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Optical spectroscopy of laser plasma in a deep crater

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***Abstract*.** **The time dynamics of plasma-emission spectra is studied experimentally at different stages of the drilling of a steel plate by 100-fs and 5-ps laser pulses: from a shallow crater to a hole. The change in the time dependence of the plasma temperature caused by variations in the irradiated surface geometry is analysed. It is found that the time interval needed to reach a particular temperature (about 8000 K) drastically increases from 40 – 50 to 150 – 200 ns when a specific crater depth is achieved. The opposite tendency is observed as the crater depth grows further and a hole is produced. Strong self-absorption in a plasma plume inside a deep crater is experimentally confirmed which results in the appearance of line absorption against a continuous emission spectrum.**

Keywords: *laser plasma, optical spectroscopy, laser drilling.*

1. Introduction

Modern optical spectroscopy makes it possible to investigate such a nonstationary object as laser plasma with nanosecond resolution. The pulse duration of many modern lasers is of the order of nanoseconds or even shorter, which does not allow spectroscopic measurements to last until the end of the laser pulse during the decay of laser plasma. Nevertheless, spectroscopic investigation of plasma parameters including its ion composition, temperature and electron density is very important for optimisation of such processes as laser deposition of thin films [1], laser treatment of materials [2], laser chemical analysis [3] and other applications.

Early researches determined that the cooling time of laser plasma depends on the energy and duration of a laser pulse [3 – 4], composition and pressure of the ambient atmosphere [2, 5 – 7], the structure of an evaporated target [6]. In this paper we consider how the geometry of an evaporated surface affects the evolution of laser plasma. Having found wide applications, laser drilling is a typical process which involves a radical change in the surface shape being irradiated: an initially flat surface turns into a conical crater which goes as far as the back surface of the sample and changes into a hole with expanding walls. The use of ultra-short (pico- and femtosecond) laser pulses, which are much expected to improve the quality of laser treatment of materials, is an important aspect of the experiments on laser drilling described below.

2. Experimental

A Hurricane Ti : Sapphire laser (Spectra-Physics) emitting 130-fs and 5-ps pulses with a pulse repetition rate of 1 kHz at 800 nm was used for drilling holes in 0.75-mm-thick stainless steel plates. The laser beam was focused by a lens (with the focal length $F = 65$ mm) into a spot 17 μm in diameter (measured at $1/e^2$ level of the intensity maximum) with a Gaussian energy distribution. The focal plane was shifted by 0.2 mm inside the sample from the front surface, as is usually done for decreasing the average time of drilling.

The optical scheme used for measuring emission spectra of plasma in a deep crater is shown in Fig. 1. The plasma was observed along the laser beam axis, which coincided with the axis of the crater. A collecting lens was used to focus plasma radiation onto the spectrometer entrance slit. The emission spectra were recorded at a fixed exposure time of 10 ns, whereas the delay between a pulse hitting the target surface and the start of the spectra recording varied from 10 to 400 ns. A 600-lines mm^{-1} diffraction grating was used to

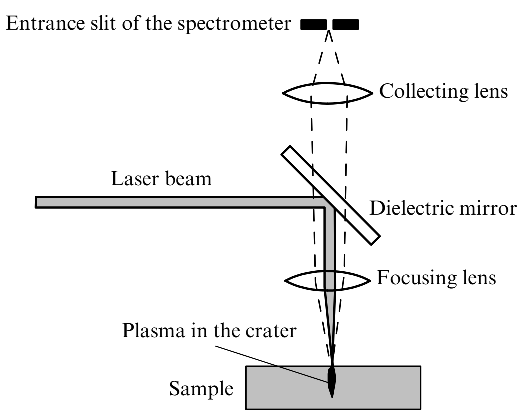

Figure 1. Optical scheme for detecting the emission spectra of the plasma in a crater.

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measure the spectra in the spectral range 370–445 nm. The spectroscopic measurements were performed with a lower pulse repetition rate (10 Hz) by averaging 100 successively recorded spectra.

