Peculiarities of evolution of a laser plasma jet from a graphite target

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The results of investigation into unstable behavior of a plasma jet produced by the long-pulse CO_2 laser irradiation of a graphite target are reported. A mushroom-like shape of the glowing area is shown to be determined by the Richtmyer–Meshkov instability of the carbon plasma–air interface and formation of nanoparticles in the plasma expanding into the buffer gas.

In recent years, in connection with development of nanotechnology, the interest is growing to physical processes proceeding in the graphite target under its exposure to continuous-wave and repetitively pulsed laser radiation when producing nanopowders and depositing thin films.[1–3] These processes are preceded by formation of a plasma jet as a result of evaporation of the target under the focused laser radiation. The experiments[1,4] revealed the conditions, under which up to 97–100% mass of the target substance comes into the plasma jet in the form of vapor, as indicate a small size and the spherical shape of the nanopowder particles.[3] However, it is still a problem to obtain a good homogeneity of thin films deposited onto material surfaces in the atmosphere.[4] Besides, it is unclear why nanopowder particles generated by the repetitively pulsed laser radiation are fourfold as small as those generated by the cw radiation.[1,3] This points to the necessity of further investigations of characteristics and dynamics of the plasma jet generated under such conditions.

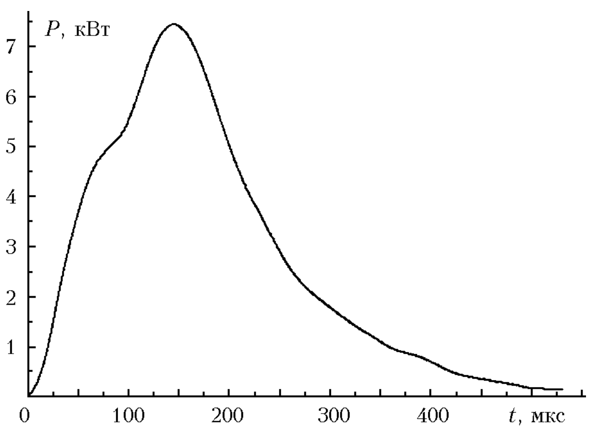

Fig. 1. Pulse shape of the LAERT repetitively pulsed CO_2 laser.

This paper presents the results of experimental and theoretical investigations into the dynamics of the plasma jet. The jet is generated by the laser pulse focused onto a plane graphite or YSZ target. In the experiments, we used a LAERT repetitively pulsed CO_2 laser[3,5] pumped by a combined discharge. The experimental conditions were close to those used in production of nanopowders[3] and deposition of thin films in the atmosphere.[4] The multimode CO_2 laser radiation with the cross section of 3×4 cm was focused onto the plane target by a KCl lens with the focal length of 10 cm. The focal spot had the elliptic shape with the dimensions of 0.6 and 0.7 mm. Radiation (Fig. 1) was directed onto the target at the angle of 45° to its surface. Frame-by-frame photography of the natural glow of the plasma jet was made with a VFU-1 high-speed photorecorder. The interval between frames was equal to 11 μs, and the frame exposure time was ~1 μs.

Figure 2 depicts the frame-by-frame record of the natural glow of the plasma jet generated under exposure of the graphite target to the laser pulse. The analysis reveals two significant facts, which call for more detailed consideration:

1. In the process of evolution, the glowing area of the plasma jet acquires a mushroom-like shape with a complex internal structure varying in time.

2. The duration of the plasma jet glow significantly exceeds the duration of the laser pulse at half-maximum. (For example, Kokai et al.[6] reported the formation of carbon nanotubes at laser ablation of the graphite target. They also observed the plasma jet glow with the duration significantly exceeding the duration of laser radiation). In addition, as can be seen from Fig. 2, after 328 μs the glowing column, which connects the cloud with the target, breaks. As a result, the energy stops to come into the cloud (see Fig. 2, the frame corresponding to 343 μs).

Thus, for the further recording period (more than 100 μs, and, in fact, much longer) the glowing cloud is independent of the physical processes proceeding on the target surface.

