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Polarisation-dependent generation of fs-laser induced periodic surface structures

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ABSTRACT

The formation of laser induced periodic surface structures (LIPPS) was investigated on polished stainless steel surfaces under irradiation with fs-laser pulses characterised by a pulse duration $\tau = 300$ fs, a laser wavelength $\lambda = 1025$ nm, a repetition frequency $f_{rep} = 250$ Hz and a laser fluence $F = 1$ J/cm^2. For this purpose line scans with a scanning velocity $v = 0.5$ mm/s were performed in air environment at normal incidence utilising a well-defined temporal control of the electrical field vector. The generated surface structures were characterised by optical microscopy, by scanning electron microscopy and by atomic force microscopy in combination with Fourier transformation. The results reveal the formation of a homogenous and highly periodic surface pattern of ripples with a period $\Lambda_{exp} \approx 925$ nm aligned perpendicular to the incident electric field vector for static linear polarisation states. Utilising a motor-driven rotation device it was demonstrated that a continuously rotating electric field vector allows to transfer the originally well-ordered periodic ripples into tailored disordered surface structures that could be of particular interest for e.g. absorbing surfaces, plasmonic enhanced optoelectronic devices and biomedical applications.

1. Introduction

In the year 1960 Birnbaum was the first to observe laser induced periodic surface structures (LIPSS) by utilising pulsed ruby lasers to irradiate semiconductors [1]. Since then extensive research was carried out to investigate the formation mechanism of these structures. In the meanwhile LIPPS have been identified as a universal phenomenon that can be observed on all classes of materials when irradiated near their ablation threshold. However, the origin and growth of these structures is still controversially discussed and a complete understanding is still missing. Upon irradiation of strong absorbing materials such as metals and semiconductors by fs-laser radiation usually low spatial frequency LIPSS are observed with a period Λ close to the initial laser wavelength λ [2–8]. Interference effects of the incident laser radiation with surface electromagnetic waves are mainly used to explain this phenomenon of ripple formation [9]. In this context, the influence of surface roughness and isolated defects as well as the excitation of surface plasmon polaritons (SPP) has to be considered [10–12].

A very important parameter during the LIPSS formation is related to the polarisation state. It is generally accepted that the irradiation with linear polarised laser radiation causes structures aligned perpendicular to the incident electric field (*E*-field) vector [2–9,13]. On the contrary, the utilisation of circularly polarised radiation at normal incidence leads to the formation of random structures consisting of nanodots and ripples with many intersections and bifurcations [14–16]. The well-defined control of the direction of the *E*-field vector during laser processing introduces a new dimension of surface patterning providing the tailoring of e.g. the optical, mechanical and chemical surface properties. The diversity of adjustable structures and the broad spectrum of available materials could therefore facilitate the realisation of various concepts including broadband, omnidirectional and polarisation-insensitive surface absorption properties [17,18], plasmonics for improved photovoltaic devices [19], the tailoring of surface wettability properties [20], surface topographies for biomedical applications [21] and nanostructure formation for surface colorising [22–24]. Despite these versatile and promising applications up to now only a few studies have discussed the utilisation of transient polarisation states during LIPPS-based surface structuring. On the basis of spatial light modulators and additional waveplates Jin et al. [25] demonstrated by means of a dynamical switching between four specific polarisation states (linear horizontal and vertical, radial and azimuthal) that a highly controlled real-time nanostructuring of polished stainless steel samples is possible. Nevertheless, the switching occurs discrete between the

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mentioned polarisation states whereas the corresponding areas show highly periodic laser-induced structures that are aligned perpendicular to the *E*-field vector.

Therefore, the focus of the present study lies on the utilisation of laser radiation with a continuously rotating *E*-field vector. Based on the well-defined adjustment of the processing parameters (e.g. rotation frequency of the *E*-field vector, scanning velocity v and pulse repetition frequency f_{rep}) the presented method provides the possibility to transfer an originally well-ordered ripple formation mechanism into tailored disordered surface structures. The generation of fs-laser induced periodic disordered structures was investigated on polished stainless steel surfaces and characterised using optical and scanning electron microscopy (SEM) as well as Fourier transformation of the SEM micrographs.

2. Materials and methods

2.1. Experimental setup

Fig. 1a schematically illustrates the principle of the experimental setup. A diode pumped Yb:KYW thin disc fs-laser system (JenLas D2.fs, Jenoptik, Germany) was used to generate linearly polarised laser pulses with a duration $\tau = 300$ fs, a central wavelength $\lambda = 1025$ nm and a pulse energy $E_{imp} = 5$ μJ at a repetition frequency $f_{rep} = 250$ Hz. The pulsed laser beam was focussed onto the sample surface using an anti-reflective coated BK7 lens (focal length $f_L = 50$ mm). The focal spot diameter was measured to $2w_f = 25$ μm resulting in a peak fluence $F = 1$ J/cm^2. The time-varying *E*-field vector was generated as follows: The originally linear polarisation of the output beam was first transformed into circular polarisation by means of a zero-order quarter-wave plate (WPQ05M-1030, Thorlabs, USA). From this radiation a linearly polarised beam with a continuously rotating *E*-field vector was reproduced utilising a nanoparticle linear film polariser (LPVIS050, Thorlabs, USA) that was mounted to a motor-driven rotation device. Circular polarised radiation was generated by removing the polariser.

