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Laser Induced Periodic Surface Structures induced by surface plasmons coupled via roughness

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ABSTRACT

In this paper the formation mechanisms of the femtosecond laser-induced periodic surface structures (LIPSS) are discussed. One of the most frequently used theories explains the structures by interference between the incident laser beam and surface plasmon-polariton waves. The latter is most commonly attributed to the coupling of the incident laser light to the surface roughness. We demonstrate that this excitation of surface plasmons contradicts the results of laser-ablation experiments. As an alternative approach to the excitation of LIPSS we analyse development of hydrodynamic instabilities in the melt layer.

1. Introduction

Laser-induced periodic surface structures (LIPSS) appear on dielectric, semiconductor, polymer and metal surfaces exposed to single or multiple short and ultrashort laser pulses, see, e.g., Refs. [1–7]. This pattern can be considered as one of the examples of self-organization phenomena on nano- and micrometer scale. The nanoscale pattern formation is observed in different physical, chemical and biological systems [8–11]. One of the most common structures is the periodic self-organised stripe pattern, which can be found, e.g., on the fracture surface of brittle glasses [12] or silicon wafers [13]. The periodic stripes are also observed by welding of metallic alloys [14]. During the welding the surface layers of the processed metals are melted. After the solidification periodic stripes were observed on the surface. Periodic ripples are also found by ablation of solids induced by water jet cutting. In this case the periodic surface structures can be explained in frames of the Kuramoto–Sivashinsky model [15]. In this paper we analyse ablation of solid surfaces by ultrashort laser pulses.

After a metal surface is exposed to an ultrashort laser pulse, the following chain of processes takes place [16]: the laser light is absorbed by electrons, which temperature increases during the laser pulse irradiation, while the lattice remains at the initial temperature. The system is driven out of thermal equilibrium and consists now of two subsystems at different temperatures: electron at the temperature of the order of one electron volt and the lattice at the room temperature. The thermal equilibrium between the lattice and electrons is established on the picosecond time scale. If the energy of the laser pulse is sufficient, the surface melts and remains in the melted state for up to a nanosecond. The depth of the melt is of the order of one or several hundreds of nanometers. After the resolidification, the self-organized patterns are frozen into the surface and can be observed, e.g., by means of scanning electron microscopy, see image in Fig. 1.

Although the basic physical processes at the laser ablation are well understood and moreover, the effect of the LIPSS formation is interesting as well for fundamental physics as for practical applications, the mechanisms of the pattern formation are still not completely clear. There are two theoretical approaches, which try to explain the laser-induced periodic structures: (1) theories based on interference, i.e., a purely optical approach; (2) theories involving hydrodynamic instabilities, which result in self-organisation effects. Patterns explained in frames of the optical theories are referred to as *coherent structures*, whereas patterns explained by hydrodynamic-like theories are referred to as *non-coherent structures* [17]. In this paper we analyze applicability of both these approaches for LIPSS induced by femtosecond laser pulses.

2. Interference with surface plasmons

As mentioned above, the interference-based theory (*coherent structures*) describes the LIPSS formation as an interference between the incident laser beam and a surface electromagnetic

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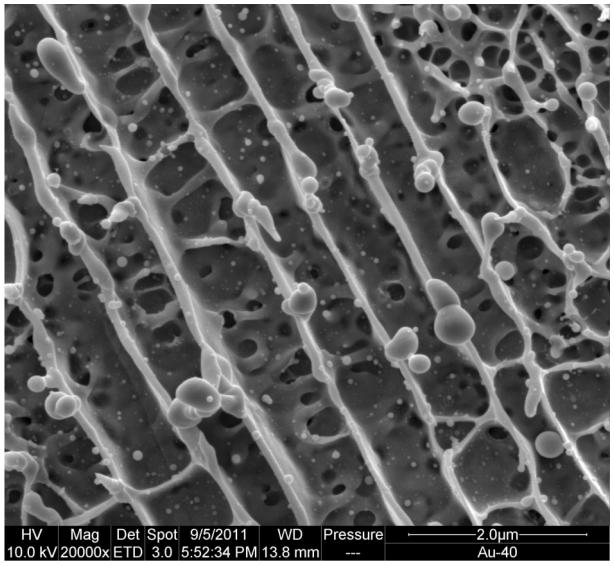

Fig. 1. LIPSS on the gold surface exposed to a single Ti:sapphire laser pulse, the wave length $\lambda \approx 800$ nm, pulse duration $\tau_p \approx 10^{-13}$ s, laser fluence $F \approx 3.3$ J/cm^2. The average LIPSS period is $\Lambda \approx 0.76$ μm.

wave excited on the surface during the laser ablation [6,18,19]. The nature of this wave is not clear, but obviously its excitation time scale must be well below the pulse duration, hence, only surface plasmons can be taken into account. The interference between the laser light and the surface plasmons gives reasons for the periodicity of the induced structures. The pattern period can be estimated and the orientation of the stripes can be explained in some experiments. However, there are contradictions between the experimental observations and the predictions made in frames of this plasmonic theory reported, e.g., in [2,7]; the validity of the plasmon excitation conditions by laser ablation is also debatable [7].

