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Nonlinear increase in the interaction efficiency of a second pulse with a target upon excitation of a plasma by a train of pulses from a Nd : YAG laser

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***Abstract*. The efficiency of hole drilling in an aluminium plate was studied experimentally upon excitation of a plasma on its surface in air by a train of pulses from a Nd : YAG laser, the interval between pulses being 15 – 20 μs. It was found that the crater depth increases nonmonotonically with each successive pulse of the train. A nonlinear, more than by a factor of six, increase in the depth was detected upon interaction of the second pulse with the target. The mechanism explaining this increase in the interaction efficiency of the second pulse in the train with the target is proposed.**

Keywords: *hole drilling with laser pulse trains, laser plasma on the target surface, interaction efficiency with the target.*

1. Introduction

The advent of lasers capable of exciting plasma on the target surface stimulated the intensive research on the mechanism of interaction of laser radiation with a target [1]. One of the directions in the research focused on laser 'drilling' of holes, laser cutting and other technologies [2 – 5]. In this case, different regimes (cw, pulsed, and their combination) of laser action on materials were investigated [2, 3]. It was found in [2, 3] that in the pulsed regime of irradiation, the use of a train of laser pulses considerably increases the speed of hole drilling and cutting compared to the use of a single pulse.

Note that a two-pulse irradiation regime drastically changes the emission spectrum of the plasma plume. In 1987 we could achieve, obviously for the first time, a tenfold increase in the contrast of plasma spectral lines and significant improvement in the sensitivity of the qualitative analysis of samples in air [6]. The improved sensitivity was obtained when plasma was generated by two successive pulses spaced by several microseconds. Note that at the same time a similar operation regime was patented in Japan [7] without publishing the finding. The idea proved rather fruitful. At present this technique is being improved by optimising the pulse duration and spacing [8, 9]. The optimisation of the irradiation regime made it possible to achieve a record sensitivity (a few ppm) of impurity detection [10]. To explain this effect, we proposed an interaction mechanism which took into account a decrease in the density of atmospheric gases over the target surface before the arrival of the second laser pulse [11]. Later, this mechanism was explained and verified experimentally [9].

However, as far as we know, the question of how the efficiency of the pulse – target interaction varies with the number of pulses in the train has not received an adequate explanation. Based on the nonlinear increase in the contrast of the emission spectrum upon two-pulse excitation of plasma [6, 9, 11], we can expect the second pulse in the train to make the greatest contribution to the speed of hole drilling or the interaction efficiency. The aim of this paper is to verify experimentally this assumption.

To test the hypothesis about the nonadditive nonlinear energy increase of the second pulse in the target, we measured experimentally the speed of hole drilling of a metal plate and integrated luminosity of chemical elements of the target material for different numbers of Nd : YAG-laser pulses whose amplitudes are almost equal.

2. Experiment

We used a typical experimental setup for studying laser plasma and pulse – target interaction [6 – 11]. We employed a pulsed Nd : YAG laser with a passive Q switch in the form of a LiF crystal containing F_2^--colour centres (initial transmittance was 22 %). The laser generated single pulses and pulse trains at a wavelength of 1064 nm. The pulse full width at half-maximum was 10 ns, and its energy after amplification was 25 mJ. The laser generated a single transverse mode with the beam intensity distribution similar to the Gaussian one. When the pump energy exceeded the lasing threshold for the first pulse by 10 % to 80 %, the laser could switch to a multipulse regime with the pulse spacing of 15 – 20 μs. An increase in the pump energy by 10 % – 15 % from the previous value was required to increase the number of pulses in the train. In the multipulse operation regime the energy of each consecutive pulse dropped insignificantly so that the pulse amplitude can be regarded the same with a 10 % accuracy.

A lens with the focal length of 87 mm was used to focus radiation on the target surface. The target could move along the beam axis in its waist to provide variations in the radiation intensity within $10^8 - 5 \times 10^{10}$ W cm^{-2} on the target surface. A photodiode connected to an oscilloscope was placed behind the target to detect the time the target

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was drilled. The emission spectrum of the plasma plume at the polychromator output was recorded with an optical multichannel analyser with a cooled and gated light amplifier [8–10].

