Spectral diagnostics of the plasma plume produced in intense metal evaporation under laser radiation

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(Submitted August 2, 1972)
Zh. Eksp. Teor. Fiz. **64**, 825-832 (March 1973)

Results are presented of a spectral diagnostics of the low-temperature plasma of a plume produced by intense millisecond optical radiation incident on metallic targets. Distribution of the temperature along the plume is measured in a broad range of intensities $I \sim (0.3-3)\,10^7$ W/cm^2 and atmospheric pressures $p = (0.1-5)$ atm. Estimates of the electron density N_e are obtained. At a distance from the metal surface corresponding to a shock wave at rest with respect to the target[1,2], temperature maxima with values $T \approx (1.5-2)$ eV are observed. The vapor temperature at a distance on the order of the size of the irradiated spot is several times greater than the critical temperatures of the metals investigated. This is a direct proof that laser radiation of an intensity $I \sim 10^7$ W/cm^2 heats the dense vapor near the target.

1. INTRODUCTION

We investigate in this paper the parameters (the temperature T and the electron density N_e) of a plasma plume produced when powerful optical radiation of millisecond duration acts on a metal (bismuth or aluminum). The values of T and N_e in different regions of the plume are governed by its gasdynamics structure. It is shown in[1,2] that the time of establishment of the gasdynamic picture, in the case of developed evaporation of metals, is much shorter than the time of action of the light ($\tau \sim 10^{-3}$ sec). The picture of vapor flow is determined then by the intensity I of the radiation incident on the metal and by the pressure p of the atmosphere in which the evaporation takes place[2]. In the considered range of intensities ($I \sim 10^7$ W/cm^2) the experimentally observed pictures of evaporation at $p \leq 1$ atm and $p > 1$ atm differ qualitatively from each other$^{1)}$.

At $p \leq 1$ atm, in the case of planar evaporation ($d \gg h$, where d is the diameter of the incident-radiation spot on the target and h is the depth of the crater produced by the radiation), stationary flow of vapor is established, together with a shock wave (SW) that is immobile relative to the target[1]. In accordance with this picture, the temperature on the plume axis should vary little in a region with dimensions ~d near the target. At distances $R > d$ from the target one should expect the temperature to decrease in accord with the Poisson adiabat $T \propto R^{2(1-\gamma)}$ (γ is the adiabatic exponent). For a monatomic gas $\gamma = 5/3$ and $T \propto R^{-4/3}$. T should increase jumpwise on the front of a strong shock wave.

When bismuth or aluminum evaporates in a dense atmosphere ($p > 1$ atm), the optical thickness of the plasma behind the SW front is appreciably larger than in the case when $p \leq 1$ atm. As a result, the intensity of the radiation passing through the plume decreases strongly and becomes close to the threshold value for the developed evaporation. The evaporation ceases almost completely, a fact manifest in experiment as a detachment of the plume at 1-2 cm from the target (experiments with bismuth[2,3]). The picture is no longer stationary. In this evaporation regime (at $p > 1$ atm) one should expect low time-averaged temperatures of the vapor in the near-surface region and a growth of T far from the target in the region of glow of the plume.

The temperature was measured in the present study by determining the emission spectra of the plume. Similar spectral investigations were undertaken earlier with lasers having emission energies on the order of hundreds of Joules ($\tau \sim 10^{-3}$ sec). A review of these investigations can be found in[4]. Owing to the low emission energy, sharp focusing of the beam was necessary ($d < 1$ mm) in order to realize on a metallic target light fluxes that exceed the threshold for developed evaporation ($I_{thr} \sim 10^6-10^7$ W/cm^2). In these investigations they did not observe the sharp maxima which we found on the plume temperature profile in the present study (see Sec. 3). Nor was evaporation in a dense atmosphere, first obtained in[3], investigated. There are many possible reasons for the discrepancy between the results of[4] and our results. First, in the experiments of[4] the depth of the crater greatly exceeded the spot diameter d. Therefore the gasdynamic structure of the plume differed strongly from the structure in our case. Second, since the scale of the picture is determined by the quantity d[1], in experiments with sharp focusing (and hence small d) the plume dimensions are small, making its diagnostics difficult. In addition, the laser pulse in[4] had a "spike" structure with complete modulation, as a result of which the SW was not stationary, and was produced only during the time of individual lasing spikes$^{2)}$.

