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On the influence of surface plasmon-polariton waves on pattern formation upon laser ablation

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ABSTRACT

Here we analyze whether the laser-induced periodic surface structures (LIPSS), which appear on solid surfaces exposed to single-pulse femtosecond laser radiation, can be explained by excitation of surface plasmon-polariton waves. We demonstrate that excitation of the surface plasmons is impossible in the laser-ablation experiments, since the excitation conditions are not fulfilled. Moreover, properties and morphology of the observed periodic patterns contradict to the theory of the plasmonic nature of the LIPSS. The results are illustrated with experimental examples.

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1. Introduction

Laser processing is known to change the morphology of solid surfaces. After exposure to short and ultrashort laser pulses, self-organized patterns of different kind, e.g. ripples [1–4], localized column-like structures [4–6], and cells [6,7] appear on the surface. The patterns appear upon femtosecond [2,4–7], and nanosecond [8,9] laser ablation of metals [6–8], semiconductors [1,5], dielectrics [2–4] and polymers [9]. The surface can be exposed to single [6] or to multiple [4,7,8,10] laser pulses. Experiments are usually done at atmospheric conditions but the patterns are reported to appear also in liquid environment [11,12].

Along with variety of patterns, there is a large variety of theories describing their nature. In [7] the cell structure is explained by either interaction of laser radiation with surface defects or the spatial inhomogeneity of the laser beam, in [6] the cells and localized patterns are explained by explosive boiling of the laser-heated surface layer. In [8,13,14] capillary waves are involved to explain the periodic ripples, also referred to as laser-induced periodic surface structures (LIPSS).

The formation of the periodic ripples is in most cases assigned to interaction of the incident laser beam with the surface plasmon-polariton wave excited on the target surface, see e.g. [15–17]. In frames of this model, which we refer to as *plasmonic model*, the laser light modulates the electron density on the surface. This wave, which is also known as surface plasmon or plasmon-polariton wave, interferences with the incident beam; if the interference pattern is stationary, a stronger ablation takes place at the antinodes and the periodic pattern appear upon the ablation. There are two methods for the excitation of surface plasmons: (1) one can adjust the velocity of the incident light to the velocity of propagating plasmon-polariton waves by choosing appropriate incident angle and dielectric constants of the materials. This is realized in Otto and Kretschmann configurations [18–21]; (2) the plasmons can be excited on a periodically patterned surface [19], in particular due to surface roughness (corrugation), which spectrum contains frequencies, at which the coupling happens [15,22,23].

In this paper we are going to prove that the LIPSS, which are formed on the surface by means of single-pulse laser ablation, cannot be explained by excitation of surface plasmonic waves. First we demonstrate that the behavior of the experimentally observed periodic patterns differs from the predictions based on the plasmonic theory. Then we demonstrate that the conditions of the excitation of the plasmonic wave are not fulfilled in the majority of experiments, in which the periodic patterns are observed. Finally we demonstrate that some of the observed surface features are more common for hydrodynamic instabilities than for interference patterns. The analysis is made for single pulse ablation in accordance with our experimental results.

An example of the experimentally observed periodic surface structure on metallic surface is shown in the atomic force microscope (AFM) image in Fig. 1. The gold surface was exposed to a single Ti:sapphire laser pulse (*Hurricane, Spectra Physics*, the wave length $\lambda = 800$ nm, pulse duration $\tau \approx 10^{-13}$ s). One can see from the image that the ripples observed in experiments should not necessarily be parallel. For example, the self-organized periodic structure on silicon surface exposed to a single Ti:sapphire laser pulse of a fluence close to the ablation threshold has a concentric

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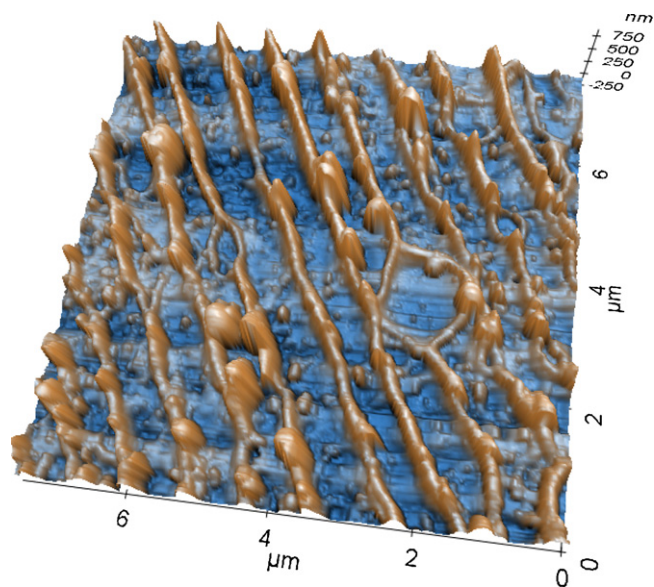   *E-mail addresses:* gurevich@lat.rub.de, gurevich@uni-muenster.de

