

## EFFECT OF HIGHLY CONCENTRATED ENERGY FLUXES ON MATERIALS

### FORMATION OF THE CONDENSED PHASE OF METALS EXPOSED TO SUBMICROSECOND LASER PULSES

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*Using the method of laser probing, the laws governing the formation of the liquid-droplet phase of a number of metals (Ni, Zn, Pb, Ag, and Cu) on exposure of metal targets to intense submicrosecond pulses have been determined. It has been established that condensation from the vapor of the erosive laser jet is the main mechanism of formation of the liquid-droplet phase of a metal under given conditions.*

**Keywords:** erosive laser jet, submicrosecond radiation pulses, laser probing, condensed phase of metals.

**Introduction.** With the advent of powerful lasers, the range of practical problems whose solution has become possible due to the introduction of rapidly developing laser technologies has extended considerably. The intense use of lasers was begun in both scientific investigations and in industry, where lasers are applied in various technologies of metal working (hardening, welding, cutting, drilling). First of all, these are solid-state lasers based on glasses and crystals activated with neodymium. The energy and time characteristics of radiation pulses generated by such lasers differ considerably. While for rather long (10  $\mu$ sec–10 msec) pulses of moderate power density the processes of interaction of laser radiation with metals have been well studied [1–4], reliable experimental information on these processes for short pulses of high power density is obviously scarce.

The present work is devoted to investigation of the principal aspects of the formation of the liquid-droplet phase of metals in erosive jets in the case of submicrosecond ( $\sim 200$  nsec) intense laser action.

**Experimental Techniques.** The parameters of the condensed phase of metals were controlled with the aid of the technique of laser probing based on the dependence of the parameters of absorption and scattering of the particles of the condensed phase of metals on their size. This technique allows one, in real time, to follow changes in the average size of particles and in their concentration in the assigned near-surface region of a target (for more details see [5]). From the dependence of the increase in the scattered component on time one can also determine the approximate velocity of the condensed phase motion.

For this investigation we selected the metals Ni, Zn, Pb, Ag, and Cu, which differ greatly in such parameters as the melting and boiling temperatures and in the heat of evaporation. This allowed us to understand, at the qualitative level, the basic laws governing the processes of formation of the liquid-droplet phase of metals under the given conditions of laser action.

As the actuating laser we used a GOS-1001 standard laser setup operating in the Q-switching mode. To implement such a regime, we used a rotating prism of complete internal reflection (speed of rotation 7000 rev/min). The setup allowed us to obtain laser radiation single pulses with the parameters  $\lambda = 1064$  nm,  $\tau \sim 200$  nsec, and  $E = 2\text{--}5$  J and with an outgoing beam diameter of 30 mm. The laser pulse form is shown in Fig. 1. On focusing such radiation to spots of diameters 1 and 3 mm, a power density of  $10^9$  and  $10^8$  W/cm<sup>2</sup>, respectively, was attained on the target surface.

The probing of the erosive jet was made by the radiation emitted by a ruby laser operating in the free-generation regime. This regime was realized with the aid of a cavity made from plane-parallel mirrors (Fabry–Perot cavity) with the reflection coefficients 0.98 and 0.70. The time structure of the ruby laser radiation operating in the free-generation regime is a set of approximately 100 spikes of length of about 1  $\mu$ sec each (total duration of the generation pulse 900–1000  $\mu$ sec), thus ensuring the time resolution of about 5–10  $\mu$ sec realizable by means of registration

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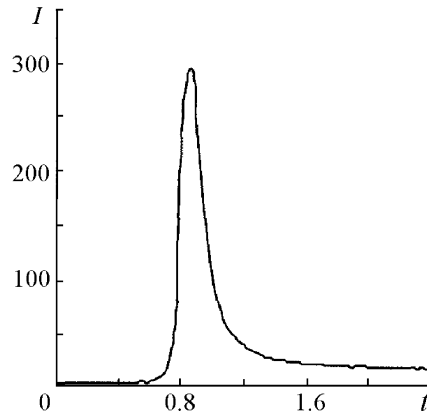


Fig. 1. Time form of the acting radiation pulse.  $I$ , rel. units;  $t$ ,  $\mu\text{sec}$ .

of individual spikes generated at random moments. It should be noted that on the whole, despite the stochastic character, the generation of the ruby laser rather homogeneously covers the studied time interval and can be used as a nonlinear time scale. The diameter of the probing beam in the near-surface zone of the target amounts in this case to 0.2–0.5 mm depending on the aim of investigations.

