Influence of laser-induced plasma parameters on the formation of laser-induced surface-periodic structures

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Abstract. The paper presents the results of processing a silicon surface with subpicosecond laser pulses under various exposure conditions. The variable conditions were: pressure in the working chamber, the speed of scanning the silicon surface with a laser beam and the laser pulse energy. Special attention was paid to the interaction of the laser-induced plasma plume with the surface of the processed material, with laser radiation and with the medium. The influence of the medium on the parameters of the laser-induced plasma is estimated. The result of this interaction is a change in the spatial characteristics of the laser-induced plasma channel, the period and type of the formed periodic surface structures, what is related to the change in the laser radiation passing through the medium at different pressures.

1. Introduction
The impact of laser pulses on the surface of materials makes it possible to obtain laser-induced periodic surface structures (LIPSS). The nature of LIPSS formation is significantly influenced by the mode of exposure to laser radiation, the parameters of the medium, and the state of the sample being processed. According to [1], LIPSS are divided into two different types: the so-called low spatial frequency LIPSS (LSFL) and high spatial frequency LIPSS (HSFL) [2]. It is generally accepted that LSFLs have a spatial periods close to the laser radiation wavelength and are formed due to optical interference of incident laser radiation with a surface electromagnetic wave that is generated during exposure to pulsed laser radiation. According to [3], HSFLs have a spatial periods that are much shorter than the laser radiation wavelength and are observed exclusively when irradiated with an ultrashort laser pulses. In addition to the structures described above, there are also structures resulting from the action of filaments formed in a laser beam when the radiation resistance of the medium in which the laser radiation propagates is exceeded. In this work, the process of filamentation of laser radiation is considered as a process leading to the formation of surface structures. In work [4], we carried out experiments to study the effect of the pressure on the period of the LIPSS structures obtained on the titanium surface during laser processing in n-hexane medium. The key role of the influence of the pressure parameter in the working chamber on the periodicity of the resulting structures has been established. The pressure parameter of the medium has a significant effect on the laser-induced plasma, the laser erosion plume, and on the thermal balance of the system (laser radiation - medium - processed surface). In [5, 6], the influence of the medium on the spatial characteristics of the laser-induced plasma channel (LIPC), its position relative to the focal plane of the focusing system, and the temperature and concentration of free electrons are considered.
In the process of LIPSS formation over the processing area, the formation of a laser-induced plasma is characteristic, which carries out an additional effect on the parameters of the laser beam, on the thermal balance of the system (working medium, laser beam exposure area). Excessive heat exposure leads to the formation of a liquid phase of the material. The accumulation of heat consists of the residual heating of the surface, the thermal effect of the laser-induced plasma, and the thermal effect of the laser erosion plume. Problems of the formation of a liquid phase as a result of overheating of the processed material lead to a violation of the period of the resulting structures, a violation of surface properties, which is critical when obtaining LIPSS on materials containing thin-film coatings, up to the complete destruction of the coating or mixing of the coating with the surface of the base material during melting.

2. Experimental
In this work, we studied the influence of conditions in the working chamber on the nature of the formation of LIPSS structures. In the course of the work, periodic surface structures on the surface of the silicon wafer, the spectrum of the laser-induced plasma, and the glow intensity of the laser erosion plume were recorded. The experimental setup used is designed as follows: laser radiation from a source (Yb:KGW femtosecond laser operating at wavelength 1030 nm) was directed into an optical head with galvanoscanners and an F-theta objective. The mode composition of the laser beam corresponds to the TEM 00 mode, the polarization is linear. The surface of the silicon wafer was processed at a speed of 1-40 mm/s, the pulse energy was set to 20 μJ, the pulse duration was 280 fs, the pulse repetition rate was 10 kHz, and the spot diameter in the affected area was 60 μm. Figure 1 shows the schematic diagram of experimental setup.

![Figure 1. Schematic diagram of experimental setup.](image)

The samples were single-crystal silicon wafers with a thickness of 1 mm. The medium in the working chamber is gaseous argon of high purity, grade 5.5. The pulse energy value was determined in such a way that the nature of the impact was as close as possible to the material ablation threshold. Such a parameter as the scanning speed determined the frequency of overlapping of the zones of exposure to laser radiation on the surface of the sample. The sample was located 1 mm below the caustic of the laser beam, which made it possible to observe the optical breakdown region above the surface of the sample being processed.

3. Results and discussion
As a result of processing the surface of a silicon wafer with laser radiation at various pressures, samples were obtained. Figure 2 shows images of the structures obtained at a laser beam scanning speed of 5 mm/s and a pressure from 1 mbar to 20 bar, the scanning direction of the laser beam for all images is horizontal, from left to right.
Figure 2. The result of processing the silicon surface at different pressures with a laser beam scanning speed of 5 mm/s.

