1.85	1.86	1.90	2.02	2.33	3.07	3.71	4.69	6.12	8.31	12.00	18.75	33.33	75.00	92.59	117.19	153.06	208.33	320.00				
20	2.47	2.47	2.47	2.49	2.54	2.69	3.11	4.10	4.98	6.26	8.16	11.11	16.00	25.00	44.44	100.00	123.46	156.25	204.08	277.78					
40	4.93	4.93	4.94	4.97	5.07	5.39	6.22	8.20	9.95	12.52	16.33	22.22	32.00	50.00	88.89	200.00	246.91	312.50	408.16						
60	7.40	7.40	7.41	7.46	7.61	8.08	9.33	12.30	14.93	18.78	24.49	33.33	48.00	75.00	133.33	300.00	370.37								
80	9.87	9.87	9.88	9.94	10.15	10.77	12.44	16.40	19.90	25.04	32.66	44.44	64.00	100.00	177.78										
100	12.33	12.33	12.36	12.43	12.68	13.47	15.55	20.48	24.88	31.29	40.82	55.56	80.00	125.00	222.22										
150	18.50	18.50	18.53	18.64	19.02	20.20	23.33	30.74	37.31	46.94	61.23	83.33	120.00	187.50	333.33										
200	24.67	24.67	24.71	24.85	25.36	26.93	31.10	40.99	49.75	62.59	81.64	111.11	160.00	250.00											
250	30.84	30.84	30.89	31.07	31.71	33.66	38.88	51.23	62.19	78.23	102.05	138.89	200.00	312.50											
300	37.00	37.00	37.07	37.28	38.05	40.40	46.65	61.48	74.63	93.88	122.46	166.67	240.00												
350	43.17	43.17	43.24	43.49	44.39	47.13	54.43	71.73	87.06	109.53	142.87	194.45	280.00												
400	49.34	49.34	49.42	49.71	50.73	53.86	62.21	81.98	99.50	125.18	163.28	222.22													
450	55.51	55.51	55.60	55.92	57.07	60.60	69.98	92.22	111.94	140.82	183.70	250.00													
500	61.67	61.67	61.78	62.14	63.41	67.33	77.76	102.47	124.38	156.47	204.11	277.78													
600	74.01	74.01	74.13	74.56	76.09	80.79	93.31	122.96	149.25	187.76	244.93														
800	98.68	98.68	98.84	99.42	101.46	107.72	124.41	163.95	199.00	250.35	326.57														
1000	123.35	123.35	123.56	124.27	126.82	134.66	155.51	204.94	248.75	312.94															
2000	246.69	246.69	247.11	248.54	253.65	269.31	311.03	409.88	497.50																

(b)

OL	-0.97	-0.48	0.02	0.11	0.21	0.31	0.41	0.51	0.56	0.61	0.66	0.70	0.75	0.80	0.85	0.90	0.91	0.92	0.93	0.94	0.95	0.96	0.97	0.98	0.99
G rel	1.00	1.00	1.00	1.01	1.03	1.09	1.26	1.66	2.02	2.54	3.31	4.50	6.49	10.13	18.02	40.54	50.05	63.34	82.73	112.60	162.15	253.35	450.41	1013.40	4053.70
Nr/G abs	0.15	0.15	0.15	0.15	0.15	0.16	0.19	0.24	0.30	0.37	0.49	0.66	0.96	1.49	2.66	5.98	7.38	9.34	12.20	16.60	23.90	37.35	66.40	149.40	597.61
1	0.15	0.15	0.15	0.1																					

OL	-0.97	-0.48	0.02	0.11	0.21	0.31	0.41	0.51	0.56	0.61	0.66	0.70	0.75	0.80	0.85	0.90	0.91	0.92	0.93	0.94	0.95	0.96	0.97	0.98	0.99
G rel	1.00	1.00	1.00	1.01	1.03	1.09	1.26	1.66	2.02	2.54	3.31	4.50	6.49	10.13	18.02	40.54	50.65	63.34	82.73	112.60	162.15	253.35	450.41	1013.40	4053.70
Nr/G abs	0.17	0.17	0.17	0.18	0.18	0.19	0.22	0.29	0.35	0.44	0.58	0.78	1.13	1.77	3.14	7.06	8.72	11.04	14.42	19.62	28.26	44.15	78.49	176.60	706.41
1	0.17	0.17	0.17	0.18	0.18	0.19	0.22	0.29	0.35	0.44	0.58	0.78	1.13	1.77	3.14	7.06	8.72	11.04	14.42	19.62	28.26	44.15	78.49	176.60	706.41
2	0.35	0.35	0.35	0.36	0.38	0.44	0.58	0.70	0.88	1.15	1.57	2.26	3.53	6.28	14.13	17.44	22.07	28.83	39.24	56.51	88.30	156.98	335.10	706.41	
4	0.70	0.70	0.70	0.72	0.76	0.88	1.16	1.41	1.77	2.31	3.14	4.52	7.06	12.56	28.26	34.88	44.15	57.67	78.49	113.02	176.60	313.96	706.41		
6	1.05	1.05	1.05	1.08	1.14	1.32	1.74	2.11	2.65	3.46	4.71	6.78	10.60	18.84	42.88	52.33	66.22	86.50	117.73	169.54	282.00	535.24	706.41		
8	1.39	1.39	1.40	1.40	1.43	1.52	1.76	2.32	2.81	3.54	4.61	6.28	9.04	14.13	25.12	56.51	69.77	88.30	115.33	156.98	253.24	478.64	706.41		
10	1.74	1.74	1.75	1.76	1.79	1.90	2.20	2.90	3.51	4.42	5.77	7.85	11.30	17.66	31.40	70.64	87.21	110.37	144.16	196.22	282.56	441.26	706.41		
15	2.61	2.61	2.62	2.63	2.69	2.85	3.30	4.34	5.27	6.63	8.65	11.77	16.95	26.49	47.09	105.96	130.81	165.56	216.23	284.33	423.44	706.41			
20	3.49	3.49	3.49	3.51	3.58	3.80	4.39	5.79	7.03	8.84	11.53	15.70	22.60	35.82	62.79	141.28	174.42	220.75	280.35	392.45	706.41				
40	6.97	6.97	6.98	7.02	7.17	7.61	8.29	11.58	14.06	17.68	23.07	32.40	45.21	70.64	122.58	253.24	322.56	423.44	535.24	706.41					
60	10.46	10.46	10.47	10.53	10.75	11.41	13.18	17.37	21.09	26.53	34.60	47.09	67.81	105.96	188.37	322.56	423.44	535.24	706.41						
80	13.94	13.94	13.96	14.05	14.33	15.22	17.58	23.16	28.11	35.37	46.14	62.79	90.42	141.28	251.10	423.44	535.24	706.41							
100	17.43	17.43	17.46	17.56	17.92	19.02	21.97	28.95	35.14	44.21	57.67	78.49	113.02	176.60	313.96	535.24	706.41								
150	26.14	26.14	26.18	26.34	26.88	28.54	32.96	43.43	52.72	66.32	86.51	117.73	169.54	282.00	478.64	706.41									
200	34.85	34.85	34.91	35.11	35.84	38.05	43.94	57.91	70.29	88.42	115.34	156.98	220.75	358.21	706.41										
250	43.57	43.57	43.64	43.89	44.79	47.56	54.93	72.38	87.86	110.53	144.18	196.22	282.56	441.26	706.41										
300	52.28	52.28	52.37	52.67	53.75	57.07	65.91	86.86	105.43	132.64	173.02	238.47	330.01	535.24	706.41										
350	60.99	60.99	61.10	61.45	62.71	66.58	76.90	101.34	123.00	154.74	201.85	274.71	392.45	706.41											
400	69.70	69.70	69.82	70.23	71.67	76.10	87.88	115.81	140.57	176.85	230.69	313.96	441.26	706.41											
450	78.42	78.42	78.55	79.01	80.63	85.61	98.87	130.29	159.15	195.52	259.52	352.79	535.24	706.41											
500	87.13	87.13	87.28	87.78	89.59	95.12	109.85	144.77	175.72	221.08	288.36	392.45	706.41												
600	104.56	104.56	104.74	105.34	107.51	114.15	131.83	173.76	210.86	262.21	346.03	478.64	706.41												
800	139.41	139.41	139.63	140.46	143.35	152.19	175.77	231.45	281.15	358.21	461.38	706.41													
1000	174.26	174.26	174.57	176.07	180.24	190.24	215.37	284.41	351.43	442.12	706.41														
2000	348.52	348.52	349.12	351.14	358.35	380.48	439.42	579.07	702.87	706.41															

(a)

OL	-0.97	-0.48	0.02	0.11	0.21	0.31	0.41	0.51	0.56	0.61	0.66	0.70	0.75	0.80	0.85	0.90	0.91	0.92	0.93	0.94	0.95	0.96	0.97	0.98	0.99
G rel	1.00	1.00	1.00	1.01	1.03	1.09	1.26	1.66	2.02	2.54	3.31	4.50	6.49	10.13	18.02	40.54	50.65	63.34	82.73	112.60	162.15	253.35	450.41	1013.40	4053.70
Nr/G abs	0.20	0.20	0.20	0.20	0.20	0.21	0.23	0.33	0.40	0.50	0.65	0.88	1.27	1.99	3.53	7.95	9.82	12.42	16.23	22.09	31.81	49.70	88.36	198.80	795.21
1	0.20	0.20	0.20	0.20	0.20	0.21	0.23	0.33	0.40	0.50	0.65	0.88	1.27	1.99	3.53	7.95	9.82	12.42	16.23	22.09	31.81	49.70	88.36	198.80	795.21
2	0.39	0.39	0.39	0.40	0.40	0.41	0.49	0.65	0.79	1.00	1.30	1.77	2.54	3.98	7.07	15.00	19.63	24.85	32.46	44.18	63.62	99.40	176.71	397.59	
4	0.78	0.78	0.79	0.79	0.81	0.86	0.99	1.30	1.58	1.99	2.60	3.53	5.09	7.95	14.14	31.81	39.27	49.70	64.91	88.36	127.23	198.80	325.47	795.19	
6	1.18	1.18	1.18	1.19	1.21	1.28	1.48	1.96	2.37	2.99	3.90	5.30	7.63	11.93	21.21	47.71	58.90	74.55	97.37	132.53	190.85	296.26	530.17		
8	1.57	1.57	1.57	1.58	1.61	1.71	1.98	2.61	3.16	3.98	5.19	7.07	10.18	15.90	28.27	63.62	78.54	99.40	129.83	176.71	251.40	397.59	795.19		
10	1.96	1.96	1.97	1.98	2.02	2.14	2.47	3.26	3.96	4.98	6.49	8.84	12.72	19.88	35.34	79.52	98.17	124.25	162.29	220.91	318.08	497.00	795.19		
15	2.94	2.94	2.94	2.95	3.03	3.21	3.71	4.89	5.93	7.48	9.74	13.28	19.08	29.62	53.01	119.28	147.26	186.37	244.44	325.47	441.26	795.19			
20	3.92	3.92	3.93	3.95	4.03	4.28	4.95	6.52	7.91	9.95	12.98	17.67	25.48	39.76	70.68	159.04	196.35	245.56	324.57	441.26	795.19				
40	7.85	7.85	7.86	7.91	8.07	8.57	9.89	13.04	15.82	19.91	25.97	35.34	50.89	79.52	141.37	318.08	392.69	497.00	641.26	795.19					
60	11.77	11.77	11.79	11.86	12.10	12.85	14.84	19.56	23.74	29.86	38.95	53.01	76.34	122.05	210.86	478.64	535.24	706.41							
80	15.69	15.69	15.72	15.81	16.14	17.13	19.79	26.07	31.94	39.86	51.04	70.68	101.79	159.04	282.00	535.24	706.41								
100	19.62	19.62	19.65	19.76	20.17	21.42	24.73	32.59	39.56	49.77	64.92	88.36	127.23	198.80	325.47	535.24	706.41								
150	29.43	29.43	29.48	29.65	30.26	32.12	37.10	48.89	59.34	74.65	97.38	132.53	190.85	296.26	530.13	706.41									
200	39.23	39.23	39.30	39.53	40.34	42.83	49.47	65.19	79.12	99.54	129.84	176.71	251.40	397.59	706.41										
250	49.04	49.04	49.13	49.41	50.43	53.54	61.83	81.48	98.90	124.42	162.30	220.91	318.08	497.00	706.41										
300	58.85	58.85	58.95	59.29	60.51	64.25	74.20	97.78	118.68	149.31	194.77	263.07	367.07	535.24	706.41										
400	68.66	68.66	68.78	69.17	70.60	74.95	86.57	114.08	138.46	174.19	227.23	309.25	445.32	706.41											
450	78.47	78.47	78.60	79.06	80.68	85.66	98.93	130.37	159.25	199.09	259.69	353.43	535.24	706.41											
500	88.28	88.28	88.43	88.90	90.77	96.37	111.30	146.67	178.01	223.08	292.15	397.60	535.24	706.41											
600	98.08	98.08	98.25	98.85	100.85	107.08	123.66	162.97	197.81	248.85	324.61	441.78	706.41												
800	139.41	139.41	139.63	140.46	143.35	152.19	175.77	231.45	281.15	358.21	461.38	706.41													
1000	174.26	174.26	174.57	176.07	180.24	190.24	215.37	284.41	351.43	442.12	706.41														
2000	348.52	348.52	349.12	351.14	358.35	380.48	439.42	579.07	702.87	706.41															

(b)

OL	-0.97	-0.48	0.02	0.11	0.21	0.31	0.41	0.51	0.56	0.61	0.66	0.70	0.75	0.80	0.85	0.90	0.91	0.92	0.93	0.94	0.95	0.96	0.97	0.98	0.99
G rel	1.00	1.00	1.00	1.01	1.03	1.09	1.26	1.66	2.02	2.54	3.31	4.50	6.49	10.13	18.02	40.54	50.65	63.34	82.73	112.60	162.15	253.35	450.41	1013.40	

Table 5.6: Selection of boundary values for the spatial emergence of SM-V denoted by different threshold parameters. These data are extracted from the datasheets, see Figures 5.8, 5.24 and 5.25.