In our experiments we used a laser setup with emission energy up to 10 kJ in a pulse of duration $\tau \approx 0.8$ msec[5]. The large emission energy has made it possible to perform experiments with radiation spots of diameter $d \sim 1$ cm. The generation pulse was quasistationary, the depth of the "spike" modulation did not exceed 30%. We used long-focus lenses with $f = 1$ m, with caustics having cylindrical sections of approximately 10 cm length. The large dimensions of the plume (~10 cm), the satisfaction of the condition of planar evaporation ($h \ll d$), and the quasistationary character of the radiation pulse made it possible to trace the variation of the temperature along the plume and to find that it agreed with the distribution that follows from[1,2], and also to establish the fact that the vapor is heated by the laser radiation in a region $R \lesssim d$ near the target.

2. MEASUREMENT PROCEDURE

The plume temperature was determined by the Ornstein method from the intensity ratio I_1/I_2 of two spectral lines with known transition probabilities A_1 and A_2 and frequencies ν_1 and ν_2 (see for example,[6], Sec. 13.1). We used for the calculations the ratio

$$\lg(I_1/I_2) = \lg(A_1g_1/A_2g_2) + \lg(\nu_1/\nu_2) - 5040(E_1 - E_2)/T^\circ, \qquad (1)$$

where E_1, E_2 and g_1, g_2 are the energy (in electron volts) and the statistical weights of the upper levels of the first and second lines, respectively. To measure the temperature (and also the electron density), we chose the lines of the ions of highest (second) multiplicity observed in the spectra of the plume. In the case of aluminum, the numerical values of the factors Ag of the levels of these ions, which enter in (1), were taken from[7].

The employed method is valid if local thermodynamic equilibrium, i.e., a Boltzmann distribution of the particles over the levels, exists in the plasma. Estimates by the formulas of[8] show that at the experimentally-realized plume-plasma parameters[1,2] ($T \sim 1-2$ eV, $N_e \sim 10^{17}-10^{18}$ cm^{-3}), the probability of exciting ions of second multiplicity by collisions with electrons is $N_e\langle v\sigma\rangle \sim 10^{11}-10^{12}$ sec^{-1} (v is the electron velocity and σ is the excitation cross section), whereas the total probability G of the decay of the excited levels of these ions is $\sim 10^8-10^9$ sec^{-1}. The condition $N_e\langle v\sigma\rangle \gg G$ is satisfied with a large margin, so that we have a Boltzmann distribution of the particles over the levels (and consequently also local thermodynamic equilibrium). The excitation temperature determined by the Ornstein method is in this case the electron temperature. However, according to the estimates (see[6], Sec. 6.9), the time of equalization of the temperature of the electrons and ions is negligibly small at the indicated plasma parameters and amounts to $10^{-9}-10^{-8}$ sec. Therefore the experimentally measured temperature is also the ion temperature.

The spectral emission lines used for the diagnostics should not experience self-inversion as a result of absorption of light in the plume. The absence of self-inversion was verified by comparing the observed and theoretically-calculated intensities of the multiplets chosen for the diagnostics (for example, in the case of the Al-III ions, the calculated ratio of the line intensities in the doublets used to find the plume temperature (see below) was $\frac{1}{2}$). The comparison has demonstrated the absence of self-inversion.

Under experimental conditions, when the use of the Ornstein method was impossible, the plume temperature was estimated by using the calculated degree of equilibrium ionization (see[9], Sec. 106).

The electron density was measured by determining the Stark broadening of the spectral lines of doubly-ionized atoms ([6], Sec. 14.2). The values of the Stark constants for the lines Al-III were taken from[10], and the constants for the lines Bi-III were calculated at our request by E. A. Yukov.

The plume-plasma emission spectrum was registered with an ISP-51 spectrograph equipped with a camera with a 270 mm lens. The longitudinal axis of the plume image was made congruent with the spectrograph slit, so that we could obtain in a single experiment the spectrum over the entire length of the plume. The spectrograph received the emission from a central region of the plume, of thickness ~0.1 mm, situated in the horizontal plane along the laser-beam axis. To determine the relative intensity of the spectral lines we used a standard heterochromic photometry procedure. As a source with a known spectral intensity distribution we used the standard tungsten ribbon lamp of the SR-3 spectrum radiometer.

The metallic targets were vaporated in an atmosphere of helium, whose spectrum was not excited in the temperature range employed in our experiments.

