OCCURRENCE OF EROSION PROCESSES IN THE NEAR-SURFACE REGION OF METALS EXPOSED TO INTENSE NANOSECOND LASER PULSES

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Using the method of laser probing, the time dependences of the transparency factor and integral luminescence of erosional jets of metals appearing upon their exposure to intense nanosecond laser pulses, as well as of the probing radiation component scattered by the jet, have been determined. Based on the results of laser probing, a conclusion was drawn on the condensation nature of the process of liquid-droplet phase formation near the surface of investigated metals in the considered conditions of laser effect on them.

Keywords: laser erosion of metals, nanosecond pulses of optical radiation, condensed phase of target material.

Introduction. Over the course of the last 50 years, the problems of laser radiation interaction with a substance have aroused the ever growing interest of researchers as is evidenced by the exponential growth of the number of publications devoted to the topic. At the present time, there exists a wide selection of laser systems making it possible to generate radiation with various power, spectral, polarization, and time characteristics. Here the direction of scientific investigations is usually specified by the needs of development of certain technologies of laser processing of materials for solving particular industrial problems. An example of such problems is laser synthesis of nanosized metal structures under conventional atmospheric conditions [1–3]. This technological problem can be solved with application of modern commercially produced laser facilities making it possible to generate laser pulses with duration ~10 ns and a high power density $(10^8-10^{10} \text{ W/cm}^2)$ in the frequency regime.

The present work is devoted to investigation of the processes proceeding in the course of the first microseconds of existence of erosional laser jets (ELJ) formed near the surface of metal targets by nanosecond laser pulses of high power density.

Experiment. To study the dynamics of erosional laser jets of metals with a high time resolution, an experimental laser setup was used the schematic diagram of which in given in Fig. 1. Functionally the setup can be divided into: a block generating pulses (2), a block for controlling the parameters of acting pulses (5–9), a block generating probing pulses (16–20, 26), a block for recording the components of probing radiation (3, 4, 10–15, 21–25), and that for synchronization (1).

The block for generating pulses and for controlling the parameters of incident radiation is represented by a LOTIS 2137 commercial-type laser facility (2) connected with a photodiode (8) and a calorimeter (9). The characteristic temporal form of the incident radiation pulse is given in Fig. 2a. The length of this pulse at half-height is 20 ns and the energy is 300 mJ, which on focusing into a spot of diameter 1 mm with the aid of a lens (6) allows one to obtain a power density of about 1 GW/cm^2 .

The block generating probing radiation consists of a ruby laser (19) based on a standard block "Nakachka (Pumping) 3000M" functioning in the mode of quasi-stationary generation, which is attained with the aid of confocal resonators (18, 20) and due to the appreciable excess of the amplification factor over losses. A fragment of the temporal form of the probing pulse is presented in Fig. 2b. The peak density of the probing radiation power attains 10^4 W/cm². The order of this quantity is limited, on the one hand, by the undesirability of disturbance of the test medium by a probing pulse and, on the other hand, by the requirement of the minimum level of intensity needed for recording the scattered component.

PhD 21-KP photodiodes (21, 24) and an SPM Micro 10020 SensL electronic photomultiplier (22) combined with a PC (25) by means of a B 422 multichannel digital oscilloscope (11), which allows one to attain resolution in time in about 10 ns, form the basis of the block for recording the probing radiation components. To level the influence of the incident radiation

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Fig. 1. Basic diagram of the experimental setup: 1) synchronization system; 2) neodymium laser; 3 and 4) interference filters ($\lambda = 694.3$ nm); 5, 7, 10, and 15) branching, planeparallel quartz plates; 6) mobile lens; 8, 21, and 24) photodiodes; 9) calorimeter; 11) ADC; 12) integrating sphere; 13) metal target; 14) erosional laser jet; 16 and 17) rotating prisms; 18–20) ruby lasers with a confocal resonator; 22) photodetector; 23) system of optical filters; 25) PC; 26) aligning helium–neon laser.

and erosional laser jet luminescence on the process of recording the probing pulse components, interference filters (3, 4, 23) ($\lambda = 694.3$ nm) are used.