The characteristic plasma-dynamic times (in our case equal to about 1  $\mu\text{sec}$ ) and the high rate of change in the intensity in the probing radiation pulse (duration of the rise front about 500 nsec) impose speed-of-response requirement on the arrangement of the analog-to-digital converter (ADC) of the recording system. In the system that controls the parameters of erosive laser jets, two plates of a fast 40M/10-2 AD converter are used with a discretization frequency of 40 MHz. To ensure a coordinated operation of the whole facility, a system of synchronization is used which is based on a G 200 P multichannel delayed pulse generator produced in quantity by the Belarusian enterprise "Spetspribor" (Special-Purpose Devices). The facility makes it possible to generate the firing pulses at different (for each of the 5 channels) time instants, which compensates the time delays caused by the different speeds of operation of pumping lamps for the actuating and probing lasers and the timely switching of the recording system.

**Discussion of Results.** To verify the mechanism underlying the formation of the liquid-droplet phase of the erosive laser jet of zinc (on exposure of a zinc target to intense submicrosecond pulses), the time dependences of the transmissivity of the jet, effective diameter of particles, and of their concentration were investigated in [6]. It was established that beginning from a certain time (20–40 nsec) after exposure of the zinc target to laser radiation of submicrosecond duration an erosive laser jet with an intense glow of destruction products is formed. With fall in the actuating radiation, the intensity of the jet glow decreases, and fine liquid-droplet particles of the target material are formed from the jet vapor. With time their dimensions increase and the concentration decreases, which shows that condensation is the most probable mechanism of their formation. Measurements of the laser erosive jet transparency in a wide time range also point qualitatively to the occurrence of condensation processes in the jet.

To investigate the basic trends in the laser erosion of a wider range of metals by submicrosecond pulses, the time dependences of the effective diameter and concentration of the liquid-droplet phase particles of the target material were recorded in the present work at different power densities of the actuating radiation ( $10^8$  and  $10^9$   $\text{W}/\text{cm}^2$ ). Laser probing was carried out at a distance of 1 mm from the target surface.

In order to understand the basic laws governing the processes of formation of the liquid-droplet phase of metals exposed to the action of submicrosecond pulses, it is sufficient to consider in detail the behavior of the time dependence of the indicated parameters for Zn, Pb, and Ni. This is due to the fact that zinc has the least difference between the temperatures of melting and boiling at a low evaporation heat; lead is most easily fusible, whereas nickel has high temperatures of melting and boiling and a high evaporation heat. Moreover, the metals have close reflection coefficients at a radiation wavelength of 1064 nm, i.e., they absorb approximately the same fraction of energy of the actuating laser.

To investigate the process of laser erosion of the above-indicated metals, targets with different grades of roughness and with average transverse dimensions of unevenness in the form of longitudinal scratches 3 and 50–100  $\mu\text{m}$  long were manufactured. When pulsed laser radiation with a power density of  $10^9$  and  $10^8$   $\text{W}/\text{cm}^2$  acted on them,