The obtained spectra are integrated over the time of action of the radiation pulse on the metal. The intensity in the pulse however, changed little over a greater portion of the time (τ = 0.8 msec at the level of half the maximum intensity, at a total duration ~1.5 msec). The doubly-ionized ion lines used to measure the plasma temperature were excited in the spectra during the middle of the pulse, when the intensity of the laser radiation was maximal and changed little. In addition, the developed evaporation, by virtue of the presence of an irradiation threshold, was observed only in the central part of the generation pulse. Therefore the time-averaged values of the temperature obtained in our work do not differ strongly from the true values.

3. MEASUREMENTS RESULTS

Typical spectrograms obtained in the experiment are shown in Fig. 1. The qualitative analysis of the spectrum makes it possible to establish the character of variation of the temperature along the plasma plume.

1. We consider first the spectrograms pertaining to Bi and Al evaporated in a helium atmosphere with $p \leq 1$ atm. Near the target surface one can see an intense continuous spectrum, against the background of which there appear strongly broadened lines of neutral and of singly and doubly ionized atoms. This is evidence of a high density of the particles in the indicated region, in comparison with other parts of the plume. The character of variation of the intensity of the spectral lines along the plume is different for atoms with different ionization multiplicity. The Al-I and Bi-I lines are seen over the entire length of the plume. Their intensity decreases slowly over distances on the order of 10 cm.3) This means that the Al-I and Bi-I lines are emitted by the colder peripheral parts of the plume, which are in contact with the external gas. The resonance lines are self-inverted as a result of absorption in the internal colder regions of the plume. The lines Al-II and Bi-II are present in the spectra over a length 6–7 cm from the surface of the target. Over this length, their glow also decreases monotonically. The line profiles of the Bi-III ions (4327.8, 4560.84, 5079.5 Å) and of Al-III (4512.54, 4529.18, 5696.47, 5722.65 Å) differ strongly along the plume from the described character of the intensity variation of the lines Al-I, Al-II, Bi-I, and Bi-II. The lines of the doubly charged ions are emitted by the internal plume region close to the laser-beam axis, where the gasdynamic picture is not distorted by external factors. Therefore the shapes of the Al-III and Bi-III spectra describe adequately the gasdynamic structure of the plume[1]. With the increasing distance R from the target surface, the intensity of these lines decreases rapidly almost to zero, in accord with the expected decrease of the temperature and density in the region of the adiabatic ex-

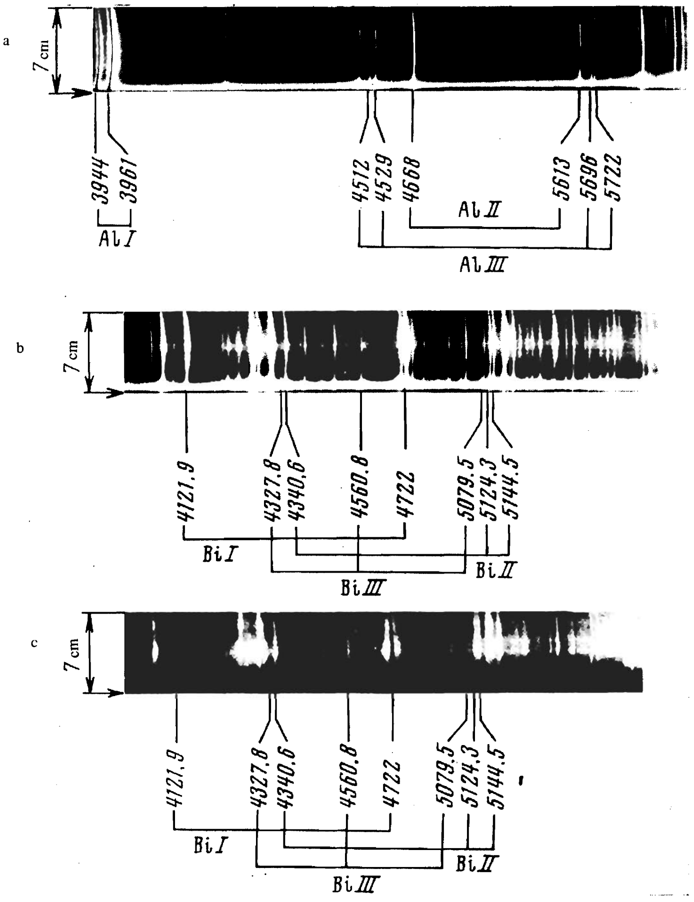

FIG. 1. Spectra of plumes: a–aluminum (experimental conditions: p = 0.1 atm, I = 1.05 X 10^7 W/cm^2, d = 0.8 cm), b–bismuth (p = 1 atm, d = 0.8 cm, I = 0.84 X 10^7 W/cm^2), c–bismuth (p = 5 atm, I = 10^7 W/cm^2, d = 0.8 cm). The target position on all photographs is indicated by an arrow, and the laser bean is incident downward. The wavelengths are in Å.