According to previous research [8], the plasma generated by short laser pulses exhibits conditions that allow local thermodynamic equilibrium, at least during the first few microseconds. In this case, the population of the electronic levels of an atom (ion) is defined by the Boltzmann equation, and the intensity I_{mn} of a spectral line complies with the relation

$$I_{mn} \sim g_m A_{mn} \exp(-E_m/kT), \tag{1}$$

where g_m is the statistical weight of the upper level; A_{mn} is the transition probability; E_m is the energy of the upper level; k is the Boltzmann constant; T is the temperature of free electrons which determines the excitation of electronic levels. It follows from (1) that the dependence of $\ln[I_{mn}\times(g_m A_{mn})^{-1}]$ on E_m can be approximated by a straight line whose inclination is inversely proportional to temperature T. In our case we calculated the temperature with the help of experimentally measured intensities of nine spectral lines of iron in the wavelength range from 371.99 to 376.38 nm, for which the parameters g_m, A_{mn} and E_m are well known [6].

3. Experimental results and discussion

The distinguishing changes in the optical emission spectrum during the plasma cooling after the termination of the laser pulse are given in Fig. 2. When the delay between the laser pulse and start of recording is zero, we have a continuous emission spectrum resulted from recombination radiation and bremsstrahlung of free electrons. Then, the intensity of the continuous spectrum monotonically decreases with increasing the delay time, which indicates a gradual decrease in the free-electron density due to consecutive electron–ion recombination. The appearance and fast growth of the line spectrum in the wavelength range of interest means that a noticeable quantity of neutral atoms of iron and chrome appear in the plasma. The following slow decrease in the spectral line intensities is caused by a gradual fall of the free-electron temperature, which decreases the probability of excitation of atomic optical transitions.

To describe quantitatively the temporal changes in the plasma emission in the first approximation, it is necessary to measure the intensities of the continuous and line spectra at a particular wavelength. For this purpose, we used one of the intensive spectral lines of iron ($\lambda = 432.58$ nm) marked in Fig. 2. Figure 3 allows us to compare the time dynamics of the amplitude variation of this particular line and the intensity of the continuous spectrum for three major types of the geometry of the evaporated surface during laser drilling: a shallow crater, deep crater and a through hole. The results obtained are presented for the 5-ps pulses with the energy $Q = 0.7$ mJ. A shallow crater (Fig. 3a) produced after first 100 laser pulses had the depth $L = 50$ μm, which is much smaller than the crater diameter ($D = 125$ μm). The intensity of the spectral line of iron in this case achieved the maximum for the recording delay time $\tau_{max} = 50$ ns. When a deep crater was formed (Fig. 3b), for which the ratio of the depth L to the input diameter D achieved 4.5, we discovered a much slower decrease in the intensity of the continuous spectrum. The maximum of the spectral line intensity in this case also occurred later ($\tau_{max} = 200$ ns). After the through hole was produced (Fig. 3c), the process reverses, i.e. the delay time corresponding to the maximum line intensity decreases to the initial value.

In the presence of a local thermodynamic equilibrium in the plasma, the maximum intensity of the particular spectral line corresponds to a some fixed temperature of the electrons responsible for excitation of optical transitions in iron atoms. Thus, the curves presented in Fig. 3 allow one to

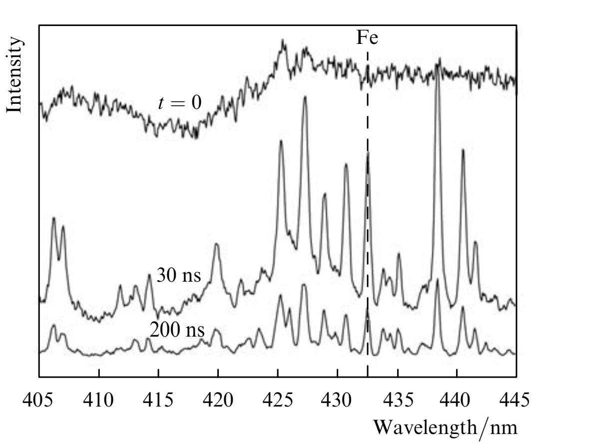

Figure 2. Plasma emission spectra for different time delays t between the arrival of a laser pulse and the start of spectrum recording.