2.2. Laser processing

Stainless steel 304 samples (1.4301; X5CrNi18-10) of (1 × 1) cm^2 were mechanically polished using a polyurethane polishing pad tool and 0.02 μm non-crystallising colloidal silica suspension as polishing agent. Subsequently, the samples were ultrasonically cleaned in ethanol. The resulting average roughness R_a of the polished surfaces was measured utilising tactile profilometry (Form Talysurf 2, Taylor Hobson, England) to $R_a = 3.4$ nm.

LIPSS were generated by irradiating the sample surface at normal incidence and at ambient air. In a first set of experiments LIPSS formation was investigated using various static linear polarisation states and circular polarisation. For this purpose the sample surface was scanned unidirectional with a velocity $v = 0.5$ mm/s by means of a computer controlled motorised *x*–*y*–*z* translation stage. Taking into account $f_{rep} = 250$ Hz and $2w_f = 25$ μm the laser beam travels a distance $\Delta x = 2$ μm between two successive laser pulses resulting in an overlap percentage of 92%. Consequently, a focal spot diameter is effectively hit by 15 laser pulses. The direction of the linear polarisation was adjusted parallel ($\alpha = 0^\circ$), perpendicular ($\alpha = 90^\circ$) and under $\alpha = 30^\circ$ and $\alpha = -45^\circ$ relative to v by a manual rotation of the polariser (Fig. 1b). In the second part of the experiments the influence of the time-varying *E*-field vector was investigated. For this purpose the motor-driven rotation device was used to rotate the polariser by a well-defined angle $\Delta\alpha$ during the intervening period between two successive pulses. $\Delta\alpha$ was varied between 2° und 28.8°, while all other parameters (particularly v) remained constant. The results utilising the time-varying *E*-field vector were compared to the borderline cases of the static linear and circular polarisation states.

2.3. Characterisation

Laser processed sample surfaces were cleaned ultrasonically in ethanol and subsequently characterised by scanning electron microscopy (SEM). For this purpose the metallic samples were examined in the SEM (S440i, Leica, Germany) at an accelerating voltage of 15 kV and at a working distance of 5 mm using the secondary electron detector. From the SEM micrographs Fourier transforms were calculated to highlight the periodicity and the direction of LIPSS in dependence on the laser beam polarisation. The topography of the surface pattern was investigated using atomic force microscopy (Dimension 3100, Digital Instruments, USA). Moreover, the samples were characterised by optical microscopy (VHX-100K, Keyence, Japan) utilising side-illumination at grazing incidence. The microscope was equipped with a wide-range zoom lens (VH-Z100) providing a magnification range between 100× and 1000×. An external microscope illumination unit (Jenalux 150, 4H-Jena-engineering, Germany) containing a fibre-coupled halogen reflector lamp was used as radiation source.

3. Results and discussion

3.1. Static linear and circular polarisation

Fig. 2a shows the SEM micrograph of LIPSS generated at the surface of a polished stainless steel sample. The line was irradiated with $v = 0.5$ mm/s, $f_{rep} = 250$ Hz and $F = 1$ J/cm^2 using linear polarisation parallel to v ($\alpha = 0^\circ$). The appearing periodic ripple pattern is aligned perpendicular to the *E*-field vector and characterised by its pronounced periodicity. The ripples extend continuously over the whole scan width. However, the transition in morphology is slightly stronger in the centre of the irradiated lines due to the higher energy of the laser pulses in the centre of the Gaussian beam profile. The AFM micrograph in Fig. 2b reveals a period of the appearing ripple pattern close to the laser wavelength and a height of the ripples of about 200 nm.

A versatile tool for the analysis of order and disorder in surface structures is given by the 2D-Fourier transformation (2D-FFT). This method allows to highlight the periodicity of the surface pattern and to obtain the period Λ_{exp} as well as the direction of the ripple pattern. Fig. 2c shows the Fourier transform calculated from the SEM micrograph (Fig. 2a) that was obtained from the line scan with linear polarisation and $\alpha = 0^\circ$. The resulting pattern of the occurring wavenumbers $k = 2\pi/\Lambda$ illustrates a well-defined alignment of the appearing peaks along the *x*-axis, i.e. in the direction parallel to v. This proves the periodicity of the LIPSS pattern and indicates that the ripples are aligned perpendicular to the *E*-field vector. The bright pixel located in the centre ($k_x = k_y = 0$) of Fig. 2c is related to the "DC term" corresponding to zero frequency, that represents the average brightness across the entire SEM micrograph. The main peaks on either side of the DC term encode the spatial frequency, i.e. the period of the periodic pattern. These peaks are situated at a slightly higher wavenumber $k_x = 6.79 \times 10^6$ m^{-1} than that associated to the laser wavenumber $k_\lambda = 2\pi/\lambda = 6.13 \times 10^6$ m^{-1} for $\lambda = 1025$ nm. Accordingly, the resulting period of the ripple pattern was calculated to $\Lambda_{exp} = 925$ nm.

For metals, the formation of LIPSS is related to surface plasmonic waves excited by the incident laser radiation [5,8,26,27]. According to this theory, the LIPSS-period Λ_{theor} at normal incidence of the laser beam is given by:

$$\Lambda_{theor} = \lambda \cdot Re^{-1}[\eta] \quad (1)$$