



# Formation of laser-induced periodic surface structures (LIPSS) on tool steel by multiple picosecond laser pulses of different polarizations



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## ABSTRACT

Laser-induced periodic surface structures (LIPSS) are produced on cold work tool steel by irradiation with a low number of picosecond laser pulses. As expected, the ripples, with a period of about 90% of the laser wavelength, are oriented perpendicular to the laser polarization. Subsequent irradiation with the polarization rotated by 45° or 90° results in a corresponding rotation of the ripples. This is visible already with the first pulse and becomes almost complete – erasing the previous orientation – after as few as three pulses. The phenomenon is not only observed for single-spot irradiation but also for writing long coherent traces. The experimental results strongly defy the role of surface plasmon-polaritons as the predominant key to LIPSS formation.

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## 1. Introduction

Laser-induced periodic surface structures (LIPSS) have been intensively studied on different materials in the last half of the century [1–8]. They can be observed in all kinds of solids, when irradiated near their ablation threshold [9]. LIPSS appear as a regular ripple structure with a period that often scales with the laser wavelength [5], while its orientation and shape are defined by the polarization of the incident light [10,11]. Since surface morphology is a key factor in controlling the optical, mechanical or chemical properties of a solid surface, laser-induced surface modification found great interest in a wide range of different applications in photonics, biomedicine, heat transfer, wettability, tribology, and other areas [12–15].

The underlying physical mechanisms of LIPSS formation are still under debate. However, two different approaches can be found in the literature. The first one is the generalized scattering and interference model [16,17] assuming that ripples are based on lithographic ablation after a modulated energy deposition, caused by an optical interference. The interference has often been attributed to the excitation of surface plasmon-polaritons (SPP) [18], since they can be easily addressed on a regularly corrugated surface [19,20] with an optical resonance wavelength slightly larger

than their spatial period. This model explains very well structures consisting of long and almost parallel lines with periods close to the laser wavelength. But, it is not able to describe the dependence of the ripple period on the absorbed laser fluence and more complex features, such as multiple bifurcations [21]. The second approach to explain LIPSS formation is known as a self-organization model [21,22]. It is very similar to models of surface structuring by ion beam sputtering and is based on the dynamics of energetic pulse interaction with the target material. By assuming that the absorbed laser energy leads to instabilities of the surface region due to softening and perturbation of crystal binding, it can explain a spontaneous formation of surface structures within the laser-modified area [21]. Moreover, the self-organization approach can also explain the recent observation of coherently connected LIPSS patterns in lines of individual spots with almost no spatial or temporal overlap [7].

In all experiments and all models, the laser polarization is a very important parameter during LIPSS formation, since the pattern morphology and orientation are controlled by the polarization of the laser-pulse electric field [4,9,11,23]. The main focus of the present study lies on the investigation of the strictness of this polarization dependence of the ripples on a metal surface. For this purpose we followed the pulse-to-pulse evolution of the LIPSS formation when changing the polarization by 45° or 90° in the course of trains of picosecond-laser pulses. The experiments were performed both on a single spot as well as in line scans with 50% overlap between the adjacent spots with the laser radiation blocked

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during the target displacement. As a target we chose a polished surface of K890 cold work tool steel that is designed for high ductility, exhibiting a high capacity for plastic yield and high fatigue strength. Therefore, it is very suitable for tooling which requires high-edge stability. The surface patterning may be of importance for improvement in tribology approaches, where LIPSS can significantly reduce the friction [13].

## 2. Experimental

In all experiments, the fundamental ( $\lambda_0 = 1064 \text{ nm}$ ) of a Nd:YAG laser (Ekspla, Lithuania, PL2250-SH-TH) was focused onto the sample surface in nearly normal incidence under atmospheric pressure. The laser emits linearly polarized pulses of 30 ps duration (full width at half maximum; FWHM) at a maximum repetition rate of 50 Hz, which can be externally triggered. The laser beam was expanded by a  $4 \times$  telescope to a diameter of 12 mm. In order to achieve a more circular spot, we placed a diaphragm with a diameter of 5 mm just behind the beam expander. A half-wave plate (HWP) in a rotation stage with an angular resolution of  $0.3^\circ$  was placed just before the focusing objective ( $f = 120 \text{ mm}$ ), yielding a focal spot of  $60 \mu\text{m}$  diameter. The pulse energy, measured by a pyroelectric detector (Gentec Electro-Optics, Inc., Canada, Solo with head QE12LP-H-MB) after the focusing objective, was set to  $33 \mu\text{J}$  during all our experiments, with a standard deviation, calculated from 500 pulses, of  $\pm 1.3 \mu\text{J}$ .

As a target material we used cold work tool steel K890 (Böhler Edelstahl, Kapfenberg/Austria), based on powder metallurgy, consisting of 0.85% C, 0.55% Si, 0.40% Mn, 4.35% Cr, 2.80% Mo, 2.10% V, 2.55% W and 4.50% Co. After machining, the K890 specimen was quenched and triple tempered according to the steel producer specifications ( $T_A = 1100^\circ\text{C}$ ,  $T_T = 500^\circ\text{C}$ ), achieving hardness of 64 HRC. Its surface was highly polished and ultrasonically cleaned in ethanol and distilled water before the processing.