The mushroom-like shape of the plasma jet is determined, in our opinion, by development of the Richtmyer–Meshkov instability of the plasma–air interface.[5,6] Using numerical simulation, Nichikawa et al.[7] have shown that, for the finite time as short as one likes, the initial distortion of the interface between

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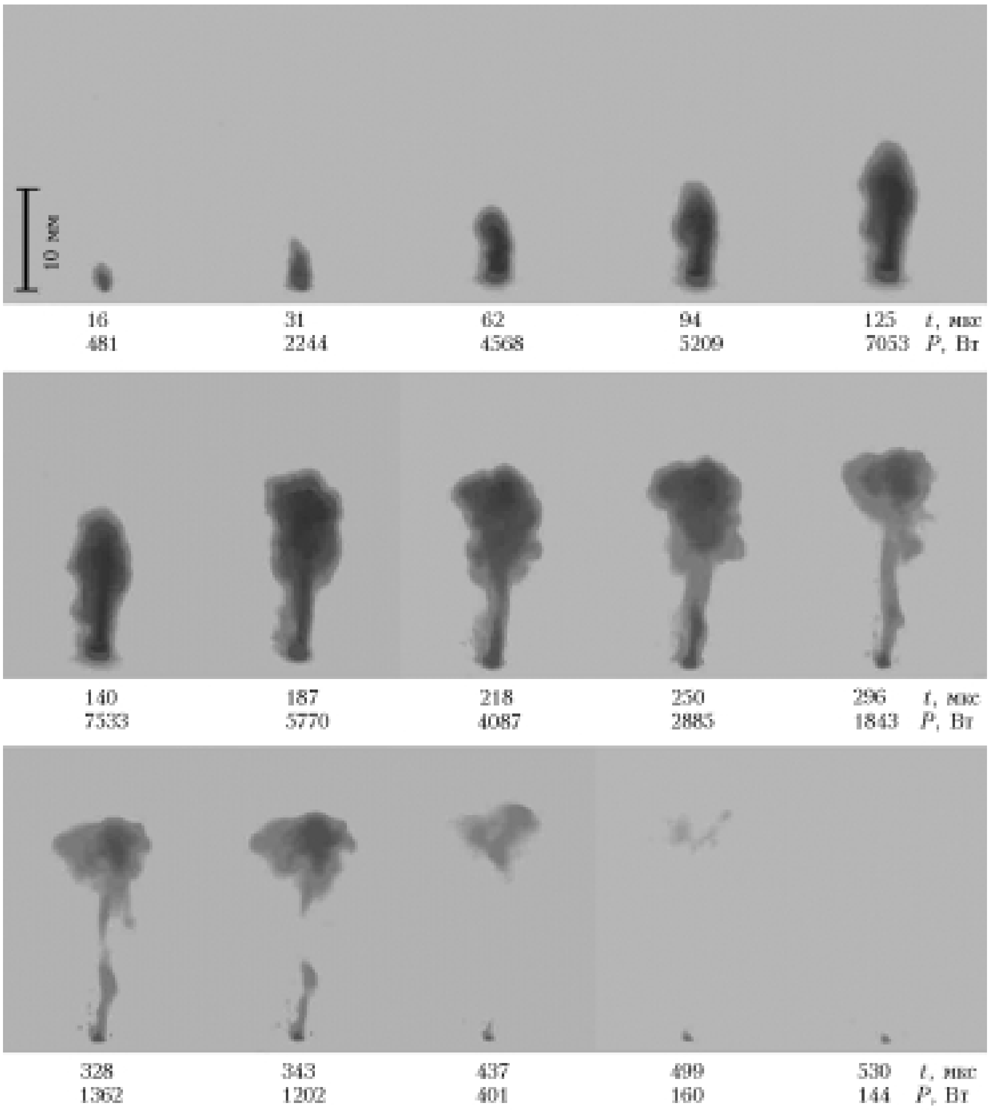

Fig. 2. Natural glow of the plasma jet generated by the long-pulse laser radiation.

fluids with different densities leads to formation and separation of a large-scale eddy of the heavy fluid (in our case, the more dense carbon plasma is the heavy fluid, while air is the light one). For the Richtmyer–Meshkov instability to appear, a necessary condition is the presence of the acceleration directed from the light fluid to the heavy one.

Figure 3 depicts the time dependence of the speed (*a*) and acceleration (*b*) of the glowing area boundary of the plasma jet. It can be seen that the maximal values of acceleration directed toward the plasma jet in our experiments were $(1–2) \cdot 10^6$ m/s^2.

The nonmonotonic character of the curves is likely connected with the portion character of the target ablation: due to high density of the evaporated substance near the surface, its optical transparency decreases, and to increase it, some period is needed for the substance expansion. A presence of high accelerations toward the target in the experiment allows us to say about the development of the Richtmyer–Meshkov instability of the plasma jet-buffer gas interface. At significantly nonlinear stages of this instability, due to a large number of eddies of the heavy fluid and bubbles of the light one, their intense turbulent mixing takes place.[8]

The turbulent mixing in the atmosphere, in our opinion, favors the formation of the nanopowder and hinders deposition of homogeneous films. The observed behavior of the natural glow of the plasma jet evidences this (see Fig. 2).