Condition	ϕ_0	φ	$N_{th}(\phi)$	$\Delta N_{th}(\phi)$	$\Gamma_{th}(\phi)$	$\Delta \Gamma_{th}(\phi)$
-	J/cm ²	-	-	-	J/cm ²	J/cm ²
LODIC	0.15	-0.97	60	90	8.85	13.27
HODIC	0.15	0.80	15	25	22.41	37.35
SIC	0.16	-	40	60	6.43	9.65
LODIC	0.17	-0.97	40	60	6.97	10.46
HODIC	0.17	0.80	10	10	17.66	17.66
SIC	0.19	-	30	30	5.57	5.57
LODIC	0.20	-0.97	40	40	7.85	7.85
HODIC	0.20	0.80	8	7	15.90	13.92
SIC	0.21	-	20	20	4.20	4.20

dash-dot lines represent the data from Approximation 3.38, denoted by prox(1), for a central domain value, the dashed lines from Approximation 3.41, denoted by prox(2), for a central domain value, the asterisk from the measurement of the lower boundary, denoted by exp low, and the plus from the measurement of the upper boundary, denoted by exp up.

For the three different peak fluences, the values from the approximations follow the course of the experimental values. The required number of irradiation repetitions decreases with increasing pulse overlap for all curves. Between a pulse overlap of -1 and 0.5, the theoretical values lie in between the two boundaries. Above a pulse overlap of 0.5, the theoretical values of Approximation 3.38 lie below the measurement points for the lower domain boundary and of Approximation 3.41 above the measurement points for the upper domain boundary. In the range $N_r = 1$ to $N_r = 10$ the deviation is about 2 repetitions. The experimental values for N_r have been determined with a step size of 2 between $N_r = 2$ and $N_r = 10$, of 5 between $N_r = 10$ and $N_r = 20$, of 20 between $N_r = 20$ and $N_r = 100$ and of 50 between $N_r = 100$ and $N_r = 200$. Please note that these figures are presented in a semi-logarithmic scale. Given the experimental step size, experimental results and approximations are over the whole range of φ in agreement.

The aim of the static and dynamic experiment was to investigate the spatial emergence of LIPSS. This has been done with a set of varying irradiation parameters, which were used to screen the domain of LIPSS. Within the dynamic experiment the number of irradiation repetitions were found to decrease and the domain locations to shift to higher fluence values with increasing pulse overlap. Both effects are in agreement with the results presented in Chapter 3. The shift of the domain location due to the decreasing average individual fluence in the context of the varying individual fluence has not been discussed in literature before.

Since this shift is relevant for the application of LIPSS, this effect requires more

Figure 5.26: Simulation and measurement results for the dependence of the number of irradiation repetitions on the pulse overlap under 2d-DIC for three different peak fluences, (a) $\phi_0 = 0.15$ J/cm², (b) $\phi_0 = 0.17$ J/cm² and (c) $\phi_0 = 0.20$ J/cm².

attention. Therefore, one additional dynamic experiment has been performed in order to analyse the shift of the domain location. In this experiment different peak fluences were applied for the same range of varied pulse overlap as in the dynamic experiment before. But in contrast to the dynamic experiment before, where N_r has been selected to screen the spatial emergence, in this experiment N_r has been varied according

Figure 5.27: SEM images of the steel sample surface after irradiation with constant accumulated peak fluence under DIC. The inserts are doubled in size.

to Approximation 3.38 to satisfy a particular accumulated peak fluence value. The expectation was that if while increasing the pulse overlap only the Gaussian accumulation is compensated, the structure gradually disappears on the surface due to the shift of the domain location.

Figure 5.27 shows SEM images of a steel sample surface after irradiation with $\phi_0 = 0.08$ J/cm², constant accumulated peak fluence of $\Gamma_p = 156$ J/cm², varying pulse overlap between $\varphi = -0.96$ and $\varphi = 0.93$ and varying number of irradiation repetitions between $N_r = 2000$ and $N_r = 24$. At the initial value $\varphi = -0.96$, see Figure 5.27 top row left, the SM-VIII was found to emerge, within the irradiated area.

Figure 5.28: SEM images of the steel sample surface after irradiation with constant accumulated peak fluence under DIC. The inserts are doubled in size.

The upper boundary was slightly exceeded, which was indicated by the emergence of SM-IV. Although the accumulated peak fluence was kept nearly constant, with increasing pulse overlap the SM-VIII were found to disappear, compare Figure 5.27 $\varphi = -0.96$ to $\varphi = 0.75$. From low to high pulse overlap, first the upper domain boundary disappears and then the lower.

This effect has been found multiple times. Another example is shown in Figure 5.28 for $\phi_0 = 0.15$ J/cm², constant accumulated peak fluence of $\Gamma_p = 29$ J/cm², varying pulse overlap between $\varphi = -0.96$ and $\varphi = 0.91$ and varying number of irradiation repetitions between $N_r = 200$ and $N_r = 4$. Even though this example shows

another type of LIPSS, the same effect can be observed. At the initial value $\varphi = -0.96$, compare Figure 5.28 top row left, the SM-V was found to emerge, within the irradiated area. The upper boundary was slightly exceeded, which was indicated by the emergence of the back merged SM-IV. With increasing pulse overlap, compare Figure 5.28 $\varphi = -0.96$ to $\varphi = 0.91$, the SM-V were found to disappear, but in this case not completely. In particular, the upper domain boundary disappears from $\varphi = -0.96$ to $\varphi = 0.66$. The lower domain boundary can be still observed at $\varphi = 0.91$.

Discussion

The static experiment, yields the surface modifications, their sequence and domain locations, for the given laser material combination. These modifications have also been found in the dynamic experiment. In static it was possible to screen the domain with a small interval for the irradiation parameters and to perform the experiment on a small area. Therefore, the domain location can be determined, in terms of accumulated fluence, very accurately and efficiently. The domain locations determined in static were found to be in agreement with the domain locations at low pulse overlaps in dynamic, compare Table 5.6.

In static the material has been found to show incubation properties and a decrease of the individual fluence threshold with increasing pulse number has been observed for all LIPSS. The incubation parameter has been determined for SM-I and SM-III and a value of $\xi = 0.82$ has been found. The course of the incubation curve has been found in agreement with Jee's incubation law for the given laser-material combination. This law and the associated parameter ξ has been applied to determine approximated values for N_r of the SM-V. The incubation parameter is a material parameter and should also be applicable for other surface modifications. However, a study which confirms the independence of this parameter on the surface modification at which this parameter was determined, is lacking in literature.

This experimental study shows the importance of incubation for machining LIPSS areas. With increasing peak fluence the accumulated fluence domain locations have been found to decrease for all pulse overlaps and surface modifications. This can be attributed to the incubation properties of the material. Furthermore, with increasing pulse overlap and constant peak fluence the accumulated fluence domain locations have been found to increase. Since it has been shown in Chapter 2 that the average individual fluence decreases at the same time also this effect can be attributed to the incubation properties of the material. Incubation also determines the domain locations under DIC and they were found to vary dependent on the irradiation parameters. If the role of incubation would not be considered while determining irradiation parameters, it would be simply unknown that the irradiation repetitions have to be decreased while increasing the peak fluence or vice versa. If the boundaries are determined in the static experiment, this already yields an approximation for the dynamic irradiation parameters, because each different peak fluence value is accompanied with a different LODIC domain location.

During this experiment it has been found that the structuring process was accompanied by material ablation. This contaminates the structures on the surface and requires an adequate cleaning procedure. Next to the quality of the surface, the re-

cast might influence the energy deposition during the structuring process. This could also be a reason why more accumulated fluence was required and could also explain, at least partially, the shift of the domain location. An indication for structured areas which were influenced by recast have been found only a few times in the SEM, at very high peak fluences, very high pulse overlaps or high number of irradiation repetitions. This was usually the case for the SM-VI, which were holes and grooves with an ablation depth in the micrometre range. However, the spatial emergence and shift of the domain location were investigated for lower peak fluences and on the SM-V structure, which forms close to the untreated surface and shows a minor ablation. The SEM images of the analysed regime showed properly structured surfaces and the shift of the domain location. Based on the result from this experiment, it is unlikely that the contaminations are the main cause of the domain shift. Nevertheless, the influence of recast on the structuring process is an interesting point for further research and is also very relevant for applications.

In Figures 5.26a to 5.26c Approximation 3.38 yields values, close to the experimental values. Under LODIC and MODIC the values were within the measured domain and for HODIC they were slightly lower. This approximation uses the domain location determined in static, or in this example at a low pulse overlap, and calculates the number of irradiation repetitions, without any individual fluence boundary, based on each individual fluence contribution. In this way, the pulse overlap dependent fluence accumulation was considered. This approximation does not consider the shift of the domain location. Therefore, this approximation can yield values which are, at HODIC, slightly lower than required. The effect of the shift has been shown in Figures 5.27 and 5.28. However, the domain location of SM-V were found to shift about a factor of 3. Compared to the size of the accumulated fluence domain at low pulse overlaps, the domain stays in a similar order of magnitude. This means for the determination of irradiation parameters, that with increasing pulse overlap the effort to find the shifted domain is small. In contrast, the finding of the domain with an increase of the pulse overlap without the knowledge from the irradiation model is difficult, because the number of irradiation repetitions can vary up to a factor of 100, dependent on the initial domain location.

In Figures 5.26a to 5.26c Approximation 3.41 yields values, close to the experimental values. Under LODIC and MODIC the values were within the measured domain and for HODIC they were slightly higher. This approximation applies a contribution determined domain location and calculates the number of irradiation repetitions, without any individual fluence boundary, based on each relative incubation based irradiation contribution. In this way the pulse overlap dependent fluence accumulation and shift of the domain location were considered. Without analysing the accumulated fluence, compare Chapter 2, the shift of the domain location would not have been observed, compare Chapter 3. This approximation improves the previous one, by considering this effect. However, for HODIC the shift was estimated too strong, which means that the individual contributions were estimated too low. This is probably due to the limited applicability of the incubation law under DIC, as discussed in Chapter 3. This could be improved, by a new incubation law, which considers a non-constant individual fluence. Nevertheless, the deviation at HODIC

is about 2 number of irradiation repetitions, which is small compared to the initial starting value of $N_r = 105$.

Despite these small deviations, LIPSS could still be observed for both approximations. Because the domain locations under DIC were determined with a certain step size for the number of irradiation repetitions. The step size influences the experimental determination of the domain boundaries for the input and validation data. But, the step size also influences the amount of experimental data. The aim of the experiment was to screen the spatial emergence of different type of LIPSS for different peak fluences and a broad range of pulse overlaps, which results in a large amount of data. Therefore, the step size had to be limited in order to limit the amount of experimental data. In practice the input data are determined more accurately, because only a specific value ϕ or N_p is required. Furthermore, the deviations can also be caused by other experimental limitations, as the uncertainties of the incubation parameter, beam radius and lateral displacements, which could result in a different accumulation with increasing pulse overlap. Overall, the static and dynamic experiments are consistent, with respect to the observed surface modifications and domain boundaries. The approximations are in agreement with the experimental values. In fact, both approximations together give a range for the experimental values. These experiments show, that with increasing pulse overlap, the reduction of the irradiation repetitions is a way to find the spatial emergence of LIPSS. The approximations are sufficient for an estimation of irradiation parameters for homogeneous areas of LIPSS.

These experiments about the spatial emergence of LIPSS have been followed by an additional dynamic experiment investigating the shift of the domain location. In this experiment, LIPSS have been found to disappear if the pulse overlap was increased and the accumulated fluence was constant, see Figures 5.27 and 5.28. This seems to be related to the shift of the domain location with increasing pulse overlap, as indicated in Figure 5.26. This experiment has been performed on a new sample and at very low energy. Nearly no influence of contamination has been observed. Especially for the shift of the SM-VIII domain location, for which the surface was hardly changed, contamination can be ruled out as a reason for the shift. The results of this experiment confirm the theoretical considerations presented in Chapter 3 and the results of the previous dynamic experiment. In order to maintain the accumulated fluence constant, Approximation 3.38 has been applied. Based on this approximation, the observation of LIPSS at high pulse overlaps depends on the incubation factor, addressed domain location and size of the domain. However, this approximation is simple and practicable, because for this approximation only N_p needs to be divided by $\tilde{\Gamma}_p$. Usually, N_p is obtained from the static experiment and for $\tilde{\Gamma}_p$ often only a few values are applied in practice, see Table 2.3. Furthermore, if the domain is large enough and a certain value is addressed in the domain, LIPSS could still be observed on the surface. Even if they disappear on the surface, only a minor increase would be required.

This study also shows the limits of lateral displacements, which depend on the ratio between accumulated and individual fluence for a domain location of a surface modification and the required spatial emergence. That means a homogeneous area of LIPSS requires a critical displacement. This displacement results in a certain

accumulated fluence. If the domain ratio is too small, the accumulated fluence, which results from the displacement required for a homogeneous area, is too high. The domain ratio also defines a limit for higher pulse overlaps, because the accumulated fluence resulting from a certain displacement should generally not exceed the upper domain boundary. These results confirm the explanation, on Page 64 of Chapter 3, which has been found for the "over-hatch" effect, compare reference [159], and show that for an appropriate ϕ_0 a modification can be maintained on the surface while increasing φ if N_r is reduced.