In the presented images, there are three different types of structures, which are indicated by multi-colored rectangles for clarity. In the area marked with a red rectangle, there are structures formed as a result of the interference mechanism, the power density is lower than laser ablation threshold. Residual thermal energy accumulated as a result of the combined action of laser pulses is dissipated due to the thermal conductivity of the material, as a result of heat exchange with the medium, the direction of polarization ripples structures is orthogonal to the polarization vector of laser radiation.

In the area marked with a yellow rectangle, one can see the presence of competing processes, the impact energy is comparable or slightly exceeds the laser ablation threshold. This zone contains both traces of fusion, material erosion, and polarization ripples structures. The direction of polarization ripples structures is orthogonal to the laser radiation polarization vector. The zone marked with a blue rectangle contains traces of thermal damage to the surface (the energy is significantly higher than the ablation energy); a developed morphology is observed. Figure 2 shows the areas of melting and ejection of the liquid phase of the material along the boundaries of extended caverns. The direction of propagation of caverns coincides with the direction of movement of the laser beam, it is most pronounced at pressures from 5 bar. At pressures of 1 mbar and 1 bar, the ordering is poorly expressed, there is developed branching, the presence of separate rounded cavities, the edges of which are covered with products of ablation and ejections of the liquid phase of the material.

The dynamics of changes in the types of surface structures depending on the scanning speed at a constant pressure (10 bar) is shown in figure 3. The structures formed in the central area of the laser beam impact are formed at a scanning speed of 5 to 20 mm/s with an energy of 20 mJ per pulse. An increase in the scanning speed to 40-60 mm/s leads to disordering of the directions of the formed system of extended caverns; the presence of randomly located local maxima and minima on the processed surface is observed. The formation of polarization ripples structures over the entire area of the laser track is typical for velocities of 80-120 mm/s. This fact indicates a significant influence of the surface morphology and the effect of heat accumulation caused by the superposition of areas of exposure to laser radiation. A surface with a developed relief absorbs laser radiation better. An increase in absorption efficiency leads to the accumulation of more heat, which does not have time to dissipate before the arrival of the next pulse.
Figure 3. Silicon surface at a pressure of 10 bar and different scanning speeds.

At low scanning speeds up to 20 mm/s, stable formation of horizontal periodic structures oriented in the direction of scanning of the laser beam is observed. This phenomenon can be explained by the fact that the filaments in the laser-induced plasma channel have a characteristic diameter of the order of the period of the resulting cavities. Thus, under these processing conditions, there is a cumulative superposition of local maximums of the radiation intensity in the cross section of the laser beam, on the edges of the formed caverns, respectively, the direction of the extended cavern corresponds to the direction of scanning of the laser beam, this structure does not depend on the polarization of the laser radiation. An increase in the velocity from 40 to 60 mm leads to the formation of a disordered system of craters in the central region of the laser beam, the diameter of which is somewhat less than the period of the resulting extended caverns; it is assumed that each crater on the surface is an imprint of the action of the filament. Extended ordered cavities are not formed due to the small exposure of laser radiation and the magnitude of the displacement of the affected area is greater than the diameter of the filament in the area of influence (the displacement area is determined by the scanning speed and the frequency of laser radiation generation, the spatial characteristics of the filaments are determined by the laser radiation energy, characteristics of the medium, and pressure). A further increase in the scanning speed leads to the formation of polarization ripples structures in the flesh up to a scanning speed of 120 mm/s. An increase in the scanning speed over 120 mm/s for these exposure conditions leads to the formation of unstable structures on the surface; surface changes are observed only in the areas of overlapping of the laser beam exposure zones.

Figure 4 shows the dependence of the period of the formed structures on the pressure in the working chamber; for pressures of 1 mbar and 1 bar, the distance between the edges of the cavities was measured.
At a pressure of 5 to 20 bar, a pronounced decrease in the period of the obtained structures and a scatter in the range of measured values are observed. It was found that the period of polarization ripples structures does not depend on pressure and, according to measurements, averages 0.7 µm, which is typical for the period of these structures and corresponds to the results described in [1, 3]. The formation of an ordered structure of cavities, the direction of which coincides with the direction of motion of the laser beam as a result of filamentation, was described in [5-7]. Optically dense media were used to stimulate filamentation; the development of filamentation in them determines the further distribution of power in the cross section of the laser beam. The formation of filaments in liquid and gaseous media with each pulse is random, rather than in crystals and glasses. In [4], the formation of extended cavities during titanium processing in an n-hexane medium is considered; the period of these structures also depends on pressure. In this work, the period of the resulting structures was dependent on pressure, the period was shorter than that observed in the current study. This behavior is probably explained by the different density of the medium in which the laser radiation propagates. When a laser beam passes through a layer of liquid n-hexane, a denser arrangement of filaments is observed due to the higher optical density of n-hexane compared to gaseous argon. The development of filamentation is also facilitated by an increase in the energy of laser radiation. The filaments in the caustic region become more extended, which leads to a change in the power distribution in the cross section of the laser beam.

