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Properties of the interaction of laser radiation with a gaseous dust medium

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***Abstract*. It is found that upon irradiation of a mixture of the atmospheric air and carbon particles of size 30 – 300 μm at a concentration of $\sim 10^2$ cm^{-3} by a cw CO_2 laser, the active combustion of particles in the mixture appears when the radiation intensity in the focal region achieves $\sim 10^3$ W cm^{-2}. The dependences of the threshold radiation intensity for the evaporation of particles on their radius are obtained for a gaseous dust medium in the form of a free vertical jet of spherical aluminium and carbon microparticles in nitrogen. It is shown that particles of size $\sim$ 10 μm can be completely evaporated in a focused cw laser beam of power $\sim 10^2$ W.**

Keywords: *gaseous dust medium, radiation intensity, evaporation.*

1. Introduction

One of the methods for producing films on surfaces and applying coatings is the laser method based on the evaporation of a material during irradiation of solid targets by high-power pulsed lasers [1]. This method is relatively simple and can be use to evaporate almost any materials. Its main disadvantage is the possibility of the deposition of microfragments of the evaporated material on the surface in the form of solidified drops or particles of arbitrary shape, which impairs the film quality. The film quality can be improved by evaporating additionally microfragments escaping from the surface in crossed laser beams [1, 2]. Note that the efficiency of the method of crossed beams depends on the interaction time of particles with the additional radiation field and can be insufficient in the case of a large initial dispersion velocity of particles initiated by main high-power laser radiation.

The aim of this paper is to study the possibility of efficient evaporation of a material under conditions when the initial object irradiated by a laser is a gaseous dust medium containing micron particles with a controllable translation motion velocity. Under these conditions, due to weak heat exchange between particles and surrounding gas, the particles can be completely evaporated in the radiation field of even a relatively low-power laser.

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2. Experimental

We used in our experiments a CO_2 laser operating either in the cw or quasi-cw regime with controllable power and pulse duration. The horizontally oriented laser beam was focused into a region containing a mixture of particles with the atmospheric air at the normal pressure or with argon or nitrogen at controllable pressure. Carbon particles of size 30 – 300 μm and CdS and Al particles of size $\sim$ 10 μm were used. A gaseous dust medium was produced mechanically. In one case, it was a quasi-stationary gaseous dust cloud, while in another – a vertical jet of particles in gas directed downward. Experiments were mainly performed with the jet of particles allowing the control of the initial parameters of the medium.

3. Measurement and calculation results and discussion

3.1 Irradiation of a carbon microparticle jet in the atmospheric air by a cw laser

Figure 1 presents the power dependences of the parameter $\eta = (W - W_1)/W$, characterising the decrease in the power W due to the absorption and scattering of radiation by particles in the medium (W_1 is the laser power after the propagation of radiation through the jet). These dependences were obtained for distances h from the jet initiation site to the lens focus equal to 2 and 4 cm. The particle flow rate was 21.7 mg s^{-1} and the jet diameter was 14 and 16 mm for $h = 2$ and 4 cm, respectively. The divergence of laser radiation was 2 mrad, the laser beam diameter was 6 cm, and the focal distance of the lens was 30 cm. The value of η begins to increase when the threshold power W_{th} is achieved at which particles begin to burn in the region of their interaction with laser radiation. The threshold power depends on h, being $\sim$ 10 W for $h = 2$ cm, which, taking into account the attenuation of radiation, corresponds to the threshold radiation intensity in the focal spot $\sim 8 \times 10^2$ W cm^{-2}. For $W > W_{th}$, the bright emission in the interaction region acquired nearly symmetric shape with respect to the beam axis and focal plane. The maximal longitudinal size of the emission region was approximately

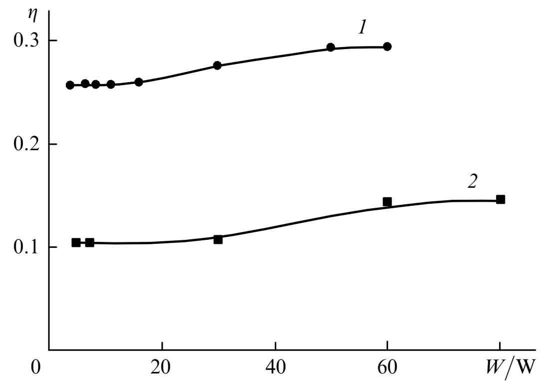

Figure 1. Dependences of the parameter η on the power W of a cw CO_2 laser irradiating a jet of carbon microparticles of size 30 – 300 μm in the atmospheric air for $h = 2$ (*1*) and 4 cm (*2*).

equal to the jet diameter. For brevity, this emission will be referred to as the discharge. Note that the value $W_{th} \approx 10$ W is consistent with the average threshold power ~ 7 W of the stable ignition of dispersed hard coal particles of size ~ 100 μm in the nitrogen – oxygen mixture jet ($h = 2$ cm, and $[O_2] = [N_2]$) irradiated by a repetitively pulsed Nd : YAG laser [3].