5.5 Conclusions

In this chapter a systematic and comprehensive experimental study on the spatial emergence of LIPSS has been presented. This study covered the irradiation under SIC and 2d-DIC of a steel sample surface with laser pulses of $\lambda = 1030$ nm and $\tau = 6.6$ ps. Five different types of LIPSS have been observed and classified, three LSFL and two HSFL with periodicities in the range 400 nm to 3 μ m and orientations parallel and perpendicular to the polarisation direction. The Fourier analysis was found to generate additional information to the SEM images. It yields the spatial properties of LIPSS, such as periodicity and regularity. Under SIC, the incubation properties of this material can be described by the selected incubation model.

In the static experiment the surface modifications and their domain boundaries can be determined in an efficient way and the results are consistent with the dynamic experiment. Both under SIC and DIC, the spatial emergence of LIPSS was found to be determined by an accumulated fluence domain, whose location and size vary, due to the incubation properties of the material, with varying irradiation parameters. Whereas under SIC and LODIC, different structure types were usually present together and a sharp transition in form of a threshold has been observed, under HODIC the sequence of surface modifications is traversed gradually with smooth transitions.

The variation of the displacement under DIC was accompanied by a variation of the surface modification and distribution at the same time. With decreasing lateral displacement and otherwise constant irradiation parameters, different surface modifications emerge on the surface. This is also referred to as "over-hatch" effect, in literature [159], and was found to be related to the fluence accumulation process and the incubation properties of the material. Therefore, the repetition threshold has been found to decrease, with increasing pulse overlap from LODIC to HODIC. But beyond that, the accumulated fluence threshold has been found to increase, as expected from the results of Section 3.3.1.

The experimental data show a good agreement with the theoretical understanding of the spatial emergence of LIPSS and the approximation of irradiation parameters, as presented in Chapter 3. As has been shown in the comparison between theoretical and experimental data, the approximations form a valuable guidance for the determination of irradiation parameters under DIC. However, the results also show interesting directions for further research. As has been shown in this chapter, incubation is important for machining areas of LIPSS. But the existing incubation laws

do not consider the characteristics of DIC, such as the non-constant individual fluence. Another important aspect is the influence of contamination on the structuring process.

Chapter 6

Application projects

6.1 Introduction

The research about the spatial emergence of LIPSS, presented in this thesis, was triggered by different application projects, in which the structures satisfied different properties. This research and the application projects, were carried out in parallel. The problem, described in Section 1.5, was related to the lack of knowledge about the application of LIPSS. Since the previous chapters dealt with this problem, this chapter describes where and how the results can be applied. Beyond this, this chapter presents the results obtained in the application projects.

6.2 Surface cleaning

6.2.1 Research context

The NanoClean project was aiming for the optimisation and up-scaling of self-cleaning surfaces for the automotive sector. In this context, self-cleaning was meant to improve the wetting properties of a surface to a superhydrophobic state, so that liquid drops roll off and collect dirt along their way, further on referred to as NanoClean technology. In fact it was Gaiker, working in the automotive sector, which showed interest in this technology. The companies FIAT and Maier took the initiative to define the automotive specifications for the up-scaling of the NanoClean technology. After evaluating several possibilities, the external rear view mirror cup, as a real automotive component, was selected as the final application and a number of general automotive requirements for an exterior trim plastic part were elaborated.

These plastic parts are produced by injection moulding in the automotive industry. Therefore, the NanoClean technology was implemented by combining tailored nano and micro structured injection moulding tools and functional thermoplastic materials. In order to come up with the final result, each project partner working on a specific technology, as moulding, structuring, material and qualification, needed to

Figure 6.1: Injection moulding inserts, (left) 1st generation, (middle) 2nd generation and (right) 3rd generation.

gain information about the NanoClean technology. Therefore it was decided to follow a stepwise approach, where the NanoClean technology was implemented in three research phases. For each research phase the geometrical complexity of the plastic parts and therefore the requirements on the involved technologies were increased.

The company Maier was responsible for the design and manufacturing of the injection mould as well as the realisation of the industrial injection moulding trials. With Maier as project partner, an industrial company participated, which actually produces plastic parts for automotive applications. The trials have been produced with the same commercial equipment used for mass production of car parts. One of the first steps in the project was to design and manufacture the injection mould. The mould design considered, among other things, the mould geometry, mould material, cooling channels, injection gate, ejection gate and part handling. The mould design and manufacturing, was prepared by CAE injection moulding simulations. The three different research phases meant for the moulding design, to be capable to adapt to different plastic part generations. This was accomplished by the development of a mould, which had interchangeable inserts providing different geometrical features.

Figure 6.1 shows left the 1st, middle the 2nd and right the 3rd generation injection mould inserts. The 1st generation had a flat functional surface and six structured fields. With this one, the NanoClean technology was tested for the first time under industrial manufacturing conditions, where the complete manufacturing chain has been followed. The achievements from the 1st generation, were incorporated in the design of the 2nd generation. With the 2nd generation insert different simple geometries, as different draft angles or curvatures of the cavity surface, were introduced. This allowed to investigate the influence of the local surface angle on the structuring process, injection moulding and part appearance. The third generation mould insert represents the final product.

In addition to the three inserts, two further injection mould inserts have been designed. The insert surfaces had flat functional surfaces. With these inserts the industrial durability of the structures has been investigated and their influence on the injection moulding process has been evaluated. These inserts were made of two standard industrial tool steels, Nimax and Mirrax ESR, both available from the company Uddeholm. Whereas Nimax is characterised by its excellent machinability, Mirrax is characterised by a high resistance to corrosion. The selection of these materials was

based on a preliminary study on the structuring quality of different tool steels.

6.2.2 Surface structuring

Laser system

In this project, the laser surface structuring technology, previously demonstrated on a laboratory scale by the University of Twente [156], has been further developed to an industrial scale. Lightmotif, as spin-off company of the University of Twente, was responsible for the development of a laser machining workstation, capable to nano and micro structure complex and large industrial injection moulds, and the structuring of die generations. The development of the laser system as well as the laser process were executed by Lightmotif.

The development of mechatronic solutions for the machining workstation were supported by the company Demcon. The main attributed tasks of the University of Twente were focused on the investigation of the laser surface structuring process, in particular LIPSS, as well as the characterisation of the structured metallic dies and replicated plastic parts. The metallic dies were characterised before and after injection moulding. These were the basic tasks related to this thesis and provided feedback information to each partner during the implementation and up-scaling of the NanoClean technology.

At first a laser machining workstation was developed by Lightmotif, which was capable of structuring large and heavy parts with complex three dimensional surfaces. The development activities were focused on three main topics: motion, software and laser. The laser system integrates a 5 degree of freedom manipulator, for the 3 dimensional macro motion between laser processing head and mould, a galvanometer scanner, for the fast micro motion during the structuring process, a dedicated software program, to map structures, generate set points and control individual subsystems during the machining process, and a laser source, which provides high power, short pulse duration, wavelength flexibility and robust production.

Also this laser system was developed in the defined project phases. The system has been built step by step, based on the increasing demands, until a machine capable of structuring a broad range of moulds was successfully obtained. This also included different beam delivery and scanner concepts. Next to the hardware and software development, this work also included the development of sophisticated alignment and calibration routines to machine the final demonstrator. Finally, a new 3d functionality was implemented in the running software. A challenge was given by the combination of macro and micro motion, which required the stitching of subapertures, see Chapter 4 for further information. The capabilities of this system were verified on a steel sphere that was completely surface structured.

The irradiation model, presented in Chapters 2 and 4 of this thesis, has the potential to be implemented in the software of laser micro machining systems. The information provided by the model could support the process research and the job preparation. The model provides quantitative information about the irradiation output parameters, beyond the determined input parameters. Furthermore, the model

yields a graphical feedback about the applied kinematic trajectory and laser energy. This supports the parameter determination and definition of the machining strategy. Furthermore, the information about the errors of an industrial laser system can be applied as input data and yield the irradiation specifically for the selected system. But also reverse, requirements on the system design can be determined, if information about the process are available.

Structure portfolio

The surface structures, produced by the developed laser system, had to fulfil different functional requirements defined by this automotive application. This meant durable mass produced structured parts of constant quality, produced with a specific industrial replication technology and of a specific polymer material. Due to the overall aim, the requirements were first related to wettability, but in the context of this application also other, such as tribological and optical surface functions, were important. Although the wetting properties basically required structures with a micro- and nanometre scale, the adaptation to this specific application and the other requirements, demanded a certain degree of flexibility of the structuring capabilities. As discussed in Section 1.1, the implementation of a manufacturing technology has to consider the effects on other functionalities of the part.

Since knowledge about the application of LIPSS was lacking, the aim was to obtain a better understanding of their spatial emergence and to increase the structure portfolio. As a result, an irradiation model has been developed by the University of Twente, see Chapter 2, analysing the irradiation by the developed laser system or more specifically the fluence accumulation process. Beyond this fundamental task, the model allows to analyse the influence of technical limitations, as power variations and subaperture stitching, on the irradiation and yields opportunities to optimise the machining accuracy and time, see Chapter 4.

Besides the irradiation model, a theoretical understanding, related to the spatial emergence of LIPSS, has been developed, see Chapter 3. Based on the developed understanding an approach was obtained, which allows to determine irradiation parameters in a practical and efficient way. Within this approach the spatial emergence is quantified by measuring the accumulated fluence domain of LIPSS and applying these data to analytical approximations. This theory also allows to define the available structure portfolio.

One possibility to define the structure portfolio follows from the results presented in Chapter 3. If the idea behind Figure 3.6 is used, the following definition can be given. The structure portfolio is the result of the available kinematic system and possible surface modifications. This means that for a one dimensional translation with a high pulse overlap and the addressing of the surface modification ablation, a groove structure is obtained on the surface. If the given laser-material combination provides LIPSS, the 1d-HODIC results in a strip of LIPSS. Table 6.1 summarises a few possible combinations of structure types as a result of combining kinematics with surface modifications. In the case of 2d-HODIC equal φ , the pulse overlap is equal for the x and y direction. The abbreviation φ^* denotes that the direction of the 2d-HODIC pattern with unequal φ is turned after each repetition. For each structure,

Table 6.1: Non complete definition of a structure portfolio by combining different kinematics with different surface modifications.

Kinematic	SM-Ablation	SM-LIPSS
SIC	Dimple	Zonular
1d-LODIC	Dimples	Zonular
1d-MODIC	Raster	Raster
1d-HODIC	Groove	Strip
2d-LODIC equal φ	Dimples	Zonular
2d-MODIC equal φ	Raster	Raster
2d-HODIC equal φ	Pocket	Area
2d-HODIC unequal φ	Grooves	Strips
2d-HODIC unequal φ^*	Cross-hatch	Raster

the geometrical properties can also be changed. A further degree of complexity is obtained if different structure types are combined.

However, this definition also indicates limitations within the types of applicable structures. In the example above, the spatial emergence of different surface modifications was assumed to be separated by their accumulated fluence domain. But, the domain of different surface modifications can overlap. This is an intrinsic property determined by the given laser-material combination and applied laser irradiation parameters. An example of this situation was given in Chapter 5, where an ablation crater and the structure SM-VI form in the same fluence range. Another example was the formation of SM-IV and SM-VIII. In these cases it can be difficult to apply the different types of surface modifications separately. However, the provided knowledge yields one possibility to define the available structure portfolio.

Parameter determination

In literature, values for irradiation parameters were determined experimentally, usually direct under DIC, and often no specific approach was mentioned. This was the situation at the beginning of the NanoClean project. Small areas have been machined and for each area, parameters as the laser power, displacement velocity, hatch distance or number of irradiation repetitions were varied. Based on the theory, see Chapter 3, and experiments, see Chapter 5, a detailed understanding of the spatial emergence of LIPSS has been obtained. This allowed to understand the spatial emergence of LIPSS under varying irradiation conditions. Beyond this, an approach has been developed, which allows to reduce the effort for the determination of irradiation parameters.

This approach has been verified, for the determination of irradiation parameters of a homogeneous area of LIPSS. Therefore, a simplified static and dynamic irradiation experiment have been performed. The static experiment contained 10 different power values and only $N_p = 200$. These starting values have been selected based on experience. The material was already Mirrax ESR, but the surface was not fine polished resulting in a higher roughness than in Chapter 5. The irradiated surfaces

Figure 6.2: SEM images of the steel sample surface after irradiation under SIC (left) and HODIC (right).

have been analysed with CLSM (Keyence, VK 9710K) and a simple table top SEM (JEOL, NeoScope). The static irradiation experiment revealed periodic structures at the edge and holes in the centre of the irradiated area. Afterwards, the inner and outer diameter of the LIPSS have been measured. Based on these data the beam radius in the sample plane and the individual and accumulated fluence thresholds have been determined.

Since a homogeneous area of LIPSS should be obtained, the velocity was selected sufficiently low. The power has been selected in between the individual fluence thresholds, compare Approximation 3.36, and the number of irradiation repetitions has been calculated by reducing N_p by $\tilde{\Gamma}_p$, compare Approximation 3.38. After machining, the irradiated areas have been analysed by SEM. Figure 6.2 shows SEM images of the obtained structures. The areas showed structures, which exactly correspond to the type of modification and accumulated fluence domain. The implementation of this experiment, comprises less than one working day. By this approach an efficient and knowledge based determination of irradiation parameters has been realised. With less experience or available N-on-1 data from literature, an extended range for ϕ_0 and N_p in the static experiment would have been needed.