The target (a rectangular cuboid with dimensions of  $20 \times 20 \times 8 \text{ mm}^3$ ) was placed in a 3D positioning system, equipped with 3 linear stages (Eksma Optics, Lithuania) with  $1 \mu\text{m}$  resolution. Here, the  $z$  direction is parallel with the excitation-pulse path, while the  $x$  axis is perpendicular to the  $z$  direction and parallel with an optical table. The  $y$  stage is tilted by about  $5^\circ$  in order to avoid the back reflection into the laser; so, the angle of incidence equals  $85^\circ$ . After processing, the samples were analyzed *ex-situ* by a scanning electron microscope (SEM; JSM-6500F) and by an atomic force microscope (AFM; Veeco, Dimension 3100).

## 3. Results

### 3.1. Surface morphology as a function of focal position

In a first set of experiments, we searched for the best focusing condition to obtain a spot with a most homogeneous ripple pattern. For this purpose, we moved the target along the  $z$ -axis around the focus plane ( $z = 0$ ) within the range  $-3 \text{ mm} \leq z \leq 3 \text{ mm}$ . At each position, a constant number of pulses ( $N = 3$ ) was applied onto a single spot. For each new spot we moved the sample for  $\Delta x = 150 \mu\text{m}$  to radiate a fresh patch of its surface. The images in Figs. 1(a)–(f) represent typical surface structures at different positions around the focal plane, Figs. 1(g)–(i) are magnifications from indicated spot areas. The double arrow under Fig. 1(g) shows the orientation of the pulse polarization.

When the surface is significantly out of the focus, at low fluence, only high-spatial frequency LIPSS (HSFL) [24] are generated with the orientation parallel to the light polarization and the period significantly below the laser wavelength, as clearly visible from Fig. 1(g), which is a magnification of Fig. 1(a). By approach-

ing the surface to the focus position, the fluence increases and low-spatial frequency LIPSS (LSFL) with period around the pulse wavelength and orientation perpendicular to the pulse polarization [5,6] appears in the center of the processed area, as visible from Figs. 1(b)–(d) and (h). However, if the surface is moved further towards the focal position, the local fluence of the central part of the beam exceeds the melting threshold of the material. This melting results in a blurring of the ripples structures, as is revealed from Figs. 1(e) and (f). Thus, on the target near the focus position three regions with different morphology [25], R1–R3, exist [e.g., see Fig. 1(i)]. Region R3 exhibits HSFL with a period of around 200 nm and orientation parallel to the laser polarization; region R2 shows LSFL with period  $0.97 \mu\text{m}$  and orientation perpendicular to the polarization, while region R1 corresponds to the region, where periodic LSFL is already blurred because of surface melting. It should be noted here, that we have carefully checked that R1 is not a fictive effect caused by the crater depth, defocusing the SEM measurements.

The three different regions, observed in Fig. 1, can be explained by a Gaussian beam profile, as schematically presented in Fig. 2(a). In this case, the regions R1–R3 appear due to different fluence thresholds  $F_{R1}$ ,  $F_{R2}$  and  $F_{R3}$ . For a Gaussian spatial profile (diffraction), the fluence  $F(z, r)$  as a function of focus position  $z$  and radius  $r$  is given by:

$$F(z, r) = F_0 \frac{w_0^2}{w^2(z)} \exp(-2r^2/w^2(z)) \quad (1)$$

where  $F_0$  is the peak fluence at  $r = 0$  and  $z = 0$ ,  $w_0$  stands for the  $1/e^2$  beam waist at the focus position (at  $z = 0$ ) and beam radius  $w(z)$  as a function of  $z$  depends on the laser wavelength  $\lambda$ :

$$w(z) = w_0 \sqrt{1 + \frac{z^2 \lambda^2}{\pi^2 w_0^4}} \quad (2)$$

The correlation to the pulse energy is given by the integral over the profile and does not depend on the  $z$ -position:

$$E_{pulse} = F_0 \frac{w_0^2}{w^2(z)} \int_0^\infty \exp(-2r^2/w^2(z)) \times 2\pi r dr = \frac{1}{2} \pi F_0 w_0^2 \quad (3)$$

The local fluence reaches the threshold for a specific process  $F_{th} = \alpha F_0$  at radius  $r_{th}$ . Here,  $0 \leq \alpha \leq 1$  is a dimensionless parameter. Combining (1) and (2), one can obtain the  $z$ -dependence of radius  $r_{th}$ , where the pulse intensity reaches the threshold  $F_{th}$ , as:

$$r_{th}(z) = w_0 \sqrt{\eta(z)} \quad (4)$$

The dimensionless factor  $\eta(z)$  in Eq. (4) is:

$$\eta(z) = \frac{1}{2} \frac{w^2(z)}{w_0^2} \ln \left( \frac{1}{\alpha} \frac{w_0^2}{w^2(z)} \right) \quad (5)$$

By fitting a circle to the edges of the regions R1–R3 in micrographs like those in Fig. 1, we measured the radii of these regions. They are presented as triangles (R1), circles (R2) and squares (R3) in Fig. 2(b). The solid curves in Fig. 2(b) show the fit of Eq. (4) to the measured threshold radii. As the fitting parameters we obtained the dimensionless coefficients for regions R1–R3:  $\alpha_1 = 0.28$ ,  $\alpha_2 = 0.42$ , and  $\alpha_3 = 0.67$ . Additionally, the fit gives also the beam waist radius  $w_0 = 30 \mu\text{m}$ , which agrees fairly well with a nearly diffraction limited beam for our experimental parameters.

The border between regions R3 and R2 are very clear. Therefore, we obtained an excellent fit of the measured data for R2. Contrarily, it is difficult to determine the radius of region R1, since the edge between the molten region and LSFL is not very sharp. This results in a slight discrepancy between the measured data and the fit in the case of region R1. On the other hand, the significant deviation of the measured data from the fit in the case of R3 can be explained

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