In [8], data are presented on the dependence of the length of laser-induced plasma channels and the glow brightness on the argon pressure in the working chamber, which is shown in figure 5.
Figure 5. Dependence of the position and brightness of the LIPC luminescence on the argon pressure in the working chamber, the direction of LR propagation from top to bottom. F - focal plane, b1 - anterior border of LIPC, b2 - posterior border of LIPC [8].

An increase in the density of the medium due to an increase in the pressure in the chamber contributes to a significant increase in the length of the laser-induced plasma channel, which consists of many filaments localized in the caustic region. Unfortunately, the insignificant luminosity of these plasma structures and the low duration of their existence do not allow their detailed analysis due to the insufficient spatial and temporal resolution of the used image registration system. According to the data presented in figure 5, at a low pressure of up to 7-8 bar, the glow intensity and length increase significantly, which indicates a more effective interaction of laser radiation with the medium.

The lifetime of channels formed in a gaseous medium by femtosecond laser pulses ranges from a few to ten of microseconds [8]. Thermal processes in the surface layer of the material start after the absorption of laser radiation and are fed, among other things, from the thermal effect of the laser erosion plume [9-13]. The problem of the formation of a liquid phase as a result of overheating of the processed material leads to spreading of the processed edges of the parts, deterioration of material properties in the near-boundary region, a large thermal load is formed on the affected area, which is typical for modes of exposure to laser radiation with a sufficiently high pulse repetition rate [13, 14]. Based on the data obtained, it has actually been established that the dominant process leading to the formation of extended caverns, the period of which depends on the scanning direction and does not depend on the polarization of the laser radiation, is the pressure of the medium in which the treatment takes place.

The magnitude of pressure affects the propagation of laser radiation, contributing to the formation of extended plasma channels, thus the effect on the material surface of both laser radiation and temperature, caused by the presence of a laser-induced plasma channel, is observed. According to the data of [9-11], the process of laser processing with an energy exceeding the material ablation threshold is a complex process, the description of which is rather difficult, including rather difficult to identify the dominant factor or a group of factors from the entire set of observed phenomena leading to the required result. The authors of the works agree on the significant effect of the plasma erosion plume on the surface to be treated. The development of laser-induced plasma can be stimulated by laser radiation reflected from the surface, including the development of a laser-induced plasma plume is achieved due to the ingress of ablated particles of the material into it, which leads to an increase in its lifetime over the area of influence and temperature erosion of the surface. This assumption was made based on the nature of the
formation of a system of longitudinal cavities at different speeds of the laser beam scanning over the silicon surface, which is shown in figure 3.

Heating of the material surface is possible as a result of the action of a laser-induced plasma plume formed over the area of action of laser radiation during the interaction of a laser pulse with both the material surface and with the medium. Interaction of laser radiation with a laser-induced plasma plume is possible when the lifetime of the laser-induced plasma is longer than the time of the interpulse gap. The working medium in which the processing is carried out affects the processes of heat transfer, the formation of a laser-induced plasma channel, and the expansion of the laser erosion plume. When the radiation resistance of the medium is exceeded, laser radiation forms an optical breakdown and a laser-induced plasma channel. This formation is typical in the area of laser beam caustics.

During the propagation of a laser pulse of subpicosecond duration, leading to the formation of a laser-induced plasma channel, this phenomenon causes changes in the laser beam, the front part of the pulse interacts with the medium, causing its changes, which leads to a change in the nature of propagation of the rear of the pulse. This plasma structure, being an "optical element" in this system, can affect the further propagation of the beam, leading to a redistribution of power. The hereditary anisotropy of the density of this plasma formation is also characteristic of the laser erosion plume, which forms above the surface of the area of action of laser radiation and the laser-induced plasma channel.

4. Conclusion
As a result of laser processing, several types of surface structures are formed, some of them are formed as a result of the interference of the incident optical and surface waves, other structures are caused by the action of local maxima in the laser beam during filamentation of radiation in the caustic region, or by the thermal effect of localized plasma channels formed above the treated surface. It was found that the pressure in the chamber practically does not affect the periodic structures formed as a result of the interference of the incident optical and surface waves. The magnitude of the pressure has a dominant effect on the period of the structures formed when the ablation threshold is exceeded due to the local action of filaments, as well as the appearance of structures as a result of the temperature effect of plasma channels formed above the material surface as a result of the erosive action of the laser beam. These structures are formed when scanning a laser beam as a result of the superposition of the effect of the combined action of many points of energy impact, are formed into channels, the direction of which coincides with the direction of movement of the laser beam. When the scanning speed is exceeded (above 20-40 mm / s), the overlap is random, the resulting surface is a system of craters and splashes, while the caverns are not combined into extended co-directional channels.

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