pansion of the vapor. Further, at a certain distance R_0 corresponding to the position of the SW front, the emission of these lines becomes abruptly stronger. Also increased is the intensity of the continuous spectrum, since a jump in the vapor density takes place on the SW front. With further increase of R, the intensity of the lines Al-III and Bi-III again decreases, and the decrease along the length is faster than in the lines Al-II and Bi-II. The indicated lines Bi-III and Al-III were therefore chosen to measure the plume temperature.

Quantitative experimental data for aluminum, obtained by reducing the Al-III spectra, are plotted in Fig. 2 and pertain to the pressures p = 0.5 and p = 0.1 atm. The absolute measurement error of the curves of Fig. 2 is due mainly to the error in the photometry and amounts to 20%. The relative error is not higher than 15%. Figure 2 contains no experimental points corresponding to small R ($R \lesssim d$), since our procedure cannot be used in this region, owing to the strong broadening and mutual overlap of the lines chosen for the reduction and to the presence of a strong continuous background near the target.

The curves in Fig. 2 show clearly the singularities of the temperature variation along the plume, as obtained above from the qualitative analysis of the spectra, and are in good agreement with the gasdynamic picture of vapor flow[1]. There are, however, certain deviations from this picture. According to[1], the increase of T on the SW front should occur jumpwise. Experiment, however, shows a rather smooth increase of the temperature, and a sharp burst occurs only at small values of p (p = 0.1 atm). The absence of jumps of T on the plots of Fig. 2 is due to the fact that when the plume is photographed in a transverse direction, the thickness of the glowing plasma increases smoothly with increasing distance from the target, since the surface of the shock-wave front is spherical in shape. For the same reason, it is meaningless to compare quantitatively the experimentally obtained decrease of the temperature (and of the density N_e, see below) with distance in the region of adiabatic expansion, on the one hand, with the theoretical value $T \propto R^{-4/3}$, on the other. The observed decrease of T behind the SW front can be attributed to the cooling of the vapor as a result of loss of energy to recombination radiation. Estimates show that an appreciable decrease in the internal energy of the vapor behind the SW front, due to

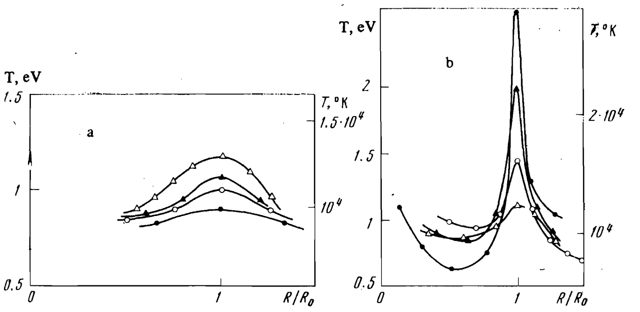

FIG. 2. Dependence of the plasma temperature T on the plume axis on the distance from the target surface at different intensities I of the laser radiation. The abscissas are given in relative units R/R_0 (R_0 is the SW dimension at the given p and I). The absolute values of R_0 are taken from Fig. 3. The experimental conditions are: a) p = 0.5 atm, I equals 5 X 10^6 (●), 6.5 X 10^{-6} (O), 9.7 X 10^6 (▲), and 1.15 X 10^7 (Δ) W/cm^2; b) p = 0.1 atm, I equals 6.3 X 10^6 (Δ), 8.8 X 10^6(O), 1.05 X 10^7 (▲), and 1.28 X 10^7 (●) W/cm^2.

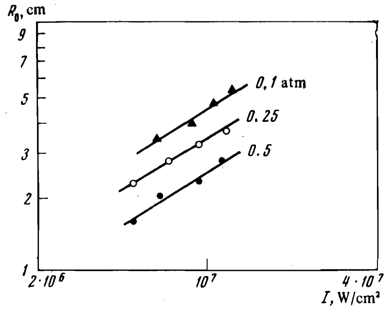

FIG. 3. Dependence of the dimension R_0 (in cm) of the shock wave on the intensity I at the different values of p (aluminum).