3. Discussion of the results

Figure 1 shows the emission spectra of the plasma produced on the surface of a bronze sample by a single pulse and two pulses. One can clearly see a manifold increase in the contrast and intensity of the spectral lines, e.g., the lines of copper Cu (282.44 nm), tin Sn (286.33 nm), and lead Pb (287.33 nm), which are virtually indiscernible during the single-pulse operation regime. The experiment showed a significant and nonadditive transformation of the emission spectrum upon switching to the two-pulse irradiation of the surface. Note that along with the increased amplitude and contrast of the plasma spectral lines due to the action of the second pulse, Fig. 1 demonstrates the absence of self-absorption [see, for example, line Sn (284.00 nm)], which distorts the real amplitude during the multipulse action (thin curve). According to the above explanation of spectrum distinctions during the two-pulse action [11], the plasma plume produced by the second pulse expands over the target surface in a less-dense gas in which the concentration of 'cooled' tin atoms at the plasma–gas interface is smaller than that during the gas breakdown by the first pulse when the plasma plume front is in contact with the cool and dense gas of the surrounding atmosphere. Therefore, the self-absorption is almost absent in the spectrum after the action of the second pulse. This is very important in a qualitative analysis of the target composition when the line intensity is proportional to the chemical element concentration [3, 12].

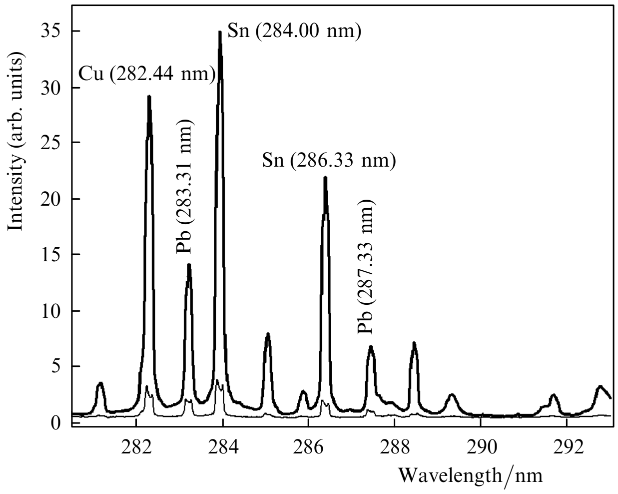

Figure 1. Emission spectra of a plasma plume upon irradiation of the surface of a bronze sample by a single laser pulse (thin curve) and a train of two pulses (thick curve).

During the experiment holes were drilled in a 0.2-mm-thick aluminium foil for the laser power density $\sim 5\times 10^9$ W cm^{-2} on the target surface. This power density on the target surface was obtained by moving the target away from the focal plane of the lens. The beam diameter was determined by the size of the burn, which appeared on the target surface after its irradiation by five–seven single pulses, two double-pulse trains, and one train in the multipulse regime. In our case, five series of measurements were made to give the average beam diameter of 130 ± 10 μm. Figure 2 presents the dependences of the number N of pulse trains required to drill a hole in the foil and of the speed of hole drilling normalised to the speed of hole drilling in the single-pulse regime on the number i of pulses in the train. The speed of hole drilling was calculated under the assumption of a cylindrical profile of the crater. Taking into account the ratio between the beam diameter on the surface (~ 130 μm) and foil thickness (200 μm), we assumed that the interaction behaviour changes with the crater depth in the same fashion for all the regimes of hole drilling.

The results of the experiment showed that the passage from the single-pulse to double-pulse irradiation regime leads to a noticeable nonlinear growth in the efficiency of the radiation–target interaction (solid line in Fig. 2).

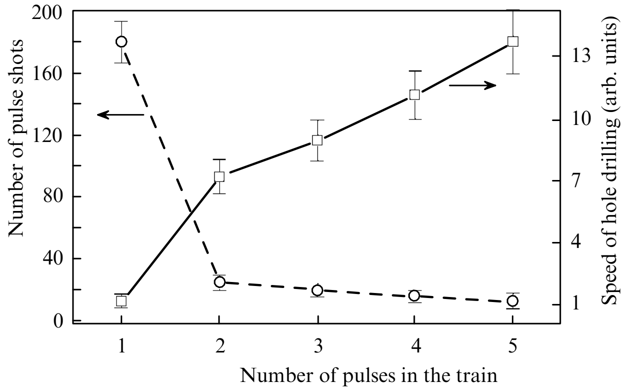

Figure 2. Dependences of the number of laser pulse shots needed for drilling a hole in a 0.2-mm-thick aluminium foil and speed of hole drilling on the number of pulses in the train.