The decrease in η with increasing h (Fig. 1) is explained by the increase in the jet transparency due to the decrease in the concentration of particles in it. The average concentration of particles in the given section of the jet in the case of the homogeneous initial size distribution of particles can be estimated from the expression $\langle N \rangle = G/(\langle m \rangle \langle \upsilon \rangle s)$, where G is the total flow rate of all particles; $\langle m \rangle$ is the mass of particles averaged over their sizes; $\langle \upsilon \rangle$ is the average velocity of particles depending on $\langle m \rangle$ and h; and $s = s(h)$ is the cross-sectional area of the jet. By neglecting collisions between spherical particles during their motion and averaging over three radii of particles $r = 30$, 70, and 120 μm for $h = 2$ and 4 cm and $G = 21.7$ mg s^{-1}, we obtain $\langle N \rangle \approx 50$ and ~ 30 cm^{-3}, respectively. Note that $\langle N \rangle$ decreases with increasing h mainly due to the increase in the average velocity $\langle \upsilon \rangle$, because the cross-sectional area s of the jet weakly depends on the distance h. As for the increase in η for $W > W_{th}$ compared to η for $W < W_{th}$, it probably occurs due to the additional absorption and scattering of radiation by the combustion products of particles in the air oxygen, which compensate the possible decrease in the initial concentration of particles during their interaction with radiation.

It is obvious that heated carbon particles can acquire a noticeable positive charge due to thermionic emission. Let us estimate this charge under our conditions. Note preliminarily that for radiation intensities $\sim 10^3$ W cm^{-2}, the avalanche multiplication of electrons can be neglected [4]. At the plasma temperature of the order of the gas temperature ($\sim 10^3$ K) and ionisation potential ~ 10 eV, the equilibrium electron concentration can be also neglected. Therefore, we will assume that electrons in the discharge are thermoelectrons at the concentration $n_e \sim q\langle N \rangle$, where q is a particle charge in elementary charge units. To determine the electron concentration in the discharge by the thermionic emission current density [5], we assume that the current transfer by electrons emitted from the surface of particles occurs at the electron velocity corresponding to the gas temperature $\sim 10^3$ K. By using then experimental results from [3], according to which the maximum temperature of the particle surface achieves 2000 K at the average temperature 1700 K, taking into account that $\langle N \rangle \sim 10^2$ cm^{-3}, we obtain the maximum and average charges $q \sim 10^6$ and $\sim 10^4$, respectively. By considering the discharge under study as the approximate analogue of a dust plasma, the estimates of q presented above can be compared with experimental data [6] obtained for a dust plasma containing positively charged particles, according to which the charge of particles in the plasma can achieve $\sim 10^4$.

Based on the results obtained in [6] and estimates presented above, it is reasonable to assume that the presence of macroscopic particles in the discharge, which have such a large charge and concentration $\sim 10^2$ cm^{-3}, should affect the flow of current through the discharge. The current was measured with metal electrodes of diameter 2 mm with flat ends. The interelectrode gap of length 3 mm was oriented perpendicular to the jet axis and the laser beam, the lens focus being located in the central part of the gap. When a dc voltage ~ 100 V was applied to the electrodes in the presence of the discharge, the average quasi-stationary current flowing through the gap was 50 mA, corresponding to the average conduction of the gap equal to $5 \times 10^{-3} \Omega^{-1}$ cm^{-1}. This conduction noticeably exceeds the electron conduction of the plasma in a constant field calculated by expressions from [4]. For example, even for the maximal estimated electron concentration $n_e \sim q\langle N \rangle \sim 10^8$ cm^{-3}, the electron conduction does not exceed 10^{-7} Ω^{-1} cm^{-1}. We assume that the obtained conduction of $5 \times 10^{-3} \Omega^{-1}$ cm^{-1} is related to one or several current filaments formed by carbon microparticles of diameter $\sim 0.1 - 1$ mm conducting current, which we observed in our experiments. Note that the quasi-stationary type of the current is probably explained by the periodic destruction of filaments by particles entering the gap. Note also that, when the voltage ~ 100 V is applied across the gap in the absence of the discharge, no filaments are formed and the electric breakdown of the gap filled with particles of this size and concentration does not occur.