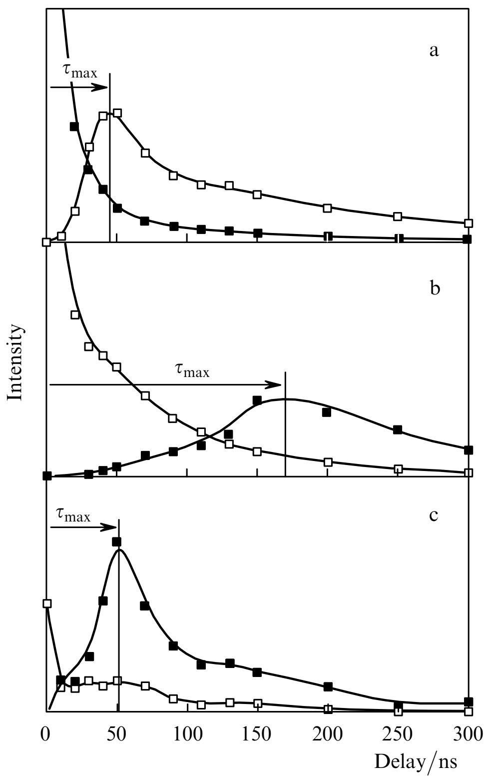

Figure 3. Dependences of the intensities of the spectral line of iron at 432.58 nm (□) and continuous spectrum at the same wavelength (■) on the delay instant in detection for a shallow crater (a), deep crater (b) and hole (c).

estimate the time during which the temperature of the laser plasma falls to a specified level. We can determine this level if we compare the time dependences (Fig. 4) of the electron temperature in the interval from 20 to 400 ns after the laser pulse. These dependences were calculated from the plasma spectra in shallow and deep craters. The curve of the electron temperature that we obtained for a shallow crater agrees well with the results of previous spectroscopic research on laser plasma for a pulse duration of 5 ps [3]. At the same time, the temperature curve for a deep crater proved shifted by approximately 2000 K towards higher temperatures. One can see from the graph that a 50-ns delay on recording in a shallow crater and a 200-ns delay in a deep crater corresponds to the same temperature level of about 8000 K. Therefore, the transformation of the temperature curve within the specified time interval during the crater geometry transformation can be adequately described at the quantitative level as a time shift of the curve determined by a change in the above-introduced parameter τ_{max}. The convenience of this description is largely determined by the fact that a considerable decrease in the line spectrum intensity in the case of a deep crater or through hole has a stronger effect on the accuracy of the calculation of the electron temperature than on determination of the parameter τ_{max}.

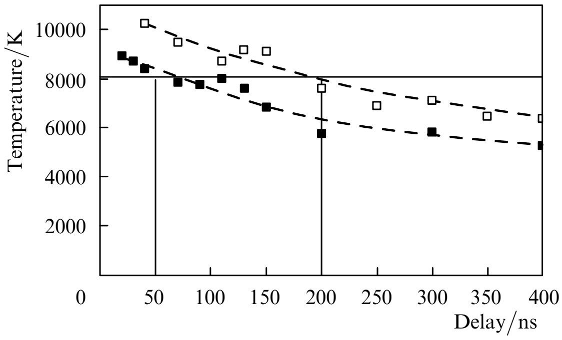

Figure 4. Plasma temperature as a function of the delay in detection for a shallow (■) and deep (□) craters.

Figure 5a shows the change in the parameter τ_{max} during drilling a cylindrical hole by laser pulses of energy $Q =$ 0.7 mJ. Varying the pulse duration from 100 fs to 5 ps did not have a noticeable effect on the measurement results. When the crater depth reached 300–400 μm, τ_{max} grew abruptly from 50 to 150–200 ns and remained at this level till the through hole formation, after which the shift decreased gradually to 20–50 ns. A similar dependence for a lower pulse energy ($Q = 0.1$ mJ) is presented in Fig. 5b. In this case, a relatively small growth of τ_{max} (from 35 to 50 ns) was observed for a smaller crater depth (150–200 μm) after which τ_{max} falls steadily to 20 ns, before the through hole formation. The analysis given below shows that all changes in τ_{max} during laser drilling can be explained by two major factors: by the change in the velocity of plasma cooling as a function of the crater geometry and by the decrease in the initial temperature of the plasma in a very deep crater or a through hole.

When analysing the time dynamics of plasma in a crater, one should take into account the finite plasma spread in space due to resistance of the surrounding air. Experiments proved that the visible size of a plasma plume produced by 0.07–10-ps pulses in the air under the standard atmospheric