Together with hole drilling the emission spectrum of the plasma plume was observed at 624.3 nm (the Al II spectral line) and 500.5 nm (the N II spectral line of atmospheric nitrogen). Figure 3 shows the dependences of the intensities of these spectral lines on the number of pulses in the train. The similarity of the dependences of the efficiency of the energy input and the intensity of the analytic Al II line (624.3 nm) of the material on the number of pulses in the train (Figs 2 and 3) shows that the growth in the analytic signal during the action of the second pulse is caused by the escape of a new portion of the material from the target.

In addition, one can see from Figs 2 and 3 that during the repetitively pulsed irradiation a change in the plasma-forming medium occurs. Thus, the air plasma [see the curve for N II spectral line (500.5 nm) in Fig. 3] dominating in the laser plume due to the first pulse is replaced almost completely by the erosion plasma from the target upon its irradiation by other pulse trains. In this case, starting with the third pulse, the effect of interaction repeats and hardly changes with each successive pulse. Note that expelling the atmospheric nitrogen from the interaction region agrees well with the interaction mechanism of double-pulse radiation with the target [11].

The observed decrease in the spectral line intensities and the ejection of the target material after the action of the second pulse in the train (Figs 2 and 3) can be interpreted as

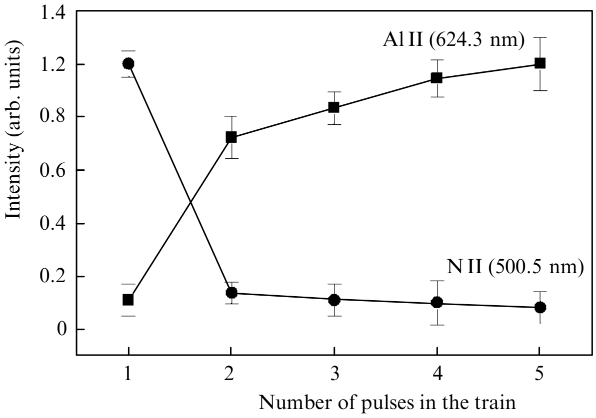

Figure 3. Dependences of the emission intensity of ions in the plasma plume at wavelengths of Al II (624.3 nm) and N II (500.5 nm) spectral lines on the number of pulses in the train.

screening of the surface by a cloud of erosion (aerosol) particles produced by the second pulse. At first glance, this screening is similar to the screening of the surface by atmospheric nitrogen molecules after the first pulse whose concentration does not have time to reach its initial value by the arrival of the second pulse because of high superficial temperature [11]. The change in the plasma-forming medium (mostly nitrogen molecules during the breakdown by the first pulse) to that of the erosion type at the breakdown by the third pulse does not result in a significant decrease in the concentration of erosion particles at the breakdown by the fourth pulse as was the case during the breakdown by the first pulse. For this reason, the interaction efficiency of the third, fourth and fifth pulses with the target is less pronounced than that of the second pulse.

To find how much each consecutive pulse in the train contributes to the interaction efficiency, we will calculate the increase in the crater depth Δh produced by this particular pulse (last in the train) using the recurrent relation:

$$h = N_1 \Delta h_1 = N_2 \Delta H_2. \tag{1}$$

Here, h is the sample thickness; N_1 and N_2 are the numbers of laser shots needed for drilling a hole in the target; Δh_1 and ΔH_2 are the increase in the crater depth (or the increase in the speed of hole drilling) measured in mm per shot for the single- and double-pulse regimes, respectively. In the general case, the action of a train of identical equally-spaced pulses on the target can be written in the form

