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Wavelength dependence of picosecond laser-induced periodic surface structures on copper

Stella Maragkaki[a,*], Thibault J.-Y. Derrien[b], Yoann Levy[b], Nadezhda M. Bulgakova[b,c], Andreas Ostendorf[a], Evgeny L. Gurevich[a]

[a] Chair of Applied Laser Technology, Ruhr-Universität Bochum, Universitätsstraße 150, 44801 Bochum, Germany
[b] HiLASE Centre, Institute of Physics AS CR, Za Radnicí, 828, 25241 Dolní Břežany, Czech Republic
[c] S.S. Kutateladze Institute of Thermophysics SB RAS, 1 Lavrentyev Ave., 630090 Novosibirsk, Russia

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ABSTRACT

The physical mechanisms of the laser-induced periodic surface structures (LIPSS) formation are studied in this paper for single-pulse irradiation regimes. The change in the LIPSS period with wavelength of incident laser radiation is investigated experimentally, using a picosecond laser system, which provides 7-ps pulses in near-IR, visible, and UV spectral ranges. The experimental results are compared with predictions made under the assumption that the surface-scattered waves are involved in the LIPSS formation. Considerable disagreement suggests that hydrodynamic mechanisms can be responsible for the observed pattern periodicity.

1. Introduction

Laser-induced periodic surface structures (LIPSS) or ripples were first reported by Birnbaum in 1965 [1] who attributed them to diffraction of the incident beam in the optical system. Nowadays two other theories are discussed in the context of the background mechanisms of the LIPSS formation (1) interference of the incident light with the surface scattered wave [2,3] and (2) self-organization processes on the surface [4–9].

In general, surface roughness allows for the coupling of the incident laser wave with surfaces that is described in the frames of the surface-scattered wave model [2]. For metallic/metalized surfaces, it is believed that surface scattered waves have the form of surface plasmon polaritons (SPP) [10] which turn to more localized plasmons with growing surface roughness [11]. The influence of plasmonics is supported by the fact that, in many cases, the period of the observed LSFL (low-spatial frequency LIPSS) can be predicted in the frames of this model [12], and that the orientation of the ripples follows the polarization of the incident light. Recent experiments on double-pulse generation of LIPSS on silicon surfaces [13], as well as the LIPSS period reduction upon laser irradiation of silicon in water [14], are well explained in the frames of the plasmonic theory.

However, LIPSS generated by white light with a coherence length of ~1.4 μm cast some doubt on the purely interference hypothesis [7].[1] Raman measurements reveal that the LIPSS formation takes place in the molten phase [6], which allows for hydrodynamic effects in the pattern formation. For multi-pulse irradiation regimes, it was concluded that the plasmonic stage, which governs the LIPSS orientation, does not necessarily determine the periodicity of the final pattern due to contribution of the thermocapillary effects [15]. The hydrodynamic processes can be even more important for the LIPSS produced by single laser pulses at relatively high fluences assuming relatively deep melting and strong ablation of material [16,17]. Additionally, the hydrodynamic theory is supported by the topology of the pattern, which can be transient between the LIPSS and cell-like structures [17], that is typical for the hydrodynamic systems.

The accumulated evidences suggest that, in many situations, the plasmonic mechanism cannot completely explain the origin of LIPSS without assisting of other physical processes, especially for single laser pulses when the LIPSS emerge without progres-

* Corresponding author.
E-mail address: maragkaki@lat.rub.de (S. Maragkaki).

[1] Calculation of the coherence length $\ell = \lambda_0^2/\Delta\lambda$ has been made based on the data of Fig. 3 in Ref. [7] with the central wavelength of the white light spectrum $\lambda_0 \approx 650$ nm and the line width $\Delta\lambda \approx 300$ nm.

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sive, pulse-by-pulse conversion of the surface roughness spectrum into periodic grating-like structure via gentle ablation [18,19]. The main experimental argument supporting the plasmonic origin of LIPSS is that the ratio between the period of the observed ripple pattern Λ and the wavelength of the incident light λ fits the predictions of the plasmonic theory [20]. However, to the best of our knowledge, the experimental evidences of this linear dependency are summarized for several wavelengths based on works of several independent groups, which generate the LIPSS patterns on different samples at different processing conditions such as laser fluence, repetition rate, focusing and pulse number.

In this paper, we report on systematic studies on the period of LIPSS generated by *single* laser pulses of a picosecond laser system at three different wavelengths, of 1064, 532, and 355 nm on the same copper sample. The experiments are described in Section 2. In Section 3, it is shown experimentally that, for the studied conditions, the observed LIPSS periodicity is considerably overestimated by the plasmonic theory. Possible mechanisms of this disagreement are discussed in Section 4.

2. Experimental setup

The LIPSS on the surface of a polished copper sample were generated at three different wavelengths with single pulses of a diode-pumped mode-locked picosecond laser (Lumera, Hyper Rapid 25, pulse duration $\tau_p \approx 7$ ps, repetition rate set at 200 kHz). The laser system was combined with a galvanometric scanner (SCANlab) equipped with an f-theta lens. The single pulse irradiation was achieved by high scanning speed of the galvoscanner controlled by a computer. The sample surface was positioned at the focal plane of the lens. The laser pulse energy was adjusted by $\lambda/2$-plate and a polarizing beam splitter. The fundamental harmonics 1064 nm, the frequency-doubled 532 nm and the frequency-tripled 355 nm wavelengths were used for the formation of periodic surface structures on Cu. All optical systems were identical, but consisted of elements designed for the corresponding spectral range. The one and the same scanner was used for the visible and the infrared light. A representative scheme of the laser setup used is shown in Fig. 1.