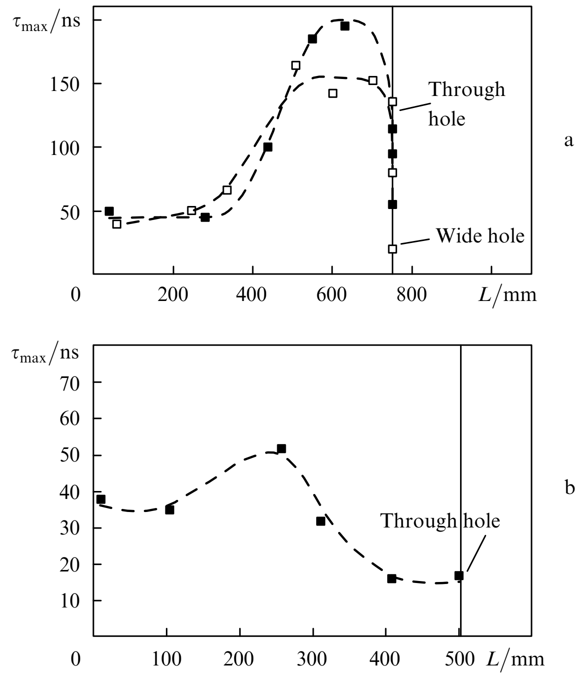

Figure 5. Time shift τ_{max} in detection of the maximum intensity of the iron spectral line ($\lambda = 432.58$ nm) with respect to the action of a laser pulse on the target as a function of the crater depth for a pulse duration of 5 ps (■) and 130 fs (□) at $Q = 0.7$ mJ (a) and for a pulse duration of 5 ps at $Q = 0.1$ mJ (b).

pressure stops growing within 25 ns after the termination of the laser pulse action [9]. The boundary of the glow region (plume) in this case was determined at the 10 % level of the maximum glow intensity. When the pulse energy is relatively low ($Q = 0.02$ mJ), the size of the plasma plume is about 120 μm. In our case we can use the results of investigation of the plasma produced by longer (300 ps) and more powerful pulses from an IR laser [10] to estimate the maximal size of the plume. The plasma emission was integrated over the time interval longer than the plasma lifetime and analysed by using optical spectroscopy with space resolution. When we used the above-mentioned 10 % criterion for the boundary of the glow region, we found the axial size of the plasma plume to be about 250 μm for the pulse energy $Q = 0.2$ mJ. For the intermediate energy $Q = 0.1$ mJ the size of the plasma plume should be about 200 μm, which corresponds to the critical crater depth at which an increase in τ_{max} is observed (see Fig. 5b). Recall that when the pulse energy grows to 0.7 mJ, the critical crater depth grows to 400 μm (see Fig. 5a). This result agrees well with the expected growth in the plume size, which, according to [10], increases about twofold when the pulse energy increases tenfold.

The estimates presented above allow one to relate the growth of the parameter τ_{max} with the plasma plume being fully localised inside the crater. The localisation of the plasma should result in lower emission losses mostly due to partial reflection of plasma radiation from the crater walls and bottom. Another, not so evident reason for the slower cooling of a plasma plume in the crater, is, in our opinion, the fact that the plasma becomes more and more spatially inhomogeneous. In our experiment, short laser pulses do not interact with the erosion plasma, which means that its parameters are defined only by the absorption of radiation in the material followed by evaporation of the material. It is clear that the distribution of the absorbed energy (F_{abs}) across the surface of a deep crater (Fig. 6) differs from the energy distribution across the laser beam (F_{las}). The density of the absorbed energy in the small area of the crater bottom is much higher than on the crater walls where the radiation falls at a greater angle. For this reason, the plasma produced near the crater bottom has a higher ionisation coefficient and electron temperature. The initial expansion velocity of the plasma generated by pico-/femtosecond pulses is about 5×10^5 cm s^{-1} [9].This means that it takes about 10 ns for the relatively 'cold' plasma from the crater walls (the crater diameter is 100 μm) to fill the crater cross section fully and to screen radiation of the hotter plasma from the crater bottom. Thus, mixing of plasma components with different temperatures leads to the appearance of a homogeneous plasma plume exhibiting strong self-absorption, i.e. the most part of emission of the 'hot' plasma component does not leave the plume and is absorbed by the 'cold' component. The energy redistribution between plasma components of different origin leads to the temperature levelling within the plasma volume and, finally, to decreased self-absorption.

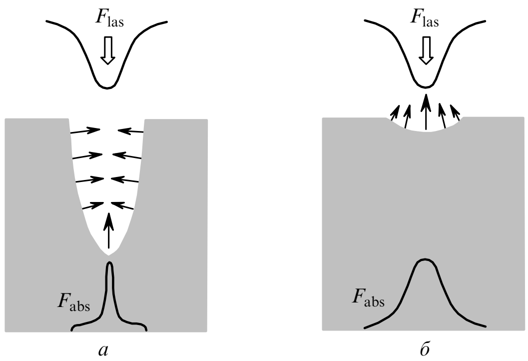

Figure 6. Plasma spread from the irradiated surface and relationship between the energy distribution in the incident Gaussian beam (F_{las}) and the density of the absorbed energy (F_{abs}) in a deep (a) and shallow (b) craters.