$$N_i \Delta H_i = N_{i+1} \Delta H_{i+1}, \tag{2}$$

where the subscript i is the number of pulses in the train. The total interaction efficiency of the whole pulse train ΔH_{i+1} can be written as a sum of the contribution Δh_{i+1} of the last pulse in the train and the increase in the crater depth produced by the preceding pulses:

$$\Delta H_{i+1} = \Delta h_{i+1} + \Delta H_i. \tag{3}$$

Then the contribution of the last pulse, or its relative efficiency in the train (from the viewpoint of its action on the target), can be estimated with the help of relations (2) and (3):

$$\Delta h_{i+1} = (N_i / N_{i+1} - 1) \Delta H_i. \tag{4}$$

Table 1 presents experimental data illustrating the decrease in the number of laser shots with increasing the number of Nd : YAG laser pulses in the train needed to drill a hole in a 0.2-mm-thick aluminium foil. Assuming that the rate of hole drilling by a single pulse is constant, each of 180 pulses (see Table 1) gives the same increase in the crater depth:

$$\Delta h_1 = \Delta H_1 = h / N_1. \tag{5}$$

Table 1. The number of laser shots required to drill a hole by a train of pulses of different durations and the interaction efficiency of pulses with a target.

The number of pulses in the train	The number of laser shots	Interaction efficiency of the last pulse in the train with the target
1	180	1
2	25	6.2
3	20	1.8
4	16	2.25
5	13	2.45

By using recurrent expression (4), we obtain that the efficiency of the second pulse in the train compared to that of the first pulse is determined by the relation

$$\Delta h_2 / \Delta h_1 = (180/25 - 1) = 6.2. \tag{6}$$

Hence, continuing the calculations and assuming that

$$\Delta h_2 = 6.2 \Delta h_1 = 6.2 h / 180, \tag{7}$$

we estimate that the efficiency of the third pulse compared to the first one is

$$\Delta h_3 / \Delta h_1 = (N_2 / N_3 - 1) \Delta H_2 / \Delta h_1 = (25/20 - 1) \times (\Delta h_2 / \Delta h_1 + 1) = 0.25 \times 7.2 = 1.8. \tag{8}$$

Thus, calculations show that the second pulse in the train increases the crater depth by 6.2 times compared to the first pulse and by almost 3 times compared to the next pulses (Table 1).

Figure 4 shows the calculated dependence of the increase in the crater depth after the action of each last pulse in the train compared to the action by the first pulse. One can clearly see the nonadditive nonlinear increase in the interaction efficiency of the second pulse with the target, which is manifested in the increased ejection of the sample material and greater speed of hole drilling.

Note that if the number of pulses is from 2 to 5, the total interaction efficiency (Table 1) is close, within the experimental accuracy, to the integral increase in the spectral line intensity of the aluminium ion (see Fig. 3) upon irradiation of the target by a train of five pulses. This result additionally explains the mechanism of the nonlinear increase in spectral line intensities of chemical elements of the target material in the plasma plume during the two-pulse interaction [11]. Unlike the interaction model proposed in [12, 13] according to which the optical breakdown of the gas-dust cloud by the second pulse at the target surface is responsible for the increase in the spectral line intensities, in our model this

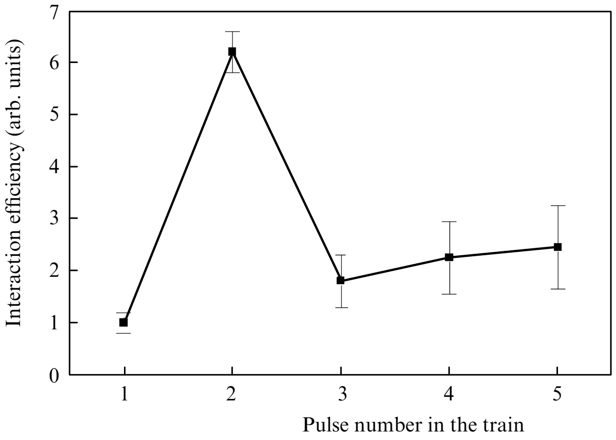

Figure 4. Dependence of the interaction efficiency (the increase in the crater depth) of each pulse with the target on its number in a five-pulse train.

growth is explained by the increased amount of the material ejected from the target by the second pulse.