Consider a tentative mechanism of formation of an individual filament, by neglecting the role of induced polarisation during the interaction of particles. A positively charged carbon microparticle located near the cathode settles down on it under the action of the attraction force, is retained on it due to sintering, and becomes a component of the negatively charged cathode, forming an asperity on its surface. On this asperity the electric field of the interelectrode gap is concentrated. As a result, a charged microparticle located most closely to the asperity settles down on it rather than in its plane vicinity. The deposition process repeats, the attraction force from the anode side orienting this process by forming a filament cathode 'growing' to the anode side. The locking of the interelectrode gap and formation of the filament can be treated as the result of the action of the same force from the anode side, resulting in the expansion of the filament cathode. It should be emphasised that thus mechanism is only hypothetical, and its refinement or the establishment of another mechanism requires special investigations.

3.2 Threshold radiation intensity for evaporating particles in a jet

Let us estimate the threshold laser radiation intensity required to evaporate particles in a jet. Preliminarily estimates show that heat conduction loss can be neglected compared to the radiative loss. By neglecting also the energy consumption for heating and melting of particles compared to the energy spent for evaporation, we can write the energy balance equation for a particle in the form

$$mQ = SA\delta t(\alpha I - \sigma T^4), \quad (1)$$

where S is the particle surface area; A is the absorption capacity of the particle material; Q is the specific evaporation energy; I is the threshold laser energy required to evaporate the particle; T is the evaporation temperature; σ is the Stefan – Boltzmann constant; and $\delta t = 2\langle R \rangle / v$ is the flight time of the particle in the interaction region with the average radius $\langle R \rangle$. The coefficient $\alpha = 1/2$ in the right-hand side of expression (1) approximately takes into account the absorption of radiation only by half the particle surface. Let us represent the interaction region in the form of a straight cylinder with the focal plane in its centre. The cylinder generatrix is oriented along the laser beam and its length is equal to the jet diameter. Such a choice of the interaction region shape is based on the visible longitudinal size of the discharge. Let us define the average radius of the interaction region as $\langle R \rangle = (R + R_f)/2$, where $R \approx Dd/(4f) + R_f$; D is the laser beam diameter; d is the jet diameter; $R_f = \gamma f$ is the focal spot radius; γ is the radiation divergence; and f is the focal distance of the lens.

Figure 2 presents the calculated dependences of the threshold evaporation intensity I on the radius r of aluminium and carbon particles forming a vertical jet in nitrogen at the atmospheric pressure. The calculations were performed for $D = 9$ cm, $d = 1$ cm, $f = 40$ cm, and $\gamma =$ 2 mrad, which gives $\langle R \rangle \approx 1$ mm, for $h = 2$ cm, corresponding to the change in δt from ~ 300 to 4 ms when r is increased from 5 to 100 μm. We used in calculations the parameters $A = 0.05$ and 1, $Q = 10.9$ and 60 kJ g^{-1}, $T = 2450$ and 3600 °C for aluminium and carbon, respectively. The increase of A for aluminium upon heating was neglected. Note that due to a weak pressure dependence of the dynamic viscosity of nitrogen [5], the results of calculations of the dependences of I on r at other pressures weakly differ from the results obtained at the atmospheric pressure.

One can see from the curves presented in Fig. 2 that, depending on r, the ratio I_{Al}/I_C can be either greater or smaller than unity (hereafter, the quantities with subscripts Al and C are related to aluminium and carbon, respectively). This is related to the different contributions of the radiative loss to the energy balance of particles depending on their radius. It is obvious that the radiative losses can be neglected beginning from certain values of r due to their weak dependence on r compared to the evaporation laser radiation energy loss. Then, taking into account that masses and velocities of aluminium and carbon particles are approximately equal, we obtain from (1) that $I_{Al}/I_C \approx (Q_{Al}/Q_C)(A_C/A_{Al}) > 1$. As the particle radius r decreases, when the contribution of the radiative loss increases, the ratio I_{Al}/I_C can be most simply found when both types of losses are equal for each of the materials: $I_{Al}/I_C \approx (T_{Al}/T_C)^4 < 1$. According to calculations, the radiative losses are negligible beginning from $r > 10 - 20$ μm, and for $r \leqslant 10$ μm, they become comparable with the evaporation loss, which is reflected in Fig. 2 in accordance with the estimated presented above.