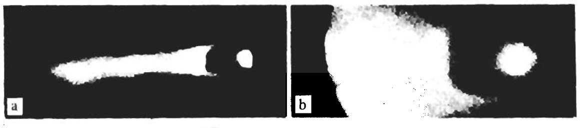

FIG. 4. Streak photograph of a plume from aluminum at $I = 6 \times 10^6$ W/cm²: a) p = 0.5 atm, b) p = 0.1 atm. Exposure time 16 msec, the frames correspond to the middle section of the applied pulse. Photograph a shows a rather strong distortion of the shock wave front (deviation from sphericity), due to the directivity of the vapor escape. There are no distortions in photograph b. (The laser beam is incident from left to right.)

radiation, occurs within a time 10^{-5}–10^{-4} sec. These values of the time (at a vapor velocity 10^5–10^4 cm/sec) correspond to distances at which a noticeable decrease of the temperature should take place behind the SW front, on an order of several centimeters, as is indeed observed in the experiment (Fig. 2).

Before we proceed to a discussion of the measured temperature in a bismuth plume, we note two other results that follows from Fig. 2.

From the position of the temperature maximum one can determine the dependence of the dimension of the shock wave on the intensity I (see Fig. 3), namely $R_0 \propto I^{1/2}$. A similar result was obtained experimentally in a study of the gasdynamics of bismuth vapor[2]. At the same time, in aluminum the dependence of R_0 on the pressure p in the interval p = (0.1–0.5) atm is weaker in Fig. 3 than that observed in experiments with bismuth[1] or that calculated theoretically for a spherical shock wave[1], namely $R_0 \propto p^{-1/2}$. When p is increased from 0.1 to 0.5 atm, the dimension R_0 decreases only by a factor 1.7. This is caused by the observed deformation of the shape of the SW front for Al (see Fig. 4), owing to the directed escape of vapor from the deeper crater on the target (in comparison with bismuth), when the dimension R_0 is small and is comparable with the diameter of the irradiated zone. For the same reason, the temperature profiles on Fig. 2a are smoother than in Fig. 2b.

The second result pertains to the dependence of T on the SW front on the intensity I (Fig. 5). At p = 0.1 atm, we have $T(R_0) \propto I$. In[2], where the vapor velocity was measured, a similar result was obtained for bismuth and explained theoretically. Since the ratio of $T(R_0)$ for a strong shock wave to the temperature near the target T(d) is constant at 1.25[1], the same relation holds also for T(d). At higher pressures, p = 0.25–0.5 atm, the observed relation $T(R_0) = F(I)$ is weaker than linear. This is apparently due to the contribution of the directed stream of vapor from the crater on the target to the shock-wave formation (Fig. 4).

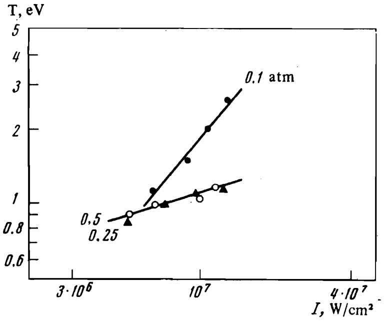

Fig. 5. Plot of the temperature T on the shock wave front against the irradiation intensity I at different external pressures in the case of aluminum: 0.1 (●), 0.5 (○), and 0.25 (▲) atm.

In the experiments with bismuth, the temperature profiles along the plume were not determined. We had no data on the transition probabilities corresponding to the Bi-III lines observed at $p \leq 1$ atm. In addition, the dependences of the SW dimension on the pressure p and on the intensity I, and also the dependence of the temperature on the SW front on I, which were obtained above for aluminum from the temperature profile, were determined for the case of bismuth earlier by another method[2]. We confine ourselves here to an estimate of the temperature of the bismuth plume on the basis of the experiment data and a calculation of the degree of equilibrium ionization α_{III} ([9], Sec. 106). According to calculations at p = (0.1–1) atm, the Bi-III lines appear ($\alpha_{III} \sim 10^{-2}$–$10^{-3}$) at $T \approx (1.1-1.3) \times 10^4$°K. The intensity incident on the target and corresponding to the appearance of these lines in the experiment is $I = (6-8) \times 10^6$ W/cm². Assuming a linear relation $T \propto I$[2], we obtain at $I = 1.5 \times 10^7$ W/cm² the value $T = 2.4 \times 10^4$°K, which is in good agreement with the values obtained in[2] by measuring the vapor velocity (namely, $T = 2.5 \times 10^4$°K at $I = 2 \times 10^7$ W/cm²). The foregoing estimates pertain to the temperature on the shock wave front near the target surface.