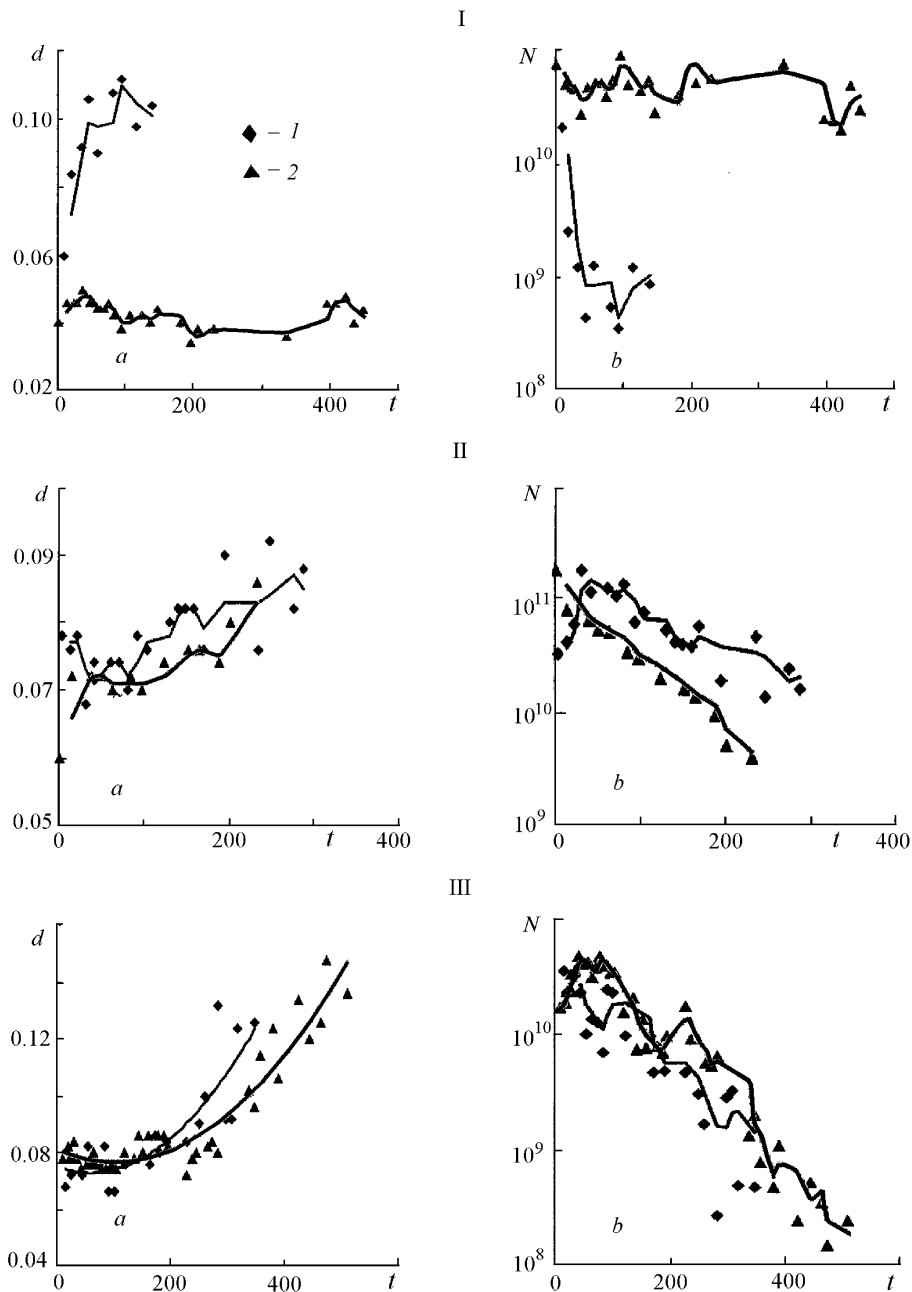


Fig. 2. Time dependences of the diameters (a) and concentrations (b) of the particles of the liquid-droplet phase of nickel (I), zinc (II), and plumbum (III) on their erosion by laser pulses with a power density of  $10^9$  W/cm<sup>2</sup> (1) and  $10^8$  W/cm<sup>2</sup> (2).  $d$ ,  $\mu\text{m}$ ;  $N$ , cm<sup>-3</sup>.  $t$ ,  $\mu\text{sec}$ .

it was established that the sharp unevennesses of the target surface of size  $\sim 50$ – $100$   $\mu\text{m}$  considerably increase the ejection of the condensed phase of metals as compared to a smoother relief, other things being equal.