To ensure coordinated operation of all the components of the facility, we employed a G200P multichannel generator of delayed pulses (1). It produces synchronous pulses with different time lags for each of the blocks.

The facility described makes it possible to investigate, with high time resolution, the dynamics of the following characteristics of erosion processes in the near-surface region of metals:

- a) the spectrum-integral luminescence of erosional laser jet (with consideration of the spectral sensitivity of the photodetector);
- b) the transparency of the erosional laser jet for probing radiation depending on the height of probing;
- c) the probing radiation component scattered by the jet [by applying an integrating sphere (12)] depending on the probing height.

As metal targets, we selected massive plates made of lead, zinc, copper, nickel, as well as of silver, gold, and platinum. Due to the vast differences in the optical and thermophysical characteristics [4], the metals of the first group (Pb, Zn, Cu, Ni) allow one to speak of the laws governing the processes of laser erosion as a whole for metals [5, 6]. Investigation of the materials of the second group (Ag, Au, Pt) is of practical interest for developing the technology of laser synthesis of nanosized metal structures under the conditions of the ordinary atmosphere [1]. The smooth surface of targets was modified by longitudinal scratches of depth 30–50 μ m in order to maximize the entrainment of material eroded on exposure to laser radiation [6, 7].

Discussion of Results. The character of destruction of metal targets on exposure to the action of 20 nanosecond laser pulses of high power density $(10^8 - 10^{10} \text{ W/cm}^2)$ differs qualitatively from both the erosion caused by millisecond and



Fig. 2. Time dependences of the intensities of laser pulses of acting (a) and probing (b) radiation.



Fig. 3. Time dependences of the intensity of acting pulse (1), integral luminescence of an erosional laser jet (2), probing pulse at h = 1 mm (3), probing pulse at h = 2 mm (4) on exposure of a lead target to 20 nanosecond pulses with power density of 10^9 (a) and 10^8 W/cm^2 (b).

longer pulses of moderate power radiation $(10^5-10^8 \text{ W/cm}^2)$ [8–11] and from the process of the destruction of metals under the action of femto- and picosecond pulses of high power density radiation [12–14]. However, one can see a considerable resemblance of the character of the progress of metal surface erosion and of the formation of a plasma jet from the metals subjected to the action of 20 nanosecond laser pulses to similar processes of erosion under the action of high-power density radiation pulses of length of about 100 ns [5–7].

Here and below, as an example we will consider the dependences obtained for lead targets. However, it should be noted that the general conclusions made are based on processing large amounts of experimental data for all the seven metals investigated.

The results of investigation of the temporal form of the transparency of the erosional laser jet for probing radiation and integral luminescence of the jet formed on exposure of a lead target to pulses of length 20 ns with power density of 10^8 and 10^9 W/cm² are presented in Fig. 3.

The study of the dynamics of integral luminescence of all of the metals discussed has shown that the moments of maximum luminescence of the erosional laser jet at the power density of acting radiation 10^8 and 10^9 W/cm² are delayed relative to the intensity peak of laser pulse by 20–60 ns. This points to the fact that the entire leading front of the acting radiation freely attains the target surface. The form of the time dependence of the luminescence of erosional laser jet of all test metals at a power density of laser pulse 10^8 W/cm² as a rule, with a small delay, repeats the loop of the acting laser pulse. On increase in the power density of laser radiation from 10^8 to 10^9 W/cm², a noticeable increase in the integral luminescence of the erosional laser jet is observed. It is accompanied by a substantial delay in the rear front of the dynamics of luminescence,

Metal	Time interval between moments of attaining distances of	Estimated value of initial velocity of
	1 and 2 mm from the target surface by erosional faser jet, hs	plasma formation, km/s
Silver	140	7.1
Gold	270	3.7
Platinum	220	4.5
Nickel	160	6.3
Copper	130	7.7
Zinc	70	14.3
Lead	120	8.3

TABLE 1. Estimate of the Initial Velocity of the Moving Forward Front of Erosional Laser Jet



Fig. 4. Velocity of the forward front of erosional laser jet vs. the power density of acting pulse: 1) 10^9 W/cm²; 2) 10^8 .

which points to the considerable increase in the maximal density of plasma formation as compared to the cases of lower intensity of the acting optical pulses. Thus, for convenience, the peaks of the time dependences of the luminescence of erosional laser jet of lead are shown in Fig. 3a and b on one level; however, the real peak intensity of the luminscence of erosional laser jet for a pulse with a power density of 10^9 W/cm² is three times higher than for the pulse with a power density of 10^8 W/cm².