Fig. 1. 3D AFM image of LIPSS on a gold surface. Area of 7.5 μm × 7.5 μm corresponds to the peripheral part of the crater left on the gold surface exposed to a single shot of a 800 nm Ti:sapphire laser. Laser fluence $F \approx 0.4$ J/cm^2. The height of the ripples is approximately 250–300 nm, the average period $\Lambda \approx 0.76 \pm 0.05$ μm.
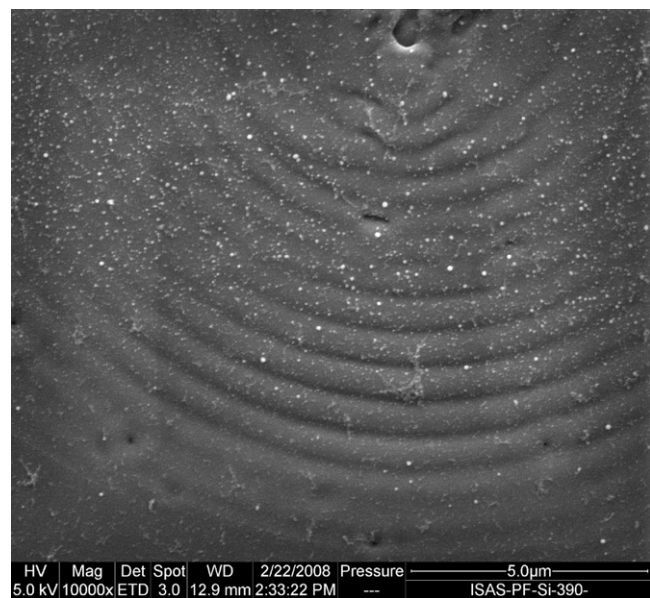

Fig. 2. SEM image of LIPSS on a silicon surface exposed to a single shot of a 800 nm Ti:sapphire laser. The laser fluence $F \approx 1.5$ J/cm^2, the average period $\Lambda \approx 0.69 \pm 0.01$ μm.

structure, see Fig. 2. The period of the surface pattern estimated from the scanning electron microscope (SEM) image is approximately $\Lambda \approx 0.69 \pm 0.01$ μm.

2. Properties of the pattern

Here we analyze experimental results reported by different working groups and demonstrate that they are inconsistent with the plasmonic theory. If we suppose that the surface plasmon-polariton wave is excited on the surface, we will have to face the following contradictions between the experimental results and the properties of the surface plasmon-polariton waves:
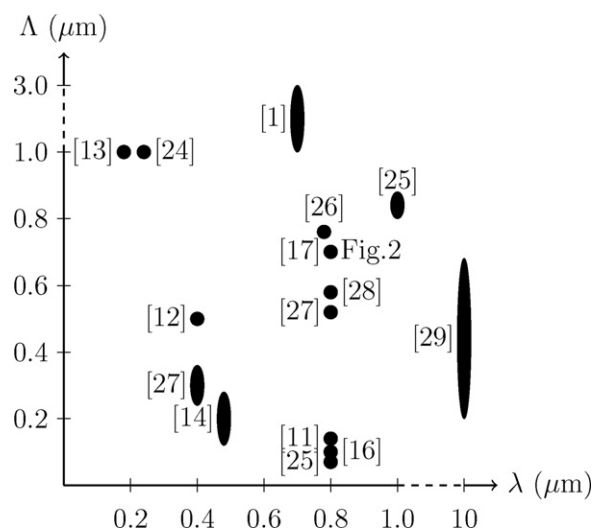

Fig. 3. Period of LIPSS in silicon reported by different groups and estimated from Fig. 2.

(I) The wavelength of the excited plasmons λ_p depends on the method applied for the plasmon excitation. However for both methods (either for Otto-Kretschmann configurations [18] or for coupling by means of surface corrugation [22,23]) the wavelength of plasmons is proportional to the wavelength of the incident light multiplied by a correction factor depending on the incident angle θ measured in relation to the normal to the surface and on the dielectric constant of the material. Consequently, the wavelength Λ of the interference pattern should also be proportional to the wavelength of the incident light and the period on the observed LIPSS should be the same.

However results reported by different groups for silicon ablated by various lasers demonstrate a different dependence, see Fig. 3. The dependences on the number of pulses, pulse duration and intensity are not shown in the image. From the plot one can see that $\Lambda \approx 1000$ nm was reported in [13] for $\lambda = 193$ nm, 14 ns excimer laser. For a slightly larger laser wavelength $\lambda = 248$ nm authors in [24] reported $\Lambda \sim 2$–5 μm for a SiO$_2$–Si-structure. The fit of the results gives for pure silicon $\Lambda \sim 1$ μm. However, as reported in [14], at $\lambda = 535$ nm the period of the surface structures is in the range of $\Lambda \sim 100$–300 nm, which is approximately 3–10 times smaller comparing to the wavelength of the pattern at $\lambda = 248$ nm. Variable period of LIPSS in the range from 1 μm and 3 μm depending on the focusing conditions was reported by Birnbaum [1] for ruby laser. For $\lambda \approx 800$ nm, the results reported for different pulse numbers and laser intensities are between $\Lambda = 70$ nm and $\Lambda = 750$ nm [16,25,26,11,17,27,28]. Remarkably, the period of the LIPSS does not increase when a CO$_2$ laser with $\lambda = 10.6$ μm is applied: $\Lambda = 200$–700 nm was reported in [29]. Thus, the wavelengths of the LIPSS for silicon reported by different independent working groups demonstrate no correlation with the wavelength of the incident laser light. The same result is also known for wide bandgap insulators like BaF$_2$ and CaF$_2$ [2].

Comparison of our experimental results (see Figs. 1 and 2) with the theoretically predicted period of the plasmon-based ripples Λ^t [22,23] does not give any clear answer on the question whether the periodic structure appears due to surface plasmons or due to some hydrodynamic instability. In our experimental conditions the incident angle $\theta = 0$, and the equations for Λ^t reported in [22,23] can be simplified. The real n and the complex k parts of the refractive index at $\lambda = 800$ nm, equations for the plasmon-based LIPSS period Λ^t adopted from [22], the results of the Λ^t calculations and our experimentally measured period Λ are listed in Table 1. One can