Structure generations

The development of the laser system for machining large injection moulds has been finished successfully and was applied for surface structuring of the 1st generation insert. After an initial process research, six structures, each with a field size of 35 mm to 50 mm, have been selected and applied on the insert. The different type of structures and geometrical properties were selected with the purpose to gain information for the different involved technologies in the project, as injection moulding, surface structuring, structure qualification, material development and surface functionalisation. As a parallel task to assess the on-going design and up-scaling of the NanoClean technology,

Figure 6.3: (1. and 3. row) SEM and (2. and 4. row) CLSM images of the 1st generation structures.

the University of Twente characterised the surface properties of every manufactured and structured injection mould insert. The characterisation parameters have been used as feedback data for the optimisation of the structuring process, mould design, moulding process and functionality development.

A complete picture of material changes has been determined by using state of the art techniques, such as OM, SEM, CLSM, atomic force microscopy (AFM), energy dispersive X-ray spectroscopy (EDS) and hardness measurements. It has been found that the topography of the insert surface has been altered in a precise way, but no significant change of the material properties has been observed. The most important parameters were related to the crucial qualitative and quantitative information about

Figure 6.4: SEM images of LIPSS on top of an ablated 1st generation structures.

the topography of the structures. An example of the structures analysed is presented in Figure 6.3, which shows SEM and CLSM images of the six 1st generation nano-micro structures on the 1st generation injection mould insert.

Three different type of structures have been applied, a combined ablation-dimple-LIPSS-area, ablation-line-LIPSS-area and ablation-pocket-LIPSS-area, see Figure 6.3. The ablation structure was varied in their spacing, size and depth, so that a blunt and sharp version of each type were obtained. The height and spacing of the ablation structures were in the order of 20 μm , see Figure 6.3. The periodicity of the LIPSS were found to be about 800 nm, see Figure 6.4 bottom row, and the height range measured by AFM was about 200 nm, see Figure 6.5. The real top to valley height, is larger than this value, because the minimum height has not been reached, as can be observed by the identical and sharp valleys. Figure 6.4 top left shows LIPSS on top of an ablated structure. The orientation of the LIPSS were found to be influenced by the ablated structure beneath. Figure 6.4 top right shows the sidewall of the ablated structure. At the sidewall a different type of LIPSS has been observed, because the sidewalls of the micro pillars received a different exposure than the top part. However, these six structures allowed an evaluation of the different aspects.

The characterisation of all parameters, together with the functional performance,

Figure 6.5: AFM images of LIPSS on top of the 1st generation structures.

was performed again, after the injection moulding trials, to observe durability and to evaluate changes and the necessity and effect of cleaning and repairing operations. In addition an industrial durability test of 20000 injection cycles has been performed with two injection mould inserts. The structure type and geometries were selected based on the 1st generation results. Within this study it was found that the structures withstand 20000 subsequent industrial injection moulding trials, without any changes to the surface topography or significant level of damage. However, some differences between the laser structuring of Mirrax and Nimax steel material have been found. Whereas Mirrax was found to show better laser processing results, Nimax offered financial advantages to the mould manufacturer. The tool steel of lower quality was found to form stochastically distributed holes after laser ablation.

The results obtained with the 1st generation were used for the design of the 2nd generation structures. The structures have been selected to study injection moulding boundary conditions, as for example undercut structures being not perpendicular to the release direction. After studying different boundary conditions with the 2nd generation structures, the final structure generation has been developed. The third generation mirror cup was fully automated and successfully machined by the developed

laser system. The machining of the insert with 25000 mm², took 50 hours in total. It should be noted, that for this final job parameters were selected which guaranteed a safe machining without optimising process speed.

Risk assessment

Based on the developed model and theory, see Chapters 2, 3 and 4, a risk assessment of the laser surface structuring technology has been performed. This analysis incorporates the risk identification and source analysis, the assessment of risk and error sources and, in the context of a risk management, the handling and optimisation strategies. In the following only a brief discussion on this topic is presented.

In the first step, risks and events were identified, which could lead to a negative outcome of this technology. In the second step, error sources were analysed and related to the identified risks and events. A part of the analysis of irradiation errors is presented in Chapter 4. In order to relate specific error sources to specific events and risks, the developed theory, Chapter 3, was applied. In the beginning, the analysis was conducted without any assessment of the probability or impact. This allowed to generate a list of risks, events and sources, which gives a total overview of possible scenarios.

With respect to the overall NanoClean technology, a negative outcome would be a technical failure of the functionality or the increase of costs to produce the functionality. A technical failure can be the deviation of the structure from the design in terms of dimensions or irregularities. An increase of research and machining costs could lead to a negative financial outcome. The research costs are mainly determined by the time spent to determine the irradiation parameters for the specific laser-material combination, setup conditions and structure type. The machining costs are related to the actual laser machining of the mould. But also the costs which occur in case of any damage to the system or to the work piece have to be considered.

The most critical events leading to a technical risk are related to position errors of the combined macro-micro motion. These errors can directly lead to an accumulated fluence outside the domain boundaries and will cause a structure irregularity or a total different structure. This finally leads to a failure of the surface function at this location. Furthermore the number of factors leading to such an event and the probability is very high. The most critical events leading to a financial risk were found to be related to an arbitrary selection of a set of irradiation parameters and an unused optimisation potential, which ends up in a too long machining time. Since this process is an additional process in the overall production process of an injection mould tool for the manufacturing of automotive parts, this finally can lead to the refusal of the laser technology.

Errors related to statistical positioning errors can be reduced with an optimisation towards larger displacements. This reduces the ratio between the position errors and the absolute displacement increment and therefore the fluctuations in the accumulated fluence. The optimisation can be done by a calculation of the critical velocity required to obtain a homogeneously covered area with LIPSS and subtracting a safety margin for error sources. Another approach can be the use of a larger focus diameter, by a beam expander or larger focal length, and calculate a new set of parameters

Figure 6.6: Plastic parts, (left) 1st generation, (middle) 2nd generation and (right) 3rd generation, obtained from the 1st, 2nd and 3rd generation injection moulding inserts, see Figure 6.1.

from the old set by using the approximations presented in Chapter 3 or simulations. Regarding the stitching error, the correct boundary can be calculated using the irradiation model. The remaining error in the accumulated fluence can be further reduced if the intersection applied for machining tiles is reduced.

The determination of irradiation parameters can be improved by the determination of the beam radius in the sample plane by the method of Liu [95]. The knowledge about the exact beam diameter in the sample plane allows to determine the exact displacement, fluence input and optimisation steps. This method can be partially executed automatically by the machine and depending on the optical vision system direct measurements on the machine are possible. This method can be extended to an N-on-1 measurement to determine the structure portfolio. Furthermore it allows to determine the domain boundaries and the incubation factor, which helps to optimise the machining quality and to reduce the machining time. The machining time can be optimised by choosing a lower domain position and determining the optimal expander-lens combination. Therefore the irradiation model can be applied to simulate the optimal machining time for a given setup and to avoid unexpected irradiation results. The approximations can be applied to find the irradiation parameters for these optimised conditions.

6.2.3 Wetting measurements

So far the technology to manufacture the plastic parts, with a dedicated and permanent surface structure, was discussed. But within the NanoClean technology, surface structuring was only one contribution to the improvement of the surface functionality. The other contribution comes from the material properties of the parts. BASF's role in the project was to develop a plastic compound for injection moulding with the mechanical properties required by the automotive industry, combined with an enhanced hydrophobic behaviour. For this research, the specific requirements of the automotive part manufacturer had to be taken into account. As a result, a new series of styrenic thermoplastic compounds characterised by a novel wetting behaviour were developed. Therefore, the replication quality and hydrophobic properties of different

Figure 6.7: SEM images of replicated 1st generation structures.

styrenic materials were investigated. Finally, it was possible to achieve a superhydrophobic surface, based on the styrene copolymer, acrylonitrile styrene acrylic ester, abbreviated ASA, combined with hydrophobisation and flow improving additives.

The task of the University of Twente was a qualitative and quantitative characterisation of the topography of injection moulded plastic parts, see Figure 6.6, and a comparison to the injection mould inserts to evaluate replication rates and any indication of wear of structures after thousands of injection moulding shots. Figure 6.7 shows SEM images of the different replicated 1st generation structures. In general, the aspect ratio of blunt structures provides advantages compared to the sharp structures for different processes, such as filling during injection moulding, measurement accuracy for characterisation and stability of form and height for laser machining. Furthermore, the surface functionalities showed a clear dependence on replication rate. The structure with the dimples showed visually the best results, with respect to the structure homogeneity. However, as can be observed in the SEM images, the top part of the structures was not replicated well. Therefore the optimal surface functionalisation was not obtained with the 1st generation.

To achieve a better result was a difficult task, because the wetting properties were at the same time influenced by the plastic material, moulding process and the structure design. Therefore, an iterative process between all research tasks was required. Under this condition the companies Gaiker and Maier performed supporting actions to address developments from BASF. Injection moulding process optimisation was performed, by varying injection moulding parameters. Figure 6.8 shows CLSM images of different replication qualities. By using variotherm injection moulding at BASF's facilities, it has been found that the moulding temperature and their course have a significant influence on the replication behaviour and later on the achieved hydrophobicity. Especially, a higher mould temperature enhanced the hydrophobicity,

Figure 6.8: CLSM images of different replication qualities, (left) fully and (right) partly replicated.

but without active cooling, problems during the ejection were obtained.

The information from the iterations were also used as feedback for the design of the mould and structures. The holes observed in the insert structures for steels of lower quality were also present in the replicated parts and their quality was reduced. These activities allowed to detect potential problems for up-scaling from lab-scale to industrial-scale and to take decisions and define actions towards production of plastic components on an industrial scale. Based on this information, the mould material was adapted and different heating technologies, possible to be combined with the commercial moulding equipment, were compared. As a result a heating and cooling system were developed and implemented. The injection moulding trials on the mirror cup were performed at a melt temperature ranging from 250 °C to 280 °C. The actual mould temperatures were set to 110 °C as a maximum technical limit for the available prototype mould.

Next to the support of the development of the injection moulding process, Gaiker was responsible for the evaluation of the wetting and cleaning properties and FIAT took the responsibility to evaluate the finally achieved results. The selected polymers have been characterised by their mechanical, chemical and surface energy properties in FIAT's laboratories. The automotive characterisation tests included weathering, impact tests, abrasion and car-wash resistance, cleanability, contact angle and cold

Figure 6.9: Results of WCA measurements for different plastic materials.

crash tests. The results were evaluated on the basis of the defined automotive requirements and established according to internal FIAT and ISO regulations. These characterisations were performed for different part generations, structure generations and polymer materials. The ASA material developed during the project, denoted by Luran v4, was compared to the standard ASA, denoted by Luran std. Two additional materials were used for comparison: a styrene butadiene copolymer, denoted by SBC, and a polypropylene, denoted by PP.

The primary goal was to obtain superhydrophobic parts, as a basis for improved cleaning performance. The water contact angle, abbreviated WCA, characterisation showed that the structuring and additives increase the WCA, achieving the expected value of 150° . Figure 6.9 summarises the obtained wetting results for different materials. In particular 30 % WCA increase was obtained from a conventional mirror polished surface to a structured surface of Luran std. Further 25 % increase of WCA was obtained from the structured surface Luran std to Luran v4. In total 67 % increase of WCA was obtained by replacing mirror polished surface of Luran std. by the structured surface of Luran v4. The developed plastic material shows significantly improved WCA combined with mechanical properties within the specification for the mirror cup application requested by FIAT.

With respect to the cleanability, an overall test procedure to quantitatively evaluate the aesthetic and cleaning properties was defined. At the end of the project, the first cleanability tests for car interior components were performed. For example, hand cream and coffee were used as dirt agents. The final samples showed improved

cleanability in comparison with standard samples, while no influence on the mechanical or chemical properties has been observed. These cleaning results are explained by the improved wetting properties.

In conclusion, a preferred mould material has been identified, knowledge on mould design for structured surfaces has been gained, various mould inserts have been machined, a laser micro machining system has been developed, micro and nano structures have been applied to 3d surfaces, knowledge about the application of LIPSS has been gained, a new plastic material has been developed, structures were found to be durable, the WCA has been significantly increased and improvements of the surface in terms of easy-to-clean performance have been demonstrated.

The obtained results confirm that the structured ASA material combined with the hydrophobisation and flow improving additives present excellent properties for the NanoClean mirror cup. The up-scaling capabilities of the proposed NanoClean technology towards self-cleaning plastic parts were demonstrated and found to be dependent on the surface structure configuration and injection moulding replication quality as well as on the chemical nature of the polymer. The main objective of the NanoClean project consisting of mass-production of permanent self-cleaning 3d complex plastic components for automotive industry was successfully achieved.

6.3 Tissue regeneration

6.3.1 Research context

Actually this research was triggered by the tissue engineering and implant manufacturing industry, where biodegradable polymers are applied to support the tissue while healing [125]. The success strongly depends on the adhesion and proliferation of the cell-implant contact [174]. The adhesion, proliferation and differentiation of cells is influenced by the surface structures, in particular by structures on a nanometre scale [35]. Since these structures influence the cell adhesion, this might also influence the tissue regeneration [175]. Therefore, large areas of specific nanometre sized surface structures are required. LIPSS provide these spatial properties and their slight natural disorder, compared to structures made by lithography or laser interference, was expected to be beneficial for mimicking biological materials.