2. We now analyze the spectra in the case of high external pressure p > 1 atm. As already noted in Sec. 1, the gasdynamic structure of the plume is significantly altered in this case. This change becomes manifest also in the spectra. In particular, the intensity of the continuous background is sharply increased (see Fig. 1c), meaning an increase in the plasma density. Greatest interest at high p attaches to a study of the plume of bismuth (see Sec. 1). The spectrum of such a plume at p = 5 atm is shown in Fig. 1c. The photograph does not show the glow of the target and of a region 1–2 cm next to the target, thus indicating that the vapor temperature in this zone is exceedingly low. This form of the spectrum is due to the strong screening of the target against the incident radiation (see Sec. 1) and to detachment of the plasma from the target[2,3]. At distances larger than 1–2 cm, there appear in the spectrum, among others, also strongly broadened and overlapping Bi-III lines whose intensity increases and reaches a maximum at a distance 4–5 cm, and then decreases slowly. The character of variation of T along the plume, the pressure in which is equal to the external pressure, is analogous to the change of the intensity of the Bi-III lines. The temperature in the "hot" part of the plume, estimated from the experimental data and from a calculation of the degree of equilibrium ionization (see above), lies near $T = 2 \times 10^4$°K ($I \sim 10^7$ W/cm², p = 5 atm).

To obtain an independent estimate of the temperature, we also measured the absorption coefficient of the laser radiation in a bismuth plume at $p = 5$ atm, and $I \sim 10^7$ W/cm^2. We used the method of passing a laser pulse ($\tau = 0.8$ msec) longitudinally through the plume. The average absorption coefficient turned out to be in this case $\kappa \approx 0.4$ cm^{-1}. Assuming that all the atoms are doubly ionized and recognizing that the pressure in the plume is equal to the pressure of the external gas (see Sec. 1), we obtain, from the formula for the bremsstrahlung light-absorption coefficient in a plasma under the condition when the pressure in the plume is equal to the external pressure, a value $T \approx 2.3 \times 10^4$ °K. This value is in satisfactory agreement with the foregoing estimate based on the observed spectra and on the calculation of the equilibrium ionization.

3. In the present experiments we also determined the electron density N_e. These measurements, however, are tentative in character, since the employed spectral instrument did not have a sufficient dispersion (72 Å/mm) and introduced a large apparatus broadening, comparable with the line widths themselves. The apparatus line broadening, which was difficult to take into account, was the main source of the experimental error, which reached 75%. With allowance for this error, the measured electron density on the shock wave front at low pressures ($p \leq 1$ atm) amounts to 2×10^{17} cm^{-3} for bismuth and 8×10^{16} cm^{-3} for aluminum. At a distance from the target on the order of the diameter of the observed spot, N_e exceeds the indicated values by 2–4 times.

4. CONCLUSIONS

We have performed spectral diagnostics of two previously investigated[1-3] gasdynamic regimes of a plasma plume produced in the case of developed evaporation of metals by powerful optical radiation. For aluminum we obtained the temperature-variation profiles along the plume at $p < 1$ atm, which agreed with the gasdynamic picture of vapor flow[1]. In the case of bismuth, we investigated the character of temperature variation along the plume in the gasdynamic region with an immobile shock wave, and in the regime of strong screening of metal by the plasma, when the plume becomes detached from the target. We obtain experimental estimates of the temperature. The particle density in a plume from bismuth and from aluminum was estimated from the Stark broadening of the observed spectral lines.

It must be emphasized that the values of T obtained in this work near the target (plume from shock wave, $p \leq 1$ atm), which exceed the critical temperatures by several times, together with analogous results obtained for bismuth[2] by measuring the vapor velocity in the plume, are the first experimental facts offering evidence of heating the vapor near the target in the region $R \lesssim d$ by a light flux of moderate intensity, $I \sim 10^6$–10^7 W/cm^2 and $\tau \sim 10^{-3}$ sec.

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[1] This was demonstrated in [2] with bismuth as an example. This difference holds also in the case of aluminum.

[2] It is important also that in [4] the diagnostics of a plume from a copper target was based on the Cu-I lines of the neutral atoms. The emission of Cu-I (in analogy with the results of the present paper, for example for aluminum), pertain to the peripheral cold region of the plume and does not reflect its gasdynamic structure.

[3] The spectra in Fig. 1 correspond to a maximum distance up to 7 cm away from the target. This statement is based on spectrograms obtained with a smaller magnification and not shown in Fig. 1.

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Translated by J. G. Adashko
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