We have found the experimental verification of strong plasma self-absorption in the deep crater, which leads to narrow lines of absorption appearing against the continuous emission spectrum. The wavelengths of these lines correspond to the optical transitions in the iron and chrome atoms (Fig. 7). This effect was observed for a limited time interval (10 – 50 ns) after the laser pulse. The lower boundary of this interval corresponds to the time by which the entire section of the crater is filled with less hot plasma from the walls, and the upper boundary characterises the time necessary for the temperature of the plasma plume to level off. Note that the plasma is inhomogeneous to some extent in ablation in a shallow crater; however, it is not enough for the line absorption spectrum to be observed. In this case, the initial inhomogeneity of the plasma is due to only nonuniform energy distribution across the laser beam (Fig. 6b). The regions of relatively 'cold' plasma near the outer boundary of the spot take a considerably smaller area than in the case of a conical crater and cannot shield the emission of the hot plasma in the centre of the light spot.

Thus, an increase τ_{max} in the deep crater is explained by a decrease in the cooling velocity of the plasma plume

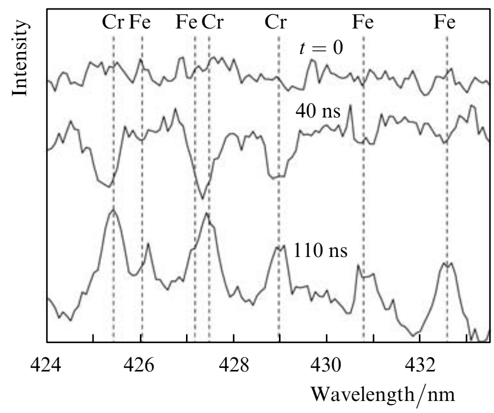

Figure 7. Emission spectra of plasma in the deep crater for different delays t in recording, demonstrating the line absorption effect.

localised inside the 'blind' crater. The formation of a through hole, firstly, violates strict localisation of the plasma inside the crater and decreases the shielding of plasma emission by the crater walls and, secondly, excludes the ablation of the crater bottom and decreases the nonuniformity of the plasma plume. The both factors favour the growth of emission energy losses and a faster decrease in the plasma temperature, i.e. τ_{max} decrease. The fact that the effective energy density of radiation incident on the inclined walls of the through hole decreases with increasing the output diameter of the hole also plays an important role. This leads to a decrease in the initial temperature of the plasma and, therefore, a more rapid achievement of the specified temperature level (about 8000 K). Apparently, parameter τ_{max} falling below the initial level (see Fig. 5a) in a wide through hole is caused by this particular factor.

We believe that a decrease in the initial plasma temperature, which cannot be measured by means of optical spectroscopy, explains a decrease in τ_{max} in a deep but still blind crater at a relatively low pulse energy (see Fig. 5b). A decrease in the energy density of incident radiation near the bottom of a deep crater until the complete stop of ablation, which caused by the divergence of a laser beam and the deformation of the crater walls, was investigated earlier for the case of laser drilling of diamonds [11]. It was found that the lower the laser pulse energy, the smaller depth of the crater is needed for this effect to appear. Another argument in favour of the incident energy density decreasing in a deep crater (Fig. 6) is the decrease in the intensity of the plasma spectra by a factor of 100 – 1000.

4. Conclusions

The experiments have shown that the change in the geometry of irradiated surface during deep drilling by short laser pulses noticeably affects the time dynamics of plasma emission. When the crater depth exceeds some critical value, the time during which the plasma temperature comes down to a particular level grows considerably. This effect is supposed to be brought about by complete localisation of the plasma plume inside the crater, which in turn decreases radiation losses of the plasma. Our experiments have confirmed strong self-absorption in the plasma plume inside a deep crater, which gives rise to a line absorption spectrum appearing against the continuous emission spectrum. The formation of a through hole or deep enough 'blind' crater is accompanied by a decrease in the specified time interval supposedly caused by a decrease in the initial temperature of plasma. Analysis of the time dynamics of the plasma emission may help to solve the problem of the control of the hole geometry during laser drilling.

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