Therefore, this research dealt with the application of LIPSS and was divided in two parts, first the production of homogeneous areas of LIPSS on a biodegradable polymer and second the study of their influence on the alignment and morphology of human mesenchymal stem cells. Due to the absorption properties of the biodegradable polymers on a broad range of wavelengths, it was decided, instead of directly structuring the polymers, to structure a steel mould and to replicate the obtained structures by nanotemplating. This required a long experimental chain, first the laser structuring, second the structure replication and third the cell study. In parallel, each step required characterisation and analysis. Furthermore, three different structure periodicities and polymer material were investigated.

This study was performed at the University of Twente as a collaboration between three different departments. The laser surface structuring was performed at the labor-

Figure 6.10: SEM (top and middle row) and AFM (bottom row) images of the steel sample surface exposed to laser pulses of $\lambda = 1030$ nm (left column), $\lambda = 515$ nm (middle column) and $\lambda = 343$ nm (right column). All LIPSS are perpendicular to the polarisation of the laser light.

atories of the Chair of Applied Laser Technology by Wendy Masman-Bakkers. Since the experimental chain was rather long, it was required to be efficient while performing all experiments and therefore it was decided to determine irradiation parameters by the approach presented in Chapter 3 of this thesis. It was the first application of this approach. Justus Eichstädt accompanied the work of Wendy Masman-Bakkers and supported the calculation of irradiation parameters for homogeneous areas of LIPSS based on the analysis of static SEM images, as presented in Subsection 6.2.2 Paragraph "Parameter determination". Afterwards, Wendy Masman-Bakkers did the cell study for her master thesis [176].

6.3.2 Surface structuring

The laser experiments were performed with the same laser system as described in Chapter 5. As material for the specimen stainless steel 304 was selected. The surface of the sample was polished to a mirror like grade. The samples were irradiated by three different wavelengths, $\lambda = 1030$ nm, $\lambda = 515$ nm and $\lambda = 343$ nm, to obtain structures of different periodicity. The structured surfaces were analysed by different microscopy techniques as SEM and AFM. At first a static experiment was performed in order to determine the available LIPSS types and to measure their accumulated fluence domains. Therefore, SEM images have been used to determine the inner and outer diameter of LIPSS emergence under SIC. The AFM images have been used to determine the periodicity and height of the LIPSS.

In the static experiment, for each wavelength LIPSS have been observed. Figure 6.10 top row shows SEM images of the steel surface exposed to different wavelengths, left $\lambda = 1030$ nm, centre $\lambda = 515$ nm and right $\lambda = 343$ nm. The images show LIPSS for which the upper domain boundary has been exceeded. For each wavelength the accumulated fluence domain was measured at $N_p = 200$. The obtained results for $\lambda = 1030$ nm were found to be in agreement with the results presented in Chapter 5. The only difference, was that at the time the laser experiments for the cell study were performed, SM-IV and SM-V were not recognised as different modifications and as a result the upper domain boundary was considered to be SM-VI. The time for the static experiment, the SEM measurements and their analysis was about one working day per wavelength.

Afterwards, the irradiation parameters for the dynamic experiment were estimated. Since homogeneous areas were required, the pulse overlap was selected with 0.80 and based on this choice the velocity was obtained by Equation 3.42. The average power of the laser has been obtained from Approximation 3.36 and the number of irradiation repetitions from Approximation 3.38. The set of irradiation parameters was first tested on small areas of 150 μm by 150 μm and later on up-scaled to 1.4 cm by 1.4 cm. Multiple areas were machined with varying N_r , in order to select the area with the best spatial regularity. For the estimated irradiation parameters, LIPSS areas were obtained and no further experiments were required. This procedure was repeated for each wavelength. The time for the dynamic experiment, the SEM measurements and their analysis was about one working day per wavelength.

Figure 6.10 middle row shows SEM images of the steel sample surface, exposed to different wavelengths, left $\lambda = 1030$ nm, centre $\lambda = 515$ nm and right $\lambda = 343$ nm. The images show areas which are homogeneously covered with LIPSS. The orientation of these LIPSS were in all cases perpendicular to the polarisation direction. With decreasing laser wavelength, the periodicity of the LIPSS were found to be smaller. The periodicity of the LIPSS at $\lambda = 1030$ nm was about $\Lambda = 510$ nm, at $\lambda = 515$ nm about $\Lambda = 315$ nm and at $\lambda = 343$ nm about $\Lambda = 232$ nm. The results obtained in the dynamic irradiation experiment confirmed that measuring the accumulated fluence domain is an efficient approach to obtain homogeneous areas of LIPSS. Furthermore, this application shows that the presented approach is robust and even works if only a small effort to determine the irradiation parameters is applied.

6.3.3 Stem cell cultivation

After the structured mould was manufactured by laser machining, a negative replication of the structures was produced with nanotemplating by photo-polymerisation. The structures were replicated in two different biodegradable polymer materials, first poly (trimethylene carbonate) hydrogels (PTMC) and poly (ethylene glycol) - poly (trimethylene carbonate) hydrogels (PEGPTMC). Afterwards a cell culture study was performed. For this study human mesenchymal stem cells (HMSC) have been selected. These are precursor cells of bone forming cells, referred to as osteoblasts, and contribute to wound healing processes. The cells were cultured for seven days.

Figure 6.11 shows in the top row SEM images of the polymer surface with the HMSC and LIPSS, in the middle row staining images from a phalloidin staining of the actin fibres inside the cells and in the bottom row images obtained from a stereo optical microscope, abbreviated SOM, of the HMSC on a flat (left) and structured (right) control surface. The SEM images show that multiple HMSC were found to be aligned with the LIPSS, compare Figure 6.11 top row. Based on the staining analysis, the cells cultured on LIPSS were found to be more elongated compared to those cultured on a flat control surface, compare Figure 6.11 middle row. The SOM images show that on the specimen with the LIPSS structures, the HMSC align to a certain direction, whereas on the non-structured surface they form clusters which grow in different directions, as can be seen in the lower row of Figure 6.11.

This study shows that it is possible to culture cells for at least seven days on biodegradable polymers covered with LIPSS. The LIPSS structures seem to influence the morphology and alignment of the HMSC. It was not possible to clarify if this influence is beneficial for bone generation. Therefore, further sequential studies are required. The applied replication method, where multiple new samples of LIPSS areas can be prepared, provides a solid basis for this purpose.

6.4 Precision positioning (I)

6.4.1 Research context

Semiconductor equipment requires high precision contact mechanics to position components very accurately. To control the position accuracy, the control of friction properties is required. A method to control this type of surface functions could be the application of laser-induced periodic surface structures. The application of LIPSS in a flexible and efficient manufacturing process requires first knowledge to control the spatial emergence of LIPSS on extended areas and secondly the determination of their mechanical properties.

The use of LIPSS, as a tool to influence tribological properties, was already proposed in 1999 by Yu and Lu for the reduction of stiction problems of hard disk drives [177]. The influence of femtosecond initiated LIPSS was investigated by Mizuno et al. [152] and the reduction of lateral force as well as decrease of adhesive force was found. Another study reports a decrease of friction for full and increase for partially structured areas [153]. These results, mainly obtained on DLC coatings with fs

Figure 6.11: SEM (row 1), staining (row 2) and SOM images (row 3) of HMSC on LIPSS.

pulses, provide only limited information about the influence of LIPSS on tribological properties.

With the results of this thesis the knowledge about the application of LIPSS has been extended. In order to support the LIPSS further towards industrial application, also the second step was pushed forward. Therefore, extended areas homogeneously

covered with LIPSS have been produced by Justus Eichstädt with the approach presented in Chapter 3 of this thesis. Afterwards, a preliminary study about the dynamic friction properties of LIPSS has been performed. The following description gives a short summary of the obtained results. Further details can be found in references [166] and [178].

6.4.2 Surface structuring

The irradiation experiments have been performed with the laser system described in Chapter 5. The irradiation parameters have been determined by the approach described in Chapter 3. First, a static irradiation experiment has been performed, where the fluence domain boundaries were determined for one type of LIPSS under SIC. Next, the Approximations 3.36, 3.38 and Equation 3.42, derived in Chapter 3, were applied to obtain extended areas of uniform LIPSS.

Inspection of specimen surface and measurement of structure geometry were done by OM (Leitz, DMRX) equipped with a camera (Leica, DFC 450 CCD), CLSM (Keyence, VK 9710K) and AFM (Nanosurf, Easyscan 2). As specimen, a flat single crystalline silicon mirror substrate (Melles Griot, PM-1025-Si) was used. The mechanically polished surface has a roughness, determined with CLSM, of $R_a = 12$ nm and $R_t = 80$ nm according to ISO 4287:1997. The hardness, measured with a hardness meter (Shimadzu, DUH-200), was 5.3 GPa or 506 Hv, using a Berkovich indenter at 200 mN load, the elastic modulus was 124 GPa. Cleaning of the specimen was done before and after laser machining using an ultra-sonic-bath, filled with isopropanol.

The single pulse experiment revealed a modification threshold of $\phi_{th} = 0.053$ J/cm² and an ablation threshold of $\phi_{th} = 0.537$ J/cm². The multi pulse experiments revealed an incubation factor of $\xi = 0.91$ and three types of LIPSS: type I with $\Lambda_I \approx 0.950$ μ m and an orientation perpendicular to the polarisation direction, type II with $\Lambda_{II} \approx 1.6$ μ m and an orientation parallel and type III with $\Lambda_{III} \approx 0.750$ μ m also parallel to the polarisation. For this experiment type III has been selected and the corresponding values for the domain boundaries have been determined in static irradiation experiments.

Figure 6.12 (a) to (d) show CLSM images of five different locations on the surface, irradiated with $\lambda = 1030$ nm, $\tau = 6.6$ ps, $f_p = 40$ Hz and $N_p = 2000$. The peak fluence was varied between $\phi_0 = 0.113$ J/cm² and $\phi_0 = 0.080$ J/cm². Within the irradiated areas, regions with different appearance can be distinguished. In Figure 6.12 (a) the following regions can be distinguished, a bright area in the centre surrounded by an annular region of LIPSS, followed by a dark and grey annular zone. The images show how the spatial emergence of LIPSS vary according to the intersection of the domain boundaries with the different deposited accumulated fluence levels, as indicated in Figure 3.4. The values for the boundaries were found to equal $\Gamma_{th}(2000) = 161.6$ J/cm² and $\Delta\Gamma_{th}(2000) = 31.5$ J/cm².

Instead of further experiments the Approximations 3.36, 3.38 and Equation 3.42 were used to calculate irradiation parameters for an extended and uniform area of LIPSS. Next, four areas were irradiated, each of them having a length of approximately 150 μ m. Using Equation 3.30 gives 177.4 J/cm² for the centre of the domain at N_p

Figure 6.12: CLSM images of silicon surface irradiated with varying ϕ_0 , φ and N_r .

= 2000. The peak fluence, determining the power by Approximation 3.36, was kept constant at $\phi_0 = 0.089 \text{ J/cm}^2$. The pulse overlap was selected to give a small spread in terms of accumulated fluence and therefore a good regularity. Four different values of φ varying from $\varphi = 0.93$ to $\varphi = 0.83$ were selected for the velocity given by Equation 3.42. The varying repetitions $N_r = 24$ to $N_r = 160$ follow from Approximation 3.38.

Although the velocity and number of repetitions were different, for all approximated sets of irradiation parameters, areas with similar LIPSS were obtained. This shows that keeping the accumulated fluence at a certain level has a decisive role in the spatial emergence of LIPSS. Figures 6.12 (e) to (h) show CLSM images of these areas, irradiated with multiple displaced pulses of $\lambda = 1030 \text{ nm}$, $\tau = 6.6 \text{ ps}$, $f_p = 1000 \text{ Hz}$, $\phi_0 = 0.089 \text{ J/cm}^2$ and varying pulse overlap (a) $\varphi = 0.93$ and $N_r = 24$, (b) $\varphi = 0.90$ and $N_r = 53$, (c) $\varphi = 0.86$ and $N_r = 101$ and (d) $\varphi = 0.83$ and $N_r = 160$. The LIPSS were found to be of the same type III as observed under N-on-1 conditions with $\Lambda \approx 750 \text{ nm}$ and with a direction parallel to the polarisation direction. The different areas show surfaces with homogenously distributed LIPSS. The areas show only minor differences in regularity of the LIPSS formation.

The requirement regarding the domain size was fulfilled with a sufficiently small Γ_d , compare Subsection 2.3.3 for all irradiation conditions. But with respect to the regularity of the structures in the different areas, small variations can be observed. This could be related to differences in Γ_p , compare Subsection 2.3.3, which resulted in different addressed domain values. Reason for this is the use of integer values for N_r . But also the domain boundaries could vary, due to the decrease of average individual fluence and incubation, as discussed in Chapters 3 and 5. The effective number of pulses and the variation of individual fluence are different for all areas and are also different compared to the static experiments. Lower individual fluence contributions and higher pulse numbers, increase the accumulated fluence threshold Γ_{th} . Since the actual incubation law does not account for varying fluence, further investigations should be focused on this issue.

In literature LSFL and HSFL are distinguished [138]. Femtosecond laser pulse experiments reveal for LSFL on silicon a periodicity of $0.62\lambda < \Lambda < \lambda$ and an orientation perpendicular to the laser polarisation [130]. The observed LIPSS, with $\Lambda_{III} \approx 0.750 \mu\text{m}$, could be attributed to LSFL, but the orientation of this LIPSS is found to be parallel instead of perpendicular to the polarisation direction. It is reported that for high number of laser pulses per area, $N_p > 100$, periodic groove structures can occur, which can have an orientation parallel to the laser polarisation [130]. The period of grooves is found to be 2-3 times the wavelength. So it is not a conventional groove structure. But the low fluence and orientation of this LIPSS argue for HSFL with a larger periodicity. Irrespective of the exact qualification of these LIPSS as HSFL or LSFL or grooves, this type of LIPSS was used for the friction measurements.