Despite the large enough average radius ($\langle R \rangle \approx 1$ mm) of the interaction region used in calculations, the laser radiation power $\sim I\pi\langle R \rangle^2$ required for evaporation of small particles proves to be low. Thus, for $r = 10$ μm, this power for aluminium and carbon is ~ 170 and ~ 100 W, respectively. We can say that the efficient evaporation of such particles can occur in the radiation field of a comparatively low-power cw laser. As the particle radius increases, the laser power at the fixed $\langle R \rangle$ should be increased, and the choice of a proper laser is determined by the particle size. This can be either cw of repetitively pulsed laser with a moderate average output power.

Note that the authors of [7] studied the change in the concentration of Al particles of size ~ 1 μm due to evaporation in argon at the atmospheric pressure upon irradiation by several 10-J, 0.5-μs pulses from a CO_2 laser. The radiation intensity depended on the beam formation method and was varied from the maximum value 6×10^7 W cm^{-2} down to $\sim 10^6$ and 3×10^5 W cm^{-2}. Calculations by using (1) for $\delta t = 0.5$ μs show that the radiation intensity (3.5×10^7 W cm^{-2}) required for evaporation of these particles corresponds to the maximum intensity. It is possible that particles were evaporated at lower intensities due to heating in the optical discharge plasma produced in experiments [7].

The possibility of the efficient evaporation of particles of radius 5 – 10 μm upon irradiation of a gaseous dust medium by a comparatively low-power laser for producing films on a surface was verified experimentally. Substrates were placed above the geometrical focus of the lens at a distance from it

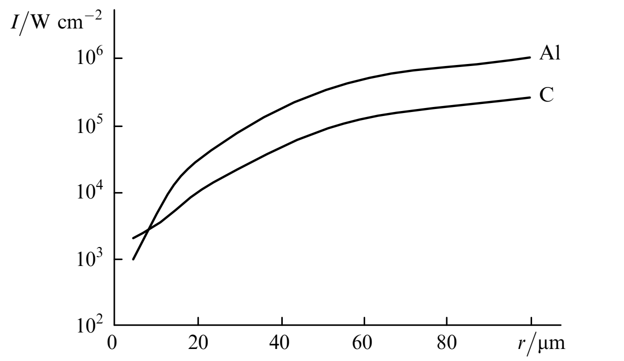

Figure 2. Calculated dependences of the evaporation threshold radiation intensity I on the radius r of aluminium and carbon particles.

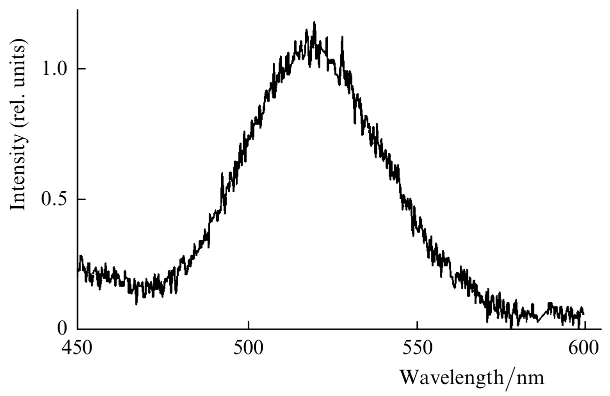

Figure 3. Luminescence spectrum of CdS films.

precluding the contact of the substrate with the visible boundary of the discharge. A jet containing CdS or Al particles of this size was irradiated by a $\sim$ 500-W quasi-cw CO_2 laser ($f = 40$ cm, $D = 9$ cm) in the argon or nitrogen atmosphere at a pressure of 100 – 300 Torr. Figure 3 shows the typical luminescence spectrum of prepared amorphous CdS films excited by a 337-nm nitrogen laser. Films of area ~ 1 cm^2 were prepared after irradiation by 5 – 10 50-ms pulses from a CO_2 laser. The preliminary visualisation of the surfaces of films demonstrated that they did not contain microfragments of size exceeding 1 μm.

4. Conclusions

Laser evaporation of particles from a gaseous dust medium is promising for preparing films and coatings from various materials without using the expensive equipment. We have shown by the example of an air-carbon medium irradiated by a cw CO_2 laser that the medium can be ignited at a comparatively low threshold radiation intensity. The investigation of the conditions for initiating combustion in various gaseous dust media and of the energy balance in the medium can be of interest for laser nanotechnologies, the physics of dust plasmas, and a number of other applications.

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