Thus, one of the effects of the first pulse on the target surface is the generation of a 'fireball' [14] and the heating of the surface region to a temperature of about 10^4 K, which cools down slowly. Experiments [15] with the microwave diagnostics of the laser plasma showed that the surface region (heated initially to 10000 – 20000 K) cools down for a relatively long time (about one hundred microseconds). At the same time, the pressure in the heated region lowers in a much shorter period, which is determined by propagation of sound through this region (a few microseconds). Taking into account the ratio of temperatures (about 300 K/15000 K), it is possible to evaluate a decrease in the atmospheric gas concentration during cooling (by a factor of $\sim$ 50) by using the Mendeleev – Clapeyron equation under the assumption that the volume of the heated region is constant after pressure equalization.

The analysis of the results shows that in the given case the first pulse acts as a vacuum pump (by providing the conditions similar to those in a low-pressure chamber) and 'prepares' the target surface (by removing oxides and contamination and heating it) to the arrival of the second pulse. In this case, the second pulse does not initiate an optical breakdown in the cloud of particles produced by the first pulse as was thought earlier [12, 13], but interacts directly with the target surface, the efficiency being greater than that of the first pulse. The second pulse ejects more material from the target (Fig. 4), evaporates it and only then ionises it in the optical breakdown which results in the increased intensity and contrast of spectral lines.

Taking this interaction mechanism into account, the authors of paper [16] were able for the first time (as far as we know) to obtain generation of X-ray pulses upon irradiation of an aluminium target by double femtosecond pulses in the atmosphere under normal conditions.

Note also that the third pulse in the train interacts with the target three times less efficiently than the second one (see Fig. 4 and Table 1). The energy input of next train pulses (the fourth and fifth) in the target only slightly increases compared to that of the third pulse. This decrease in the interaction efficiency (the increase in the crater depth in other words) can be caused by two main factors: a further rise of temperature of the crater surface by the arrival of the third pulse and an increased concentration of particles removed from the target by the second pulse. Both factors cause a decrease in the breakdown threshold [2 – 4] and plasma formation at the leading edge of the pulse, which leads to the screening of the target and prevents a part of the pulse energy from arriving at the surface. For the fourth and fifth pulses these interaction conditions do not seem to change considerably. Therefore, the increase in the crater depth is comparable with the contribution of the third pulse (Fig. 4).

4. Conclusions

Thus, in this research we have detected a nonlinear (by a factor of six) increase in the interaction efficiency of the second pulse with the target compared to the first pulse upon irradiation of an aluminium target by a train of two nanosecond pulses of equal energies from a Nd : YAG laser. An increased ejection of the target material provides an additional argument for applicability of the earlier proposed physical mechanism of interaction of laser pulses spaced by a few microseconds with the target. In this case, the effects of the pulses differ greatly. The first pulse heats the interaction region, which lowers the density of the surrounding air and decreases the screening effect of the atmosphere during the transportation of the energy of the second pulse to the surface. The second pulse additionally heats and evaporates the target material, interacts more efficiently with it by increasing nonlinearly the speed of energy penetration to the target and the removal of the material, heats and ionises the erosion particles in the ambient air with the reduced density.

The applicability of the results is no less important. Thus, the decrease in the thermal load on the target is obvious, when two pulses rather than multipulse trains are used in special technological processes susceptible to high temperatures like laser welding/cutting of micro- and nanomaterials, sputtering of macromolecules without their substantial overheating, breakage, thermal destruction, etc. [2, 3]. Note that the use of the double-pulse regime leads to an increase in the speed of hole drilling by a factor of 7.2 (see Table 1) upon doubling the total irradiation energy.

The advantage of the double-pulse regime is particularly important in laser treatment of living tissues [17 – 19]. Laser drilling of holes (perforating) inside the eyeball without its dissection is obviously most sensitive to high temperatures because of possible injuries to the eye tissue and retina [17]. It is the double-pulse regime of laser drilling [18] and cutting [19] of tissues that allows avoiding the risk of thermal destruction of dental tissues. On the whole, a manifold increase in the interaction efficiency of laser radiation with a target by simply changing the regime of pulse generation allows saving material resources and meets modern requirements for energy-saving technologies.

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