Extended surface areas have been obtained, which were homogenously covered with LIPSS. The areas, obtained by different irradiation parameter combinations, satisfying accumulated fluence boundary conditions, show the same type of LIPSS. This observation confirms the theory presented in Chapter 3. Large areas with LIPSS are required in the field of surface functionalisation. The use of static experiments allows to perform the empirical part of this approach in an efficient way and the

approximations give a systematic control of the irradiation parameters. The determination of domain boundaries allows to allocate the applied accumulated fluence with respect to domain boundaries of different morphologies and to obtain multiple sets of irradiation parameters.

6.4.3 Friction measurements

Friction was measured using a tribometer (CSM, Nanotribometer) with a linear reciprocating module in a ball-on-flat configuration. Normal loads, between $F_N = 2$ mN and $F_N = 25$ mN, were varied. To record stable signals, the number of cycles was 10 and measurement signals were averaged across the number of cycles of $0.3 \mu\text{m}$ amplitude. The measurement was repeated 4 times, each repetition in a new track on the surface. A single crystalline silicon ball (ISP Optics, Si-B-5) of 5 mm diameter was glued on a bending element, a double leaf spring made of photo structured glass. The roughness of the ball was specified to be below $R_a = 1$ nm. The displacement of the bending element is measured with a fibre optic sensor system, based on reflection intensity variation. The tribological tests were performed at a speed of 0.1 mm/s and at ambient conditions of temperature 20.5°C and 20% relative humidity in air.

The friction, depending on load, was measured, using a Si ball sliding perpendicular to the structure orientation on the machined as well as on the untreated area. Initial experiments showed that for F_N well over 25 mN LIPSS wear off. As a consequence lower F_N for friction measurements were selected in order to avoid wear and to get stable and reproducible friction measurements. Below 25 mN, no indication for wear after 10 cycles was found. According to Hertzian equation for point contact, for $F_N = 25$ mN and Si ball on Si flat, the contact half width is $9 \mu\text{m}$, indicating that 24 LIPSS periods are in the contact area with the ball. The ball deformation was calculated to be 16 nm at 25 mN.

It was found that the friction force, for $F_N = 25$ mN, measured on the structured surface is about 1.6 times higher compared to the friction on the untreated, compare Figure 6.13. For F_N below 25 mN the friction properties of the structured area and of the initial area tend to get closer together with decreasing load. Between 25 mN and 5 mN the friction force depends on F_N and shows, to a first approximation, a linear relation. Due to technical limitations for loads below 8 mN, the measurement accuracy decreases and both friction signals cannot be unambiguously distinguished.

Within this study the coefficient of friction on the structured area was found to be about a factor of 1.6 at $F_N = 25$ mN higher compared to the initial surface. In [152] a reduction of friction force for nN loads has been observed and in [153] tested under F_N of 2 N and 10 N as well. Reasons for different results could be due to different measurement conditions, under which the load dependence was not explicitly determined, and different laser systems for structure generation.

The increase of friction between structured and untreated surface, could be either due to a modification of topographical or mechanical surface properties. Both surfaces exhibit a certain roughness, the structured surface can be described more accurately by a wavy surface whereas the untreated surface exhibit more stochastically distributed roughness peaks, which could be described by an exponential distribution of

Figure 6.13: Friction force versus normal load for 2 mN until 25 mN. The dashed line is measured on the structured area and the continuous on the initial surface.

asperity heights. The real contact area for the wavy surface scale differently [179] with increasing load then the stochastic surface [5]. This may result in a higher friction signal on the wavy surface. Another argument for the influence due to topography could be given by contact mechanical simulations. At $F_N = 25$ mN 24 periods are in contact and the ball deformation is 16 nm. Even if both surfaces deform 50 nm in summation, an inhomogeneous contact is given and may give different friction results.

On the other hand the initial crystalline state of the material could be changed. An indication is given by the surface images, where the irradiated surface appears to be thermally modified. Further evidence is given by initial experiments with F_N well above 50 mN, where LIPSS starts to erase. For that range of F_N the friction signal of the structured area was further increasing compared to the initial. But wear track analysis shows that LIPSS were erased. From this experiment the influence of topography and mechanical surface properties on the increase of friction cannot be unambiguously distinguished.

For loads below 25 mN the friction forces of the structured and the initial area tend to get closer together with decreasing load. The continuation of measurements for smaller geometries in combination with lower loads would be interesting, especially if

a cross-over point between high and low friction would be observed. Due to technical limitations, the measurement accuracy decreases at low F_N and could not be improved by repetition of the experiments. Conclusions from that part of the graph, especially for the cross-over point, cannot be drawn. In Subsection 6.5.3 measurements are presented, which have an accuracy that allow to determine adhesion effects.

The increase of friction with increasing load could be applicable for areas where accurate static contact is required. It might be possible to optimise the structure design in the direction, that both, increase and decrease of friction can be obtained with different normal loads. But this improvement needs to overcome the limitations by tribological characterisation techniques. The relevance for the application also depends on the long term stability, e.g. wear behaviour of the structures. In conclusion, the influence of laser surface structuring on friction was found as an increase of the friction coefficient for structured areas. Friction increase can be explained as an effect of increased roughness, however the influence of a change in mechanical properties of the surface cannot be excluded.

6.5 Precision positioning (II)

6.5.1 Research context

In parallel to the research presented in this thesis the M2i TULP research project has been carried out. In this fundamental research project, the physical mechanisms leading to the characteristic formation of LIPSS have been studied theoretically and experimentally [180]. To investigate the formation mechanism of LIPSS experimentally, the production of LIPSS is required. Therefore, a close collaboration with the TULP project has been formed.

The industrial partners involved in this fundamental project each had specific interests in LIPSS. One industrial partner was interested in the application of LIPSS for precision positioning applications, involving components with high flatness and low roughness. The positioning of parts with these physical properties, can be limited by adhesion or even sticking. For this specific application a separate application project has been performed. For very low roughness of surfaces, adhesion forces are dominated by van der Waals forces and capillary forces and these forces are strongly dependent on the real contact area of contacting surfaces. One possibility to reduce the spontaneous adhesion of components is to reduce the real area in contact, which requires a precise control of the surface topography.

The aim of the application project was on the one hand to prove that sticking and adhesion forces can be reduced by surface structures and on the other hand to evaluate different structuring technologies. Therefore, in this project different structuring technologies and processes have been compared. For this application different requirements were defined, such as avoiding the production of particles and changes to flatness. This illustrates that, as discussed in Section 1.1, the implementation of a manufacturing technology in an existing manufacturing chain has to consider the effects on other functionalities of the part and technologies in the manufacturing chain.

One technology, which was applied in this project, was laser surface structuring using LIPSS. For this project, different extended areas of LIPSS were required. The samples for the measurements have been structured by Justus Eichstädt, using the knowledge about the spatial emergence of LIPSS. On the structured areas adhesion measurements have been performed at TNO. More background to this topic can be found in reference [181].

6.5.2 Surface structuring

For the structuring of the sample surfaces, initially two different laser systems have been evaluated. The first was a Ti:Sapphire based laser system consisting of a pump laser (Coherent, Verdi), femtosecond oscillator (Coherent, VPUF) and regenerative amplifier (Coherent, RegA 9000). The central wavelength of this system was about 800 nm and the pulse duration smaller than 200 fs. The second laser system was the same as described in Chapter 5. Both systems were used in direct writing configuration, with similar equipment for handling and control. As material single crystalline silicon wafers delivered by the industrial partners were used. The initial femtosecond experiments have been produced in cooperation between Justus Eichstädt and Johann Skolski.

In the static experiments similar types of LIPSS have been observed with both systems. Figure 6.14 shows in the top row two examples for surface modifications at $N_p = 100$. It should be noted, that the structures have been recorded with different SEM systems and therefore the structure appearance was found to be slightly different. Interestingly, the spatial emergence in static experiments also has been found to be determined by an incubation dependent accumulated fluence domain.

Furthermore, in initial dynamic experiments, similar transitions between surface modifications, as presented in Chapter 5, have been observed. Figure 6.14 shows in the bottom row two examples of the transition from conventional LSFL to grooves. The experimental results obtained under HODIC were found to be interesting for the analysis of LIPSS formation mechanisms, because the FT analysis of SEM images showed well defined frequency domain features. Furthermore, a good morphological correlation between dynamic results and finite-difference time-domain simulations [182], were obtained.

However, with the picosecond laser system, the particular LIPSS type observed around the threshold was not stable at very low pulse energies. This was probably due to a lack of intensity at this wavelength, which was 1030 nm. Also the switch of the laser wavelength from 1030 nm to 515 nm was not leading to an improvement with respect to stability. The third harmonic 345 nm was not an option for the application, because a certain periodicity was required. But on the other side, the femtosecond laser system was characterised by an unstable laser power and small available energy range. Therefore, finally for this specific application the picosecond laser system has been applied to obtain areas of LIPSS.

Next to the demand of extended areas, for this application it was very critical to avoid the production of particles or other contamination during machining. Therefore, it was required to select a type of LIPSS which produces as little as possible material

Figure 6.14: SEM images of a silicon surface after irradiation with pico- and femto-second laser pulses under SIC (top) and HODIC (bottom).

ablation. Based on literature study about the influence of surface topography on adhesion, for the given contact mechanical situation, the height of structures was required to be between 1 to 10 nm [11, 183]. This requirement was determined on surfaces with homogeneous and stochastically distributed surface roughness. LIPSS are often accompanied by material ablation and although less is known about the height of LSFL, a few hundreds of nanometres can be expected [184, 185].

This altogether led to the application of a very specific type of LIPSS for the adhesion measurements on extended surface areas, namely surface plasmon polariton driven LSFL, abbreviated SPP-LSFL. They were found to grow under SIC for high pulse energies and low pulse numbers. They were found to be accompanied by melting, but very limited material ablation. Their accumulated-individual fluence ratio was so high, that homogeneous areas were difficult to produce. But, for this application a homogeneous closed area on the scale of the structure dimension was not required.

Therefore, it was decided to produce an inhomogeneous area of SPP-LSFL. The pulse overlap of approximately 0.50 was at the transition between LODIC and MODIC.

This meant for the beam and focus conditions a displacement in the order of 10 μm . At each displaced location, the SPP-LSFL were produced and used to reduce the mechanical contact area. In this way a hybrid contact area was obtained, with alternating flat and structured surface parts. Figure 6.15 shows CLSM images of one area with the obtained surface structures.

Multiple areas of 6 mm by 6 mm edge length have been produced. For each structured field, the height of the structures were varied by the laser power. This required a stable and defined power control, wherefore the picosecond laser system was selected. The height values, determined by CLSM, were found to vary from 44 nm top valley height and $R_a = 6.2$ nm to 245 nm top valley height and $R_a = 19.8$ nm. The periodicity was found to be very close to the laser wavelength. The areas showed very smooth structures, although some particles have been observed. But the number of particles was far less than usually observed on LIPSS areas. The structuring quality was found to be limited by the position accuracy and pulse energy stability available under research conditions, compare Chapter 4.

6.5.3 Adhesion measurements

The adhesion measurements have been performed by means of a mechanical characterisation system (ASMEC GmbH, UNAT), which is an extension of the nano indentation technique. For the adhesion measurements a silicon infrared-lens with a surface roughness $R_q = 2$ nm and radius of 123 mm was used as counter surface. These measurements were performed under controlled environmental conditions. For this measurement, the silicon lens approached the surface until a full contact was obtained and then retreated. The force F_A required to pull up the lens was measured and used as equivalent for the amount of adhesion.

First a silicon reference sample has been measured and then the laser treated sample. On the laser treated sample first the unstructured and afterwards the structured surface areas have been measured. In case of the reference sample, an adhesive force of about 80 mN has been obtained, in case of the laser treated sample and unstructured area an adhesive force of about 68 mN has been obtained and in case of the different structured areas no adhesion has been measured. For the non-structured areas a clear snap-on behaviour has been observed and the adhesive forces were comparable. Figure 6.16 shows the obtained force-displacement graphs, Figure 6.16a the measurements on the reference sample in black colour and on the unstructured area of the laser treated sample in red colour and Figure 6.16b the measurements on the unstructured area of the laser treated sample in black colour and structured areas in red, green and blue colour.

In this study, the potential of laser surface structuring for the reduction of adhesive forces has been shown on a laboratory scale. The applied surface structures effectively reduced the adhesion forces. Although different structures with different heights have been applied, in all cases no adhesion has been observed. The smallest LIPSS, which have been applied in this study, have a height of about 50 nm. Apparently this height was above the threshold for adhesion reduction. For future experiments reduced structure heights would be interesting in order to obtain the adhesion reduction as a

function of structure height.

Although a few particles have been observed in the CLSM images, the adhesion measurements on the unstructured area of the laser treated sample showed a clear snap-on during the approach and adhesion has been measured. This result can be confirmed by the experience with laser surface structuring. The applied SPP-LIPSS dimple pattern produces a very low amount of particles compared to laser ablated structures or other type of LIPSS. This is partly due to the large displacement and no overlap and only a small part of the total area is machined. Next, the topographical change introduced by the SPP-LIPSS arose from a short melting, wave excitation and fast solidification. If the excitation is not too strong, the wave does not break and no material is ejected. The material, which is ablated, is mainly evaporated during the fast temperature cycle.