The long glow of the plasma cloud, the energy income to which stopped since 328 μs (see Fig. 2), is associated, in our opinion, with the Planck glow of hot nanoparticles. To confirm this, we have conducted experiments with the same target exposed for 1.5 μs to the TEA–CO_2 laser radiation with the peak power of $9 \cdot 10^4$ W, pulse duration at half-maximum of 50 ns, and the energy of 20 mJ. The radiation was incident normally onto the target and focused in a spot of 0.2 mm in diameter.

To obtain a slit scan of the plasma jet glow, we used the FER-7 photorecorder with the measurement error for time interval $\Delta t = \pm 0.1$ μs and that for spatial coordinate $\Delta l = \pm 0.05$ mm. To decrease the energy coming into the target at a high power, we used the optical breakdown in the vapor of the target material. The minimal time needed for appearance of the optical breakdown was 40 ns. Its appearance was judged from the appearance of the N_2^+ lines ($\lambda = 470.9$ nm) recorded with the MSD-1 monochromator, FEU-79 PMT, and oscilloscope. The energy coming into the target was determined from these data, and it amounted to 4 mJ.

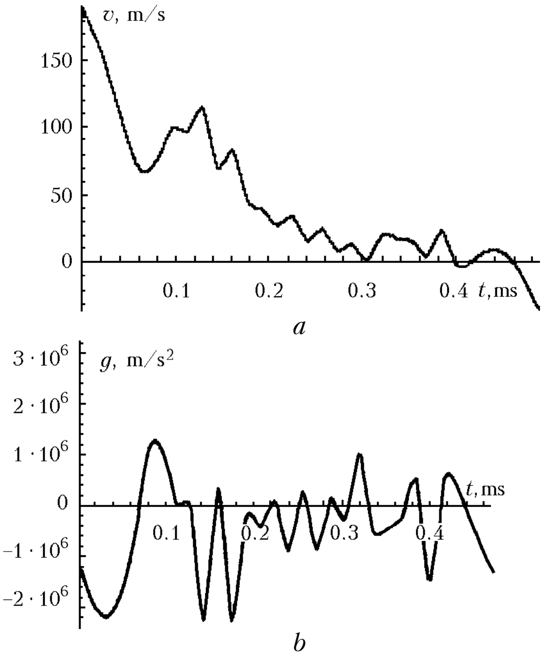

Fig. 3. Acceleration dynamics of the glowing area boundary at the symmetry axis of the plasma jet.

Figure 4 depicts the slit scan of the plasma jet glow. It can be seen that the glow arises initially at the distance of 0.5 mm from the target surface. The plasma boundary is unstable, and the glow duration is virtually equal to the pulse duration. The plasma jet achieves its maximum of 1.28 mm at $t \sim 1$ μs.

Fig. 4. Slit scan of the natural glow of the plasma jet generated by the short-pulse laser radiation.

Knowing the characteristic energy of the laser radiation incoming the target, as well as the focal spot size and the crater depth, we can estimate the mean concentration of carbon atoms in the jet mouth. At $T = 3000$ K it is $n_C = 4.594 \cdot 10^{16}$ cm^{-3}, that is, 245 times lower than the mean concentration of air molecules $n_{O_2} = 1.128 \cdot 10^{19}$ cm^{-3}. This prevents the formation of nanoparticles, and, consequently, the time of plasma glow cannot exceed the pulse duration by the time longer than the lifetime of excited states $\sim 10^{-7}$ s, just which was observed in the experiment.

The situation is quite different, when the target is exposed to a long pulse with the energy of 1.5 J with the power chosen so that the optical breakdown does not occur. In this case, $2.51 \cdot 10^{-8}$ kg of carbon comes into the plasma jet, and this ensures the mean concentration of carbon atoms in the jet mouth $n_C = (1.49–1.72) \cdot 10^{19}$ cm^{-3}. Since $n_C > n_{O_2}$, the expansion of the plasma jet into the buffer gas (air) leads to the efficient formation of nanoparticles, which determine the long glow of the plasma jet observed in our experiments.

Thus, the shape and the structure of the plasma jet glow are determined, in our opinion, by the development of the Richtmyer–Meshkov instability, while the duration of the glow is determined by the presence of incandescent ultradisperse particles in the plasma jet.

Acknowledgments

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