Introduction

Laser induced periodic surface structures (LIPSS) often termed "ripples", have been an active field of research ever since their discovery in 1965 [Bir65], shortly after the realization of the first laser in 1960 [Mai60].

The early investigation of ripple patterns with nanosecond laser pulses, showed a periodicity depending on the laser wavelengths and on the angle of incidence. Moreover, most patterns exhibit an orientation perpendicular to the laser polarization. Thus, an interference effect of the incident laser and a secondary surface electromagnetic wave (e.g. generated by scattering of laser irradiation) was suggested, as the fundamental physical process involved in the formation of these structures [SYP+83] (often referred to as "classical ripple theory").

Through the investigation of LIPSS with femtosecond laser pulses a variety of other structures as well as ripple patterns with spatial periods well below the incident laser wavelengths were observed. The classical ripple theory has therefore been expanded to the, in this thesis referred to as *static* model of structure formation [BRK09], [GCP+11]. The so called "femtosecond ripples" share many common characteristics in the morphological appearance (bifurcation and truncations of lines, dot & columnar structures) with self-organized pattens from induced surface instability after ion beam bombardment [ZCF+09]. Furthermore, the structure sizes produced upon femtosecond laser irradiation or ion beam sputtering, seem to depend on the laser intensity/ ion energy and irradiation time as well as on a positive feedback, which results in the coalescence of structures. This leads to the suggestion that other physical process may be involved in the formation process. The adaptation of the theory of ion beam sputtering [Sig69], [BHa88], [Var13b] gave rise to the *dynamic* model of self-organized LIPSS formation.

Despite numerous experimental data on surface patterning upon femtosecond laser irradiation, the microscopic mechanisms of LIPSS formation are still under discussion. Supporting and contrary arguments for the two formation models, can be found in the observed features of the LIPSS patterns. For instance, grating-like structures with spatial periods in the magnitude of the incident wavelength ($\Lambda_{\text{LIPSS}} \sim \lambda_{laser}$), may result from an inhomogeneous ablation through the interference pattern of the interacting waves. Thereby, the monochromaticity and high spatial coherence of the laser, are essential properties for inference and, thus, formation of periodic ripple patterns according to the *static* model.

On the other hand, spherical nano-dots upon irradiation with circular polarized laser light, bifurcating ripples, and patterns with periods larger or significantly smaller than the laser wavelength are features which cannot be accounted for by an optical interference effect. They can, however, be explained in the framework of a self-organization process from laser-induced instability [Cos06], [Rei10], [Var13b].

To shed light on the controversy between these two formation models, the LIPSS formation ought to be investigated with a polychromatic light source of reduced spatial coherence. As a result of this, an optical interference effect might be excluded or confirmed as the driving physical process in LIPSS formation.

This open question has been the major motivation for this master thesis. Spontaneous pattern formation upon irradiation with incoherent white-light from an incandescent light bulb, has already been reported in photo-refractive crystals [TBS04], [TTB+04], [KSE+00]. However, to study the features of the "femtosecond ripples", the intended light source further needs to provide intense pulses in the ultrafast time regime, with duration below one picosecond ($\tau_p \leq 1$ps).

Since 1970, it has been known that narrow bandwidth laser pulses can undergo an extreme spectral broadening upon propagation through a nonlinear optical medium [ASh70]. The so called "generation of a white-light supercontinuum" is nowadays an active field of research, since the applications of the white-light are very attractive to many scientific fields, e.g. optical tomography [HLC+01]. The resulting white-light beam, thereby, exhibits very distinct characteristics in terms of spatial wavelengths dependence as well as spatial beam properties at varying input laser powers. Most importantly the white-light pulses stay ultrafast light pulses upon propagation through a nonlinear optical medium of short length [QWW+01], [NOS+96].

The investigation of the spatial coherence of the white-light continuum shows two different sides, depending of the input power of the pumping laser. It has been published that, the white-light retains the high spatial coherence of the pump-laser [WIt01], [CPB+99] for low input powers, making it a polychromatic laser source. However, for large input powers, it has been reported [BIC96], [GSE86] that the white-light looses its spatial coherence.

For these reasons, it has been a motivation, to characterize the generated white-light continuum, previous to the study of the LIPSS formation.

The goal of this work is to study the possibility of LIPSS formation upon irradiation with a suitable white-light continuum of sufficient WL pulse energy, broad spectral bandwidth and low spatial coherence. Furthermore, to discriminate between both LIPSS formation models by experimentally studying the importance of laser wavelength and coherence vs. irradiation dose on the characteristic features of LIPSS.

Thesis Organization

The first two chapters of this thesis are intended to give a brief theoretical introduction into the two major scientific fields of laser ablation & laser surface structuring and nonlinear optics. Presented in **chapter one**, are the favored mechanism of femtosecond laser ablation on metals and semiconductors as well as the two formation models of LIPSS. The, in this work, preferred terminology of the structures and spatial period lengths is also given here.

In **chapter two**, the nonlinear optical processes responsible for the spectral broadening of laser pulses (i.e. continuum generation) with a focus on the used nonlinear optical medium Sapphire is discussed.

The experimental setup for the characterization of the white-light continuum and studies of LIPSS is presented in **chapter three**.

Chapter four discusses the experimental results from the characterization of the white-light continuum with a focus on energy conversion efficiency, white-light polarization, spatial beam properties, long term stability and spatial coherence.

In **chapter five** the study of LIPSS upon irradiation with the white-light continuum is presented. The investigation of the produced structures focuses on the multi-pulse feedback effect as well as on the identification and characterization of similarities in the observed structures to self-organized surface patterns upon laser irradiation or ion beam sputtering. Furthermore, the effect of the white-light polarization on the orientation of the ripple patterns as well as energy dispersive x-ray measurements of the surface element distribution after irradiation in air atmosphere are discussed.

The conclusion of this work and outlook on future experiments is given in **chapter six**

In addition, the determination and detailed description of the pulse duration and intensity distribution in temporal and spatial domain, as well as the beam propagation of the laser beam that was used in the experiments of this work are presented in the **appendix**.

Publication, Presentations and Posters to this Work

1. J. Reif, O. Varlamova, S. Uhlig, S.Varlamov, M. Bestehorn. Formation of self-organized LIPSS by irradiation with an ultrafast White-Light Continuum. *Proceedings of LPM 2014 - 15th international Symposium on laser precision microfabrication*

2. S. Uhlig, O. Varlamova, M. Ratzke, J. Reif. Formation of self-organized LIPSS by irradiation with an ultrafast White-Light Continuum. *Poster at EMRS Spring Meeting 2014*

3. J. Reif, O. Varlamova, S. Uhlig, S. Varlamov, M. Bestehorn. On the Physics of Self-Organized Nanostructure Formation upon Femtosecond Laser Ablation. *Applied Physics A,114(4):1-6,2014*, ISSN 0947-8396, `http://dx.doi.org/10.1007/s00339-014-8339-x`

4. J. Reif, O. Varlamova, S. Uhlig, C. Martens. Genesis of Femtosecond-Induced Nanostructures on Solid Surfaces. *Presentation at 3rd workshop on LIPSS (2nd circular), Berlin November 7th 2013*

5. J. Reif, O. Varlamova, S. Uhlig, S.Varlamov, M. Bestehorn. On the Physics of Self-Organized Nanostructure Formation upon Femtosecond Laser Ablation. *Proceedings of LMP 2013 - 14th International Symposium on Laser Precision Microfabrication*

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