The particles formed as a result of the laser erosion of nickel by more intense pulses ( $10^9$  W/cm<sup>2</sup>) differ noticeably in their sizes, taking on values of diameters from 60 to 110, which cannot be said of the particles formed on exposure to a pulse of power density of  $10^8$  W/cm<sup>2</sup>: they remain practically unchanged ( $\sim 50$  nm) over the course of probing (see Fig. 2, I, a). The same features are also typical of the dynamics of their concentrations (see Fig. 2, I, b); for higher-intensity pulses it decreases substantially ( $3 \cdot 10^{11} \rightarrow 5 \cdot 10^9$  cm<sup>-3</sup>), remaining practically constant ( $5 \cdot 10^{11}$  cm<sup>-3</sup>) for less intense pulses. The behavior of the liquid-droplet phase of zinc in the case of laser erosion has a num-

TABLE 1. Limits of Variations of the Effective Diameters and Concentrations of the Liquid-Droplet Particles of Metals and Some Physical Parameters of the Corresponding Metals [7, 8]

Metal	Acting radiation power density, W/cm <sup>2</sup>				Temperature, °C		Specific heat of evaporation, kJ/mole	Reflection coefficient, % ( $\lambda = 100$ nm)
	10 <sup>8</sup>		10 <sup>9</sup>		melting	boiling		
	<i>d</i> , nm	<i>N</i> , cm <sup>-3</sup>	<i>d</i> , nm	<i>N</i> , cm <sup>-3</sup>				
Zn	60–90	2·10 <sup>11</sup> –4·10 <sup>9</sup>	70–90	2·10 <sup>11</sup> –2·10 <sup>10</sup>	420	907	114	69
Pb	80–140	3·10 <sup>10</sup> –2·10 <sup>8</sup>	70–130	7·10 <sup>10</sup> –5·10 <sup>9</sup>	327	1745	178	80
Ni	50	3·10 <sup>11</sup>	60–110	3·10 <sup>11</sup> –5·10 <sup>9</sup>	1455	2780–2910	370	72
Ag	40	7·10 <sup>12</sup>	35	3·10 <sup>13</sup>	962	2170	266	97
Cu	40	5·10 <sup>11</sup>	50	3·10 <sup>11</sup>	1085	2540	302	90

ber of distinctive features as compared to the previous case (Ni). The dynamics of the diameters of particles for different power densities of actuating pulses practically coincides (see Fig. 2, II, a); it is seen from the figure that the average diameter of particles is equal in this case to 80 nm. The time dependences of concentrations (see Fig. 2, II, b) decrease monotonically — more slowly for a more intense pulse and more rapidly for a less intense one. In comparing the time dependences of the diameters and concentrations of the particles of the liquid-droplet phase of lead, it is seen (Fig. 2, III) that the curves that describe the effect of pulses of different intensities practically superpose on one another, pointing to the similar dynamics of droplet formation. The diameters of the particles formed lie in the range 70–140 nm, and their concentrations within  $2 \cdot 10^8$ – $8 \cdot 10^{10}$  cm<sup>-3</sup>.

The approximate velocity of motion of the condensed phase particles in the case of laser erosion by submicrosecond pulses amounts to ~65 m/sec for nickel, ~80 m/sec for zinc, and ~45 m/sec for lead (in the direction of the normal to the target surface).

The limits of variations of the effective diameters and concentrations of liquid-droplet particles of metals during the time of probing at the indicated parameters of laser action, as well as some physical parameters of the investigated metals, are given in Table 1. It is seen from this table that the particles of the liquid-droplet phase of the metals investigated have dimensions of the order of several tens of nanometers at appreciable concentrations in erosive laser jets.

Parallel with the given experiments, we carried out estimates of the efficiency of formation of the nanodimensional condensed phase of metals for the case of multiple laser action at the same cite on the target surface, which is of great practical interest for problems in the processing of metals by a sequence of pulses. Independently of the initial roughness of the target surface, after two to three radiation pulses (with the above-mentioned parameters) the conditions for the formation of the condensed phase of the target material differ little from smooth targets with an average size of irregularities of about 3 μm.

Summarizing the foregoing, the following general conclusions can be drawn:

1. The effect of improved conditions for the formation of the liquid-droplet phase of metals on increase in the roughness of the target surface has been revealed in all of the metals investigated. This can be attributed to the growth of the effective area of interaction of radiation with the target surface, as well as to the presence of a great number of nucleation sites compared to smooth targets. The effect of the smoothing of the target surface by repeatedly acting laser pulses has been established.

2. In laser erosion of metal targets by submicrosecond radiation pulses of power densities 10<sup>8</sup> and 10<sup>9</sup> W/cm<sup>2</sup> a substantial quantity (10<sup>8</sup>–10<sup>12</sup> cm<sup>-3</sup>) of nanodimensional particles (40–140 nm) of the target material is formed depending on the type of the metal and action conditions. Here, the particles of a more refractory metal (nickel) have a smaller size and higher concentration as compared to more low-melting zinc and lead (other conditions of laser action being equal).