Indeed, the plasmon excitation via light is described by the dispersion curves of the both waves (plasmons and photons) and the excitation conditions are defined by their intersection, see Fig. 2.

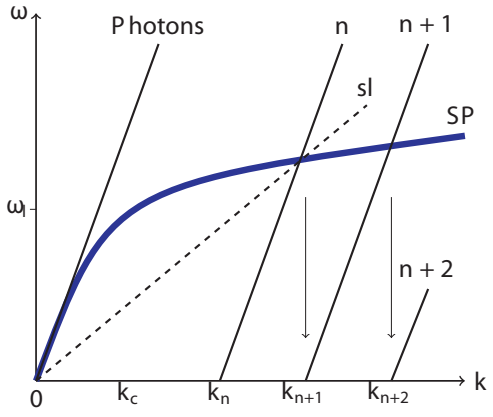

Fig. 2. Schematic representation of the dispersion curves of surface plasmon-polariton waves (bold blue solid line labeled as *SP*) and of free photons (thin black solid line). Dashed line *sl* stays for slow photons; solid line *n* - periodically structured surface with the wave vector k_n (or $k_{n+1}, k_{n+2}, \ldots$, respectively). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

The dispersion curves of photons and plasmons are described by the equations

$$k = \frac{\omega}{c} \quad \text{and} \quad k = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1 \varepsilon_2}{(\varepsilon_1 + \varepsilon_2)}} \tag{1}$$

respectively. Here, the ω is the frequency of light, k – the wave vector, $\varepsilon_1 = 1$ – dielectric constant of the surrounding medium (air), $\varepsilon_2 = 1 - \omega_p^2/\omega^2$ – dielectric constant of the metal with the plasma frequency ω_p. Typical values for the plasma frequencies for gold and copper are comparable $\omega_p \approx 1.4 \times 10^{16}$ s^{-1} [20]. The dispersion curves are plotted in Fig. 2. It is useful to note that that of surface plasmons (denoted as *SP*) starts saturating at approximately $k_c = \omega_p/c \approx 5 \times 10^7$ m^{-1}.

From Fig. 2 one can see that there is only one intersection point between the dispersion curve of surface plasmons and that of free photons (denoted as *Photons*) and this intersection corresponds to the zero frequency. Due to this reason the plasmonic theory has difficulties to explain the excitation mechanism of the surface plasmons upon laser ablation. Thus, direct optical excitation of surface plasmons is impossible [20,21]. There are two methods how to avoid this limitation [22]: (1) decrease in the slope of the photon dispersion line, i.e., to decrease the phase velocity of light (see line *sl* in Fig. 2); (2) shift of the dispersion curve. The second important issue is the energy of the excited plasmonic wave. The amplitude of the plasmonic electric field must be comparable to that of the incident light, since for any sort of interference one needs two nearly coherent waves with comparable amplitudes. Hence at least half of the energy of the incident laser light should excite the oscillations of the electrons on the surface. The practical realization and difficulties of these plasmon excitation scenarios are discussed in the two following subsections.

2.1. Excitation via slow light

The phase velocity of photons can be slowed down to fit the velocity of the propagating plasmon-polariton wave by choosing appropriate incident angle and dielectric constants of materials. This is realized in the Otto and in the Kretschmann configurations [23,24] by illuminating the surface through a dielectric prism and choosing the incidence angle so high, that the total internal reflection of the incident light takes place on the dielectric interface. This method is used in biology and medicine and allows coupling of up to nearly whole laser energy to plasmons: Depending on the laser wavelength and incidence angle, the reflection can be varied from approximately 100% to approximately 1% [25,26]. The method requires illumination of the metal surface at a fixed incidence angle $\alpha \neq 0^\circ$ through a dielectric material with high refractive index [20–24] to achieve the necessary condition for the total internal reflection.

In common laser-ablation experiments, in which the formation of LIPSS is observed, the surface is exposed at the incidence angle of $\alpha = 0^\circ$ through the air. In some experiments silicon surfaces are exposed through a layer of liquid and a thin layer of silicon oxide [27], but the conditions for the total internal reflection remain unsatisfied there. In the theoretical studies one can also consider the electron plasma excited under the oxide layer in silicon and analyze a layered system, for which some of the excitations condition are easier to satisfy [27]. The most fundamental problem of the plasmon excitation in laser ablation is the incident angle of the laser light, which is typically $\alpha = 0^\circ$. The momentum of the incident wave is perpendicular to the sample surface, hence the excitation of a wave in the surface plane (i.e., with the momentum perpendicular to the momentum of the exciting wave) is difficult without involving other physical mechanisms.