Investigation of the transparency of an erosional laser jet for probing radiation demonstrated a considerable increase of this characteristic for all the metals studied as compared to the case of higher-power pulses of length 100 ns [5]. Thus, the losses of probing radiation during its passage through the erosional laser jet at a height of 1 mm from the target surface do not exceed 20% (see Fig. 3a) even at the intensity of a 20-ns acting pulse of 10^9 W/cm² (which is characterized by the most dense plasma formation), whereas in a similar case of the action of 10-ns pulses on metals [5] the losses of probing radiation in a peak attained 80%. The low levels of probing radiation losses are typical of the erosional laser jets of all the studied metals exposed to pulses of length 20 ns; their maximal value amounts to 10-15% for the easily fusible lead and zinc and 3-10% for the remaining metals.

The forms of the time dependences of the integral luminescence of an erosional laser jet and of its transparency for probing radiation at a height of 1 mm (for the intensity of acting pulses 10^8-10^9 W/cm²) have an important feature: the peak of the integral luminescence of an erosional laser jet is delayed relative to the action only by 20–60 ns (depending on the type of the metal and power density of the acting pulse, whereas the minimum of transparency is delayed already by 120–200 ns. This can point to the fact that the main luminescence of the erosional laser jet under described conditions occurs prior to the moment when the plasma front attains a height of 1 mm, whereas in the case of the action of 100 nanosecond radiation pulses the considered dependences attain extrema practically synchronously [5].



Fig. 5. Time dependences of the acting pulse (1), general probing radiation losses in erosional laser jets (2); probing radiation losses in erosional laser jets at the expense of scattering (3) and probing radiation losses in erosional laser jets at the expense of absorption (4).

Using the time lag between the minima of the transparency of an erosional laser jet at different heights over the target surface (1 and 2 mm), one can estimate the initial velocity of the plasma front propagation. The results of this investigation are presented in Table 1. They show that the characteristic values of the initial velocity of the forward front of erosional laser jet of the metals investigated amount to 4–15 km/s. Good correspondence of the data obtained to those of [5] for pulses of length 100 ns should be noted, which may indicate the absence of the effect of the forward front duration of nanosecond pulses on the initial velocity of plasma formations induced by them.

Investigations of the velocity of motion of the erosional laser jet front of lead were carried out for a wider range of the heights of probing relative to the target surface (1-5 mm), which made it possible to determine the velocity of motion of the forward front of plasma formation depending on its distance from the metal surface (see Fig. 4). The characteristic form of the curves obtained is indicative of the adiabatic cooling of the erosional laser jets formed on exposure to intense nanosecond radiation. In this case the initial value of the erosional laser jet velocity on increase in the intensity of irradiation up to 10^9 W/cm^2 increases somewhat and thereafter decreases more slowly. This again indicates the substantial increase in the laser plasma parameters on increase in the power density of nanosecond pulses up to 10^9 W/cm^2 .

In order to study the structure of the probing radiation extinction in a plasma formation at different time instants, the dependence of its scattering on time and on the distance to the target surface (1 and 2 mm) was recorded in addition to the dynamics of losses. This made it possible to divide the probing radiation losses by absorption and scattering attributable to the fundamentally different processes proceeding in plasma. It is seen in Fig. 5a that actually in the first 150 ns after the start of laser action on a lead target, practically all probing radiation losses in the erosional laser jet are determined by the absorption in plasma (due to the effect of the backward bremstrahlung absorption on charge carriers [5]). Thereafter, in the structure of the probing radiation extinction a scattered component appears that rapidly increases and attains its maximum in about 200 ns after the start of action. Subsequently this component decreases nonmonotonically. Such dynamics of the scattered component can be explained by the start of the process of condensation in the erosional laser jet [5]. Indeed, two competing processes contribute to the probing radiation scattering: the increasing scattering by the local inhomogeneities of the cooling-off plasma (density fluctuations) [15] and the adiabatic scattering of plasma, which decreases the scattering by decreasing the amount of target material in the zone of probing.