For this study a laser with a pulse duration of 6.6 ps and structures with a large displacement have been applied. With this pulse duration and lateral displacement, the heat input into the samples was very small and is not expected to change the flatness. The LIPSS height of about 50 nm shows a local stochastic variation of a few percent of the total height at each 10 μm displaced location, compare Figure 6.15, and is subjected to an elastic deformation due to the parts in contact. That this structure variation can lead to a deformation of a stiff macroscopic counter part of millimetre geometries is also not expected. However, for both effects, particle introduction and structure height variation, an additional development step is required, e.g. to establish the effect of polishing after structuring.

In literature, often the effect of homogeneously structured areas on adhesion was investigated and in those studies a critical roughness value was determined which led to a reduction of adhesion. In those studies often a sphere or tip as counterpart was applied to avoid alignment problems and a point contact was established. In this way the problem can be spatially down scaled and investigated under laboratory conditions. A similar approach has been selected for this study. But in this case the critical roughness value also depends on the elasticity of the materials and the radius of curvature. But under industrial conditions this specific application is characterised by two flat counter parts with macroscopic scales of hundreds of millimetres and with a high degree of flatness and low roughness. The enormous adhesive forces are especially due to large and flat surfaces in contact.

The point contact measurement has also limitations in the analysis of different structure designs. Solutions, to reduce the contact area can also be grooves, dimples or a dimple-LIPSS pattern, as long as the flatness is not changed and other requirements, such as to avoid contamination, are fulfilled. With those structures a hybrid contact area is obtained and the total adhesion force for the components in contact is determined by the structure height and also by the untreated area in between. If the area of the point contact is smaller than the periodicity of those patterns, the overall effect of hybrid contact areas cannot be evaluated. In this study, the adhesion force has been determined multiple times on top of the LIPSS and found to be reduced. However, with a flat probe larger than the LIPSS dimples also the flat part of the area in between the LIPSS dimples would have had an influence and would have given an average value. For areas with a hybrid contact, the roughness value for adhesion

(a)

(b)

Figure 6.16: Force-displacement graph obtained during testing of a silicon wafer by the SI IR-lens. (a) Measurements on the reference sample in black colour and on the unstructured area of the laser treated sample in red colour. (b) Measurements on the unstructured area of the laser treated sample in black colour and structured areas in red, green and blue colour.

reduction can be expected to be higher than the 10 nm determined for homogeneous and stochastically distributed surface roughness [183]. Because the unstructured area will provide a higher adhesion, wherefore feature heights need to be higher as compensation. Here the elastic or plastic deformation of peaks, is playing an important role.

6.6 Conclusions

The results obtained in this thesis can be applied under different conditions in research and industry. In research two directions have been identified, first the study of LIPSS formation mechanisms and second the study of surface functionality. In industry the results can be applied where surface structuring is a solution to control component functionalities. In this thesis three different applications from three industrial sectors have been presented, surface cleaning in the automotive sector, tissue regeneration in the biomedical sector and precision positioning in the semiconductor industry. The application results confirm the potential of LIPSS to influence surface functionality. But beyond this observation, already made in literature, with this thesis a further step towards industrial applicability has been made.

The results obtained in this thesis can be applied in order to determine the structure portfolio for a given laser system and material combination, analyse the laser direct writing irradiation process, understand the spatial emergence of LIPSS, determine and optimise laser irradiation parameters and assess the risks of errors in laser surface structuring processes. Among these different ways for application of the results, the most important is the determination of irradiation parameters. The provided knowledge could be further applied, if the irradiation model, the static analysis and the approximate solutions would be implemented in the software of laser micro machining systems.

However, this final chapter brought another important aspect. The presented theory has been evaluated and confirmed for different laser wavelengths, pulse durations and irradiation materials. It was found that the approximation of irradiation parameters based on the analysis of static experiments is an effective and efficient approach to determine irradiation parameters. The results of this work were already applied while doing this research, which demonstrates their value for different applications. However in all application cases, it was found that a long and complex development chain with interdisciplinary research was needed to investigate and control surface functions by surface topography. These initial results show that more research is required in the future to understand the influence of LIPSS on surface functionality.

Chapter 7

Summary

7.1 In English - In het Engels

This research was triggered by potential applications for different industrial markets, as for example automotive engineering, semiconductor equipment and biomedical engineering. Driven by customer needs, companies have an economical interest to improve existing products. With respect to the mentioned industrial markets, contact mechanical, tribological or biomechanical surface phenomena play an important role and the growing understanding of these surface phenomena contributes to the design of industrial components.

For the improvement of an existing component, the manufacturing chain needs to be adapted, by selecting a manufacturing technology and adding additional manufacturing steps. The new design defines requirements on manufacturing technologies. In the case of the above-mentioned phenomena it is required to structure surfaces on a micrometre and even nanometre scale. Recently, laser-induced periodic surface structures, abbreviated LIPSS, gained more interest, because potential prospects are seen in the field of surface functionalisation. Therefore, this research deals with manufacturing technologies, in particular laser technologies, to structure surfaces.

The feasibility to produce LIPSS and the potential to influence surface functionality have been shown in research. But so far, the implementation as an industrial manufacturing process is limited. Whereas industrial laser sources and systems are available, knowledge about the application of this process was lacking. Therefore, the aim of this thesis was, to investigate the relation between certain irradiation parameters and the spatial emergence of LIPSS starting from the knowledge about incubation. This subject has been studied theoretically and experimentally.

In order to support the understanding of the spatial emergence of LIPSS, an irradiation model has been presented and the fluence accumulation process has been analysed for different irradiation conditions. Whereas, under SIC the individual fluence is constant and the accumulated fluence distribution is Gaussian, under DIC the individual fluence is varying and the accumulated fluence distribution is non-Gaussian. The irradiation model allows to calculate and characterise the irradiation applied by

a laser system, including irradiation errors, which was not only found to be useful for developing and understanding of the irradiation process, it was also required for the experimental investigation of the spatial emergence of LIPSS under DIC.

The spatial emergence of LIPSS is determined by the intersection of the applied accumulated fluence with the accumulated fluence domain boundaries. The location and size of the accumulated fluence domain vary, due to incubation, with the applied irradiation parameters. Due to the varying individual fluence, existing incubation laws cannot be applied to obtain accurate quantitative information about the domain location under DIC. However, they can be applied to qualitatively discuss the spatial emergence under DIC. The spatial emergence with increasing pulse overlap, is determined by the ratio of domain location variation to accumulated fluence increase. Based on this study, a new approach for the determination of irradiation parameters has been derived. The approach consists of two steps, first the determination of accumulated fluence domain boundaries under SIC and second the approximation of irradiation parameters fulfilling these requirements under DIC.

The theoretical results have been evaluated by irradiation experiments. This study covered the irradiation under SIC and 2d-DIC of a steel sample surface with laser pulses of $\lambda = 1030$ nm and $\tau = 6.6$ ps. Five different types of LIPSS have been observed and classified, three LSFL and two HSFL with periodicities in the range 400 nm to 3 μ m and orientations parallel and perpendicular to the polarisation direction. The experimental results showed a good agreement with the theoretical considerations. Both under SIC and DIC, the spatial emergence of LIPSS was found to be determined by an accumulated fluence domain, whose location and size vary due to the incubation properties of the material. It was found that the approximation of irradiation parameters based on the analysis of static experiments is an effective and efficient method. The use of static experiments allows to perform the empirical part of this approach in an efficient way and the approximations give a systematic control over the irradiation parameters.

The results obtained in this thesis can be applied under different conditions in research and industry. In this thesis three different applications from three industrial sectors have been presented, surface cleaning in the automotive sector, tissue regeneration in the biomedical sector and precision positioning in the semiconductor industry. The application results confirm the potential of LIPSS to influence surface functionality. By performing application projects, the presented theory has been evaluated and confirmed for different laser wavelengths, pulse durations and irradiation materials. The results of this thesis can be applied in order to determine the structure portfolio for a given laser system and material combination; analyse the laser direct writing irradiation process; understand the spatial emergence of LIPSS; determine and optimise laser irradiation parameters and assess the risks of errors in laser surface structuring processes.

But the obtained results also identify interesting directions for further research. The influence of a varying individual fluence on accumulated fluence domain boundaries, due to incubation, is relevant for machining extended areas of LIPSS and for applications with crucial damage threshold requirements. The provided knowledge could be further applied, if the irradiation model, static analysis and approximate

solutions would be implemented in the software of laser micro machining systems. In the presented applications, it was found that a long and complex development chain with interdisciplinary research was needed to investigate and control surface functions by surface topography. These initial results show that more research is required in the future to understand the influence of LIPSS on surface functionality.

7.2 In Dutch - In het Nederlands

Het onderzoek is geïnitieerd door mogelijke toepassingen, bijvoorbeeld in de automotive markt, halfgeleider industrie en de biomedische sector. Voor bedrijven in deze sectoren is het belangrijk bestaande producten te verbeteren om tegemoet te komen aan de wensen van klanten. Hierbij spelen contactmechanica, tribologie en biomechanica een belangrijke rol en het groeiend begrip van de oppervlakteverschijnselen draagt bij aan productvernieuwing.

Verbetering van een bestaand product of component daarvan vereist aanpassing van de productieketen door de keuze van een fabricagetechnologie en de toevoeging van extra fabricagestappen. Het nieuwe ontwerp stelt eisen aan de fabricagetechnologie. Bovengenoemde oppervlakteverschijnselen maken het noodzakelijk oppervlakken te structureren op micrometer- of zelfs nanometerschaal. Recent kregen laser geïnduceerde periodieke oppervlaktestructuren (LIPSS) meer belangstelling vanwege mogelijke toepassingen op het gebied van functionele oppervlakken. Daarom richt dit onderzoek zich op fabricagetechnologie, i.h.b. lasertechnologie om oppervlakken te structureren.

Hoewel in onderzoek is aangetoond dat het haalbaar is LIPSS te produceren en zo de oppervlaktefunctionaliteit te beïnvloeden, is de industriële implementatie tot nu toe beperkt gebleven. Terwijl industriële laserbronnen en -systemen beschikbaar zijn, ontbrak de kennis over de toepassing van dit fabricageproces. Daarom is het doel van dit proefschrift de relatie tussen de parameters van laserstraling en de ruimtelijke verschijning van LIPSS te onderzoeken vanuit de kennis van incubatie. Dit onderwerp is zowel theoretisch als experimenteel bestudeerd.

Om het begrip van de ruimtelijke verschijning van LIPSS te verbeteren, is een laser stralingsmodel opgesteld en de accumulatie van laserenergie (fluence) is geanalyseerd voor verschillende stralingscondities. Onder statische condities (SIC) is de fluence ten gevolge van een laserpuls constant en de geaccumuleerde fluence heeft een Gaussische verdeling. Onder dynamische condities (DIC) varieert de afzonderlijke fluence en de geaccumuleerde fluence is niet langer Gaussisch. Het stralingsmodel maakt het mogelijk de straling te berekenen en te karakteriseren, inclusief de afwijkingen hierin. Het model bleek niet alleen nuttig voor de ontwikkeling en het begrip van het stralingsproces, maar was ook nodig voor het experimentele onderzoek naar de ruimtelijke verdeling van LIPSS onder DIC.

De ruimtelijke verschijning van LIPSS wordt bepaald door de ligging van de gebruikte geaccumuleerde fluence niveau's ten opzichte van de domeingrenzen. Ten gevolge van incubatie hangen de locatie en de afmeting van een fluence domein af van de gebruikte laser-proces-parameters. Door de variatie in afzonderlijke fluence kunnen

bestaande incubatie wetten niet toegepast worden voor een nauwkeurige kwantitatieve beschrijving van de domeinen onder DIC, maar wel voor een kwalitatieve beschouwing van de ruimtelijke verschijning van LIPSS. Bij toenemende pulsoverlap wordt deze verschijning bepaald door de verhouding van de variatie in domeinpositie en de toename in geaccumuleerde fluence. Daarmee legt deze studie de basis voor een nieuwe benadering voor de bepaling van laser-proces-parameters. Deze benadering bestaat uit twee stappen: eerst de bepaling van de domeingrenzen van geaccumuleerde fluence onder SIC en vervolgens de afschatting van de laser-proces-parameters onder DIC.

De theoretische resultaten zijn geëvalueerd met laser experimenten die bestonden uit SIC en 2d-DIC bestraling van staal met laserpulsen, $\lambda = 1030$ nm en $\tau = 6.6$ ps. Vijf verschillende types LIPSS zijn waargenomen en ingedeeld, drie zijn van het LSFL type en twee van het HSFL type met een periodiciteit in het bereik van 400 nm tot 3 μ m en oriëntaties parallel en loodrecht op de polarisatie richting. De experimentele resultaten stemmen goed overeen met de theorie. Zowel bij SIC als DIC blijkt de ruimtelijke verschijning van LIPSS bepaald te worden door een geaccumuleerd fluence domein, waarvan de plaats en de afmeting variëren door de incubatie-eigenschappen van het materiaal. Gevonden is dat de afschatting van de procesparameters op basis van SIC experimenten een effectieve en efficiënte methode is. Door gebruik van SIC experimenten kan het empirische deel van deze benadering efficiënt uitgevoerd worden en de afschattingen geven een systematische beheersing van de proces parameters.

De resultaten van dit proefschrift zijn toepasbaar onder verschillende omstandigheden in onderzoek en industrie. In dit proefschrift worden drie toepassingen uit drie industriële sectoren behandeld, oppervlaktereiniging in de automotive sector, weefselherstel in de bio-medische sector en precisie positionering in de halfgeleiderindustrie. De resultaten van deze toepassingen bevestigen de mogelijkheid om met LIPSS de oppervlaktefunctionaliteit te beïnvloeden. Door de applicatieprojecten kon de theorie geëvalueerd en bevestigd worden voor verschillende laser golflengtes en pulsduren en voor verschillende materialen. De resultaten van dit proefschrift kunnen gebruikt worden voor het bepalen van de structuur portfolio van een gegeven laser systeem in combinatie met een materiaal; voor analyse van het laser "direct-write"proces; voor het begrijpen van de ruimtelijke verschijning van LIPSS; voor het bepalen en optimaliseren van laser-proces-parameters en voor een evaluatie van de risico's op fouten bij laser-oppervlakte-structurering.