3. One of the characteristic features of the laser erosion of metals under the indicated conditions is the long-term presence of the condensed phase of the target material in the near-surface region — up to 500 μsec after the laser action. The average velocity of motion of the liquid-droplet phase particles of metals in erosive laser jets

amounts in this case to 50–100 m/sec, depending on the type of the metal, which is in good agreement with the well-known data [9] for longer pulses.

4. The results of investigations carried out in the indicated range of the laser radiation power density for the metals studied indicate that for nickel the dimensions of the liquid-droplet phase particles somewhat decrease on decrease in the acting radiation intensity. In this case, the dynamics of the dimensions of particles in different regimes of laser erosion differ greatly. In the case of a more intense action their diameters undergo a sharp increase from 60 nm to 100–110 nm in 100  $\mu$ sec; a less intense regime practically does not change the sizes of the particles. This may be due to the fact that more powerful pulses cause more intense destruction of a nickel target in the course of erosion, thus substantially increasing the saturation of the target material vapor in the zone of action, allowing the particles to noticeable increase in their size due to the condensation of the vapor cooling-off on them. This is also responsible for the decrease in their concentration. For zinc the curves of the diameters under different regimes of laser action practically coincide, pointing to the identical conditions of droplet formation. Some difference in the dynamics of the concentrations of particles can be attributed to the fact that a more intense pulse is capable of removing more substance from the zone of action that afterwards condenses into droplets. This is due to the small difference (480°C) between the melting and boiling temperatures. The same trends are revealed in analyzing the results of investigation of lead targets, with the only difference that the behavior of the concentrations of particles in different regimes of exposure coincides in this case. This can be explained by the substantial difference between the melting and boiling temperatures (>1400°C), which does not allow a more intense pulse to vaporize a noticeably larger volume of substance as compared to a less intense one.

As to the laser erosion of silver and copper at the indicated parameters of laser radiation, the absence of the dynamics of the liquid-droplet phase particle dimensions can be explained by the small fraction of the absorbed energy (because of the high reflection coefficient for these metals at the acting laser wavelength) and by the comparatively small destruction of the target. The small amount of the substance ejected from the target surface prevents the earlier formed particles from growing noticeably. This applies also to the case of low-intensity laser effect (power density  $10^8$  W/cm<sup>2</sup>) on nickel.

**Conclusions.** As a result of the investigations of the interaction of a powerful laser pulse radiation of submicrosecond length with some metal targets (Ni, Zn, Pb, Ag, and Cu) the presence of a considerable quantity of nano-dimensional particles (40–140 nm depending on experimental conditions) in the near-surface region of the erosional laser jet of the target material has been established. In contrast to laser erosion by long laser pulses (10  $\mu$ sec–10 msec), the regime of use of submicrosecond pulses has a number of characteristic features:

1. The principal mechanism of formation of the liquid-droplet phase under the described conditions of laser action is condensation from the erosive jet plasma vapor. The particles formed in an erosive laser jet are present near the target surface for a long time after the laser action (up to 500–600  $\mu$ sec after the action).

2. During the entire time of probing, the particles of the condensed phase of the target material have nanometer dimensions, which makes the investigated regime of laser erosion convenient for the formation of metal nanostructures because of the absence of particles of any other dimensions in the jet.

3. The efficiency of formation of the liquid-droplet phase of metals is substantially influenced by the roughness of the target surface. The effect of smoothing of the target relief by repeated pulses was noticed. This imposes certain restrictions on the use of frequency lasers for the realization of the given regime of action. To ensure a high efficiency of the frequency regimes of the processing of metals it is necessary to constantly change the localization of the acting radiation focusing spot on the target surface.

## NOTATION

$d$ , effective dimension of particles,  $\mu$ m;  $E$ , integral energy in a pulse, J;  $I$ , relative intensity of radiation;  $N$ , numerical density of particles, cm<sup>-3</sup>;  $t$ , time,  $\mu$ sec;  $\lambda$ , radiation wavelength, nm;  $\tau$ , duration of radiation pulse, nsec.

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