The structures were analyzed by means of optical and scanning electron microscopy. As one can see from the electron microscope images (Figs. 2–4), the LIPSS, which appear after single-pulse laser ablation are not ideally periodic and contain cell-like defects. These structures are less regular compared to multipulse-generated LIPSS. As a result, determination of the pattern period by Fourier transform of the whole image is ineffective and the LIPSS periods were determined through the pattern profile measurements. The number of peaks along the lines perpendicular to the ripples were measured in several defectless spots within the LIPSS-covered area for each laser wavelength. The peak fluence was calculated as $F = 2E_p/\pi\omega_0^2$, where ω_0 is the beam radius at the $1/e^2$ of the intensity and E_p the laser pulse energy. We assume here that $M^2 \approx 1$, i.e., that the pulses are nearly Gaussian.

3. Experimental results

In this work, we investigate the wavelength dependence of the periodicity of LIPSS produced by single picosecond laser pulses, making an attempt to correlate it with the plasmonic theory and gain a better insight into the underlying physical mechanisms. The plasmonic concept of LIPSS formation was never systematically tested before for the same metal sample irradiated under the same conditions. Figs. 2–4 show the electron microscope images of LIPSS produced by single picosecond laser pulses at different wavelengths. In each Figure, laser fluence corresponds approximately to the middle of the range, in which the LIPSS can be observed at particular wavelengths.

Table 1
Summary of the experimental and theoretical results. λ is the incident light wavelength. $\Lambda_t^{(1)}$ is the LIPSS period calculated using the simple SPP model (solid line in Fig. 5). $\Lambda_t^{(2)}$ is the LIPSS period calculated with accounting for the surface roughness (squares in Fig. 5). Λ_{exp} is the experimentally measured LIPSS period. σ is the measurement uncertainty.

λ (nm)	$\Lambda_t^{(1)}$ (nm)	$\Lambda_t^{(2)}$ (nm)	$\Lambda_{exp} \pm \sigma$ (nm)
1064	1049	1037	580 ± 70
532	509	508	360 ± 20
355	355	330	300 ± 40

Table 1 summarizes the obtained theoretical and experimental data on the LIPSS periods. Considerable disagreement of the plasmonic theory with experiment, which increases with laser wavelength and cannot be explained by both the statistical and the systematic error, calls for revision of the LIPSS formation concept.

4. Discussion

In the frames of the plasmonic (or interference) model, the spatial period of the LIPSS on metallic surfaces Λ depends on the wavelength of the incident light λ as [20]

$$\Lambda = \frac{\lambda}{\mathrm{R\ e}(\sqrt{\epsilon/(1+\epsilon)})} \quad (1)$$

where the ε is the wavelength-dependent dielectric constant of metal. The angle of the laser beam incidence is equal to zero for all experiments reported here. The LIPSS period for copper calculated according to Eq. (1) with optical properties from [21] is shown in Fig. 5 by solid line.

The simple SPP model, expressed by Eq. (1), assumes an ideally-flat sample surface. Interestingly, the calculations based on a more advanced model (so-called Sipe model [2]), which introduces the shape and filling factors to describe randomly rough surfaces, s and f respectively, coincide with predictions of the simple SPP theory, Eq. (1), with $s = 0.4$ and $f = 0.1$ (as given in Ref. [23]). The LIPSS period predicted by this model is given in Fig. 5 by empty squares. The experimental data shown by dots with error bars demonstrate that, for the conditions of LIPSS formation studied here, both the simple and sophisticated models can predict only the overall tendency of LIPSS period evolution with laser wavelength but cannot describe the period values. For stainless steel surfaces, Gedvilas et al. [16] have also reported periodicities of LIPSS created by single picosecond pulses, which are considerably smaller than predicted in the frames of the plasmonic theory. Note that Eq. (1) yields respectively ~350 nm and ~1050 nm at 355 and 1064 nm wavelengths for optical properties of iron [21].

Two possible interpretations of the disagreement are possible:

(1) During picosecond laser pulses, the optical constants of metals can considerably change due to swift heating of the conduction electrons followed by heat exchange with the lattice and lattice melting. Although at picosecond irradiation regimes of moderate fluences the non-equilibrium between the electron and lattice subsystems is not so strong as in the case of femtosecond laser pulses (see, e.g., [24]), the simulations for the present experimental conditions based on the two-temperature model (TTM) [25] reveal extremely high heating of the lattice already by the middle of the laser pulse. Thus, at $\lambda = 355$ nm, the laser pulse with peak fluence of 0.4 J/cm^2 (Fig. 2) corresponding to the fluence in the center of irradiation spot yields electron and lattice temperatures in the excess of 5000 K. Under the irradiation conditions of Figs. 3 and 4, modeling gives lattice heating up to 30,000 K. Although simulations of such regimes, when matter experiences heating to extreme states and

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