De verkregen resultaten laten ook interessante richtingen voor verder onderzoek zien. De invloed van variaties in de afzonderlijke fluence op de domeingrenzen van geaccumuleerde fluence ten gevolge van incubatie, is van belang voor het bewerken van een groot oppervlak met LIPSS en voor toepassingen met kritische eisen aan de schadedrempels. De gegenereerde kennis zou verder toegepast kunnen worden als het stralingsmodel, de statische analyse en afschattingen opgenomen worden in de software van laser micro bewerkingssystemen. In de getoonde toepassingen is gevonden dat een lange en complexe ontwikkelingsketen met interdisciplinair onderzoek nodig is voor beheersing van oppervlakte-functies door oppervlakte topografie. Deze eerste resultaten laten zien dat in de toekomst meer onderzoek nodig is om de invloed van LIPSS op oppervlakte-functionaliteit te begrijpen.

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Publications

Journal contributions

J. Vincenc Obona, J.Z.P. Skolski, J. Eichstädt, V. Ocelík, G.R.B.E Römer, J.Th.M. De Hosson and A.J. Huis in't Veld, Modification of Si surface with a picosecond laser, Submitted to the journal: Applied Surface Science, 2015.

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Invited talks

J. Eichstädt, The spatial emergence of laser-induced periodic surface structures under lateral displacement irradiation conditions, Seminar Oberflächentechnologien der Bundesanstalt für Materialforschung und -prüfung, Fachbereich 6.4 Technologien mit Nanowerkstoffen, Berlin, Germany, November 8, 2012.

J. Eichstädt, Simulation von Energiedichteprofilen zur Laserstrukturierung kontaktmechanischer Oberflächen, Seminar Laser in der Feinbearbeitung des Laserverbundes Berlin-Brandenburg, Brandenburg, Germany, April 19, 2012.

Conference presentations

J. Eichstädt, Influence of errors on the irradiation process for laser-induced periodic surface structures, Seventh International WLT Conference on Lasers in Manufacturing (LiM), Munich, Germany, May 13-16, 2013.

J. Eichstädt, Formation of laser-induced periodic surface structures under lateral displacement irradiation conditions, 2nd Symposium on the growth mechanism of LIPSS, Cottbus, Germany, October 4, 2012.

J. Eichstädt, Towards Friction Control using laser-induced periodic Surface Structures, Sixth International WLT Conference on Lasers in Manufacturing (LiM), Munich, Germany, May 23-26, 2011.

Posters

J. Eichstädt, Irradiation analysis for laser-induced periodic surface structures, Industrial Laser Event 2013, University of Twente, Enschede, The Netherlands, April 16, 2013. Impact, EM.

J. Eichstädt, Simulation of energy density profiles for laser-induced surface phenomena, Industrial Laser Event 2012, University of Twente, Enschede, The Netherlands, April 17, 2012. Impact, EM.

J. Eichstädt, The opportunities of laser induced periodic surface structures for tribological applications, Gordon Research Conference - Tribology, Waterville, USA, June 27, 2010. Impact, EM.

J. Eichstädt, Topographical characterization of laser texture, Industrial Laser Event 2010, University of Twente, Enschede, The Netherlands, March 31, 2010. Impact, EM.

Nomenclature

Notations

x	A scalar x
$\vec{x}$	A vector $\vec{x}$
$\mathbf{x}$	A matrix $\mathbf{x}$
$\bullet_x$	Denotes variables in x direction
$\bullet_y$	Denotes variables in y direction
$\hat{\bullet}$	Denotes an effective value
$\tilde{\bullet}$	Denotes a relative value
$\bullet'$	Denotes variables of the secondary kinematical system

Abbreviations

1d-DIC	One dimensional DIC
2d-DIC	Two dimensional DIC
AFM	Atomic force microscopy
BD	Beam dump
BK-7	Borosilicate crown glass
CAD	Computer-aided design
CAE	Computer-aided engineering
CAM	Computer-aided manufacturing
CCD	Charge-coupled device
CLSM	Confocal laser scanning microscopy
DIC	Dynamic irradiation conditions
DIN	German: Deutsches Institut für Normung e. V.; English: German institute for standardisation
EDM	Electro-discharge machining
EDS	Energy-dispersive X-ray spectroscopy
EN	German: Europäische Norm; English: European standard
Exp	Experiment

FT	Fourier transform	
G abs	Peak of the accumulated fluence or Γ_0	J m^{-2}
G rel	Relative accumulated peak fluence or $\tilde{\Gamma}_p$	
HMSC	Human mesenchymal stem cells	
HODIC	High overlap dynamic irradiation conditions	
HSFL	High spatial frequency LIPSS	
IR	Infrared	
ISO	German: Internationale Organisation für Normung; English: International organisation for standardisation	
LASER	Light amplification by stimulated emission of radiation	
LBM	Laser beam machining	
LIPSS	Laser-induced periodic surface structures	
LODIC	Low overlap dynamic irradiation conditions	
LSFL	Low spatial frequency LIPSS	
M1-6	Mirror 1-6	
MA	Manually	
MODIC	Medium overlap dynamic irradiation conditions	
MTP	Maximum turning point	
OEM	Original equipment manufacturer	
OL	Pulse overlap or φ	
OM	Optical microscopy	
P	Periscope	
PBS	Polarising beam splitter	
PhD	Englisch: Doctor of Philosophy; Latin: philosophiae doctor	
PP	Polypropylene	
Prox	Approximation	
SBC	Styrene butadiene copolymer	
SEM	Scanning electron microscopy	
SIC	Static irradiation conditions	
SM	Surface modification	
SOM	Stereo optical microscopy	
SPP	Surface plasmon polariton	
TULP	Texturing using ultra short laser pulses	
WCA	Water contact angle	

Latin symbols

c	Velocity of light in a medium	m s^{-1}
c_p	Relative contribution per pulse	
D	Outer diameter of a surface modification	m
d	Ablation depth	m

$\vec{E}$	Electric field strength	V m^{-1}
E_p	Pulse energy	J
e	Number of matrix elements	
F_A	Adhesion force	N
F_N	Normal load	N
f_e	Error frequency	s ⁻¹
f_l	Focal length	m
f_r	Pulse frequency	s ⁻¹
g	Integer for lateral displacements in y direction	
$\vec{H}$	Magnetic field strength	A m^{-1}
I	Irradiance or intensity	W m^{-2}
I_0	Peak intensity	W m^{-2}
i	Indices for data points in matrices or indice for domain numbers	
j	Indices for data points in matrices or indice for domain values	
k	Extinction coefficient or integer for lateral displacements in x direction	
l	Geometrical length of the irradiated area	m
M^2	Beam quality factor	
N_{a-f}	Pulse boundaries	
N_p	Number of pulses	
N_r	Number of irradiation repetitions	
N_{th}	Pulse number threshold or repetition threshold	
N_x	Number of displacements for a half side in x direction	
N_y	Number of displacements for a half side in y direction	
n	Refractive index or number of the pulse	
P	Power	W
P_0	Peak power	W
P_a	Average power	W
P_e	Amplitude of the power error	W
$\vec{p}$	P-polarisation	
q	Spatial resolution of the space domain	m
R_a	Average roughness of a surface profile	m
R_q	Root mean square roughness of a surface profile	m
r	Radial coordinate	m
$\vec{S}$	Poynting vector	W m^{-2}
$\vec{s}$	S-polarisation	
T_{1-6}	Irradiation periods	s
T_c	Period of an optical cycle	s
T_r	Pulse period	s
t	Time	s
v	Velocity	m s^{-1}

x	Cartesian coordinate	m
Δx	Absolute intersection in x direction	m
Δy	Absolute intersection in y direction	m
x_0	Absolute displacement increment in x direction	m
x_e	Statistical position errors in x direction	m
x_{e0}	Amplitude of statistical position errors in x direction	m
x_n	Sequence of displacements in x direction or actual position	m
x_s	Starting coordinate in x direction	m
x_Φ	Phase shift error	m
$x_{\Phi 0}$	Amplitude of the phase shift error	m
y	Cartesian coordinate	m
y_0	Absolute displacement increment in y direction	m
y_e	Statistical position errors in y direction	m
y_{e0}	Amplitude of statistical position errors in y direction	m
y_n	Sequence of displacements in y direction or actual position	m
y_s	Starting coordinate in y direction	m
z	Cartesian coordinate	m
z_0	Axial alignment error	m
z_R	Rayleigh length	m

Greek symbols

Γ	Accumulated fluence	J m ⁻²
Γ_0	Peak of the accumulated fluence or G abs	J m ⁻²
Γ_a	Average accumulated fluence	J m ⁻²
Γ_c	Accumulated centre fluence	J m ⁻²
Γ_d	Accumulated peak-to-valley difference fluence	J m ⁻²
Γ_p	Accumulated peak fluence	J m ⁻²
Γ_{th}	Accumulated fluence threshold	J m ⁻²
Γ_v	Accumulated valley fluence	J m ⁻²
γ	Variable to solve the fluence, energy and power integral	
δ	Relative displacement	
ε	Ellipticity factor	
Θ	Orientation of LIPSS	°
θ	Angle of incidence	°
ϑ	Switch for the translation direction	
Λ	Periodicity of LIPSS	m
λ	Central wavelength	m
$\Delta\lambda$	Spectral linewidth	m

ξ	Incubation factor	
ρ	Factor for the domain step size	
σ	Factor for the domain step number	
ς	Switch for the translation axes	
τ	Pulse duration	s
Φ	Phase shift line period	
ϕ	Individual fluence	J m ⁻²
$\Delta\phi$	Individual fluence variation	J m ⁻²
ϕ_0	Individual peak fluence	J m ⁻²
ϕ_a	Average individual fluence	J m ⁻²
ϕ_d	Individual peak-to-valley difference fluence	J m ⁻²
ϕ_p	Individual peak fluence	J m ⁻²
ϕ_{th}	Individual fluence threshold	J m ⁻²
ϕ_v	Individual valley fluence	J m ⁻²
φ	Pulse overlap or OL	
Ψ	Relative accumulation factor	
$\vec{\psi}$	Cartesian position vector	m
$\vec{\psi}_a$	Analysis locations	m
$\vec{\psi}_d$	Irradiated locations	m
Ω_d	Pseudorandom values for displacement errors	
Ω_p	Pseudorandom values for power errors	
ω	Focused beam radius	m
ω_0	Focused beam radius in the focal plane	m
$\tilde{\omega}_0$	Unfocused beam radius in the waist centre	m

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Dedicated to Franzi

About the Author

Justus Eichstädt was born on 22 September 1982 in Rathenow, Germany, is married with Franziska and has a daughter Marlene. He finished secondary school at J.H.A. Duncker Gymnasium Rathenow in 2002 with specialisation in physics and biology. Afterwards, he studied ophthalmic optics at Ostfalia University of Applied Sciences and obtained a diploma degree in 2007. The diploma thesis was entitled with: "Anpassung eines Subaperture-Stitching-Interferometers an die Prüfung der Zylinderform optischer Komponenten". The study was balanced between optical, medical and economical sciences. In parallel, he attended the ophthalmic optics school in Hankensbüttel and received a certificate of apprenticeship in 2005. Alongside his scientific education he started working at Menz Optic AG in 2002 and at Berliner Glas KGaA in 2005.

After the diploma study he attended a post-graduate master program, which contained courses in photonics at Beuth University of Applied Sciences, Technical University of Berlin, Brandenburg University of Applied Sciences and Wildau University of Applied Sciences and obtained a master degree in 2008. The master thesis was entitled with: "Untersuchung des Abtragverhaltens einer SiSiC-Keramik mit einem gepulsten, frequenzkonvertierten Festkörperlaser". Justus Eichstädt was graduated with an award for the best alumnus of the years 2008/2009. In parallel and afterwards to the master program he continued working at Berliner Glas KGaA in the development department for manufacturing technologies. From 2009 to 2014 he worked as researcher at the University of Twente. Presently, he works at a professional school for ophthalmic optics in Rathenow.

This research was triggered by potential applications for different industrial markets, as, for example, automotive engineering, semiconductor equipment and biomedical engineering. Driven by customer needs, companies have an economical interest to improve existing products. With respect to the mentioned industrial markets, contact mechanical, tribological or biomechanical surface phenomena play an important role and the their growing understanding contributes to the improvement of surface functions of industrial components. To implement an improved design for an existing component, the manufacturing chain needs to be adapted for an additional structuring process. This requires to select a manufacturing technology and adding additional manufacturing steps. The new design defines requirements on manufacturing technologies. In the case of the above-mentioned phenomena it is required to structure surfaces on a micrometre and even nanometre scale. Recently, laser-induced periodic surface structures, abbreviated LIPSS, gained more interest, because potential prospects are seen in the field of surface functionalisation. The feasibility to produce LIPSS and the potential to influence surface functionality have been shown in research. But so far, the implementation as an industrial manufacturing process is limited. Whereas industrial laser sources and systems are available, knowledge about the application of this process was lacking. Therefore, the aim of this thesis was, to investigate the relation between certain laser irradiation parameters and the spatial emergence of LIPSS. This subject has been studied theoretically and experimentally.