Approximately 1.5 μ s after the start of laser action, the erosional laser jet plasma "cools off" sufficiently and its scattering practically ceases. From this instant of time one observes a stable excess of the processes of absorption over scattering, which can be explained by the start of stable formation of drops at the expense of condensation processes [5]. The results of investigation of the dynamics of extinction of probing radiation and of its scattered component for the case of smaller intensity (10⁸ W/cm²) of acting pulse (Fig. 5b) show that these trends qualitatively resemble those described above, with only a small decrease being observed in the level of their peak values, which is indicative of the great transparency of plasma formation.

Based on the investigated laws governing the laser erosion of metals by nanosecond (20 ns) high-intensity (10^8 – 10^9 W/cm²) radiation pulses, a conclusion can be drawn that in approximately 20–60 ns (depending on the metal) after the start of laser action a highly luminescent plasma formation is formed in the near-surface region of the target. The laser jet propagates in the direction of the external medium, with the maximum of luminescence of the erosional laser jet for all the materials investigated occurring at a height of less than 1 mm. The characteristic velocity of propagation of the forward plasma formation at a height of 2 mm from the target surface was 4–14 km/s depending on the type of metal target. The study of the magnitude of the forward front velocity of the erosional laser jet in a wider range of heights (2–5 mm) for a lead target has revealed an exponential decrease of this parameter, which confirms the model of adiabatic expansion of the erosional laser jet in the case of intense submicrosecond laser action [16].

After the fall in the intensity of laser action, rapid cooling of the jet begins predominantly due to the scattering of plasma, as can be judged by a certain delay in the decrease in jet luminescence relative to the trailing front of the acting pulse. Here, in the case of the low intensity (10^8 W/cm^2) of the acting pulse, the form of the trailing fronts of the acting pulse and of the erosional laser jet luminescence practically coincides, which indicates the high level of transparency of plasma formations and actual absence of its interaction with incident radiation (i.e., practically the entire energy of the pulse arrives at the target surface). With increase in the laser pulse power density (up to 10^9 W/cm^2), the delay in the decrease of luminescence becomes longer and its intensity increases. This may point to the increasing interaction of the erosional laser jet with the acting radiation. As the most probable mechanism of the absorption of the trailing front energy of the acting laser pulse, we can consider the inverse bremsstrahlung effect [17].

In this case, in the structure of the probing radiation extinction a scattered component appears indicative of the formation, inside of the jet, of local inhomogeneities of plasma, which at this moment determine the transmission of probing radiation by the jet. Subsequently, $1-1.5 \mu s$ from the start of action, the processes of absorption of probing radiation start again to prevail over the scattering at a small enough level of losses in the jet, which indicates the start of disperse droplet formation through plasma cloud condensation [5]. The results of previous investigations [6] show that this process leads to formation of nanosized particles of the treated metal in the near-surface region of the target.

Conclusions. As a result of the investigation carried out, the main laws governing the formation of an erosional laser jet of metals exposed to the action of 20 nanosecond intense $(10^8-10^9 \text{ W/cm}^2)$ laser radiation pulses have been established. It is shown that the application of nanosecond pulses of intensity 10^8 W/cm^2 for laser treatment of metals is most efficient, since in this case the erosional laser jet practically does not interact with incident radiation due to the high transparency of an appearing plasma formation. On increase in the radiation power density, a considerable part of the acting pulse energy does not reach the target surface and is spent on increase in the parameters of the forming plasma (temperature and density). The progress of condensation processes in an erosional laser jet formed under the given conditions makes laser erosion attractive from the point of view of the realization of highly efficient processes of the formation of nanosized particles of metals.

NOTATION

h, distance between probing radiation and target surface, mm; *I*, relative radiation intensity, rel. units; *t* and τ , ns and μ s; *v*, velocity of plasma front motion, km/s.

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