DYNAMICS OF LOW-ENERGY LASER-PRODUCED TIN PLASMA

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Emission spectroscopy with a high temporal resolution is used to study the initial expansion stages of the erosive laser-produced tin plasma. The time dependences of the population of excited states of tin atoms and ions are examined. The time of ion recombination was evaluated, and the time dependences of the electron temperature and concentration at distances of 1 and 7 mm from the target are determined.

1. Introduction

The plasma torch emitted from a solid target under the action of powerful laser radiation has a wide range of applications [1, 2]. For instance, the laser-produced tin plasma, being a source of soft x-ray radiation, is a promising object for the next-generation photolithography [3–6].

In works [7, 8], the results of researches dealing with the processes occurring in a laser-produced tin plasma were reported. The main objective of those works consisted in the optimization of both plasma parameters and plasma-chemical reactions in a laser torch in order to obtain thin films with various compositions. Therefore, the research of the laser-produced torch from tin is a challenging task. The method of emission spectroscopy provides the necessary information for the analysis of processes in such plasma to be done. Hence, our work is aimed at studying the temporal characteristics of the laser-produced tin plasma within this method.

2. Experimental Part

For producing a laser plasma, we used an LTIPCh-5 pulse-periodic laser, which operates in the Q-switched mode (12 Hz, 1064 nm, 20 ns, 4×10^8 W/cm^2). A target fabricated of pure tin was mounted in a vacuum chamber at a pressure of residual gases of 6 Pa.

Radiation in the spectral region of 200–600 nm was analyzed with the use of an MDR-2 monochromator and a diffraction grating with 1200 lines/mm. The monochromator wavelength resolution was 0.2 nm. The optical and temporal characteristics of radiation emitted by the laser-produced plasma were registered with the help of an MDR-2 monochromator, a FOTON photo multiplier, and a C1-99 oscillograph. The accuracy of intensity measurements was not worse than 10%. The identification of spectra was carried out according to the data presented in handbooks [9, 10].

3. Discussion of Experimental Results

The dominating processes leading to the formation of excited states of Sn atoms and ions in the plasma of a laser-produced torch which extends into vacuum or a residual gas are the dielectron recombination and thermal processes:

$$\mathrm{Sn}^{+2(o)} + e + e \rightarrow \mathrm{Sn}^{+1(*)} + e',$$

$$\mathrm{Sn}^{+1(o)} + e + e \rightarrow \mathrm{Sn}^{*} + e',$$

$$\mathrm{Sn}^{o} + e' \rightarrow \mathrm{Sn}^{*} + e.$$

The essence of those processes consists in the capture of an electron by an ion in the ground state, with the excess energy being carried away by another electron. As a result, an ion with a lower charge emerges in the upper excited state. The interaction between fast electrons and atoms in the ground state gives rise to the formation of tin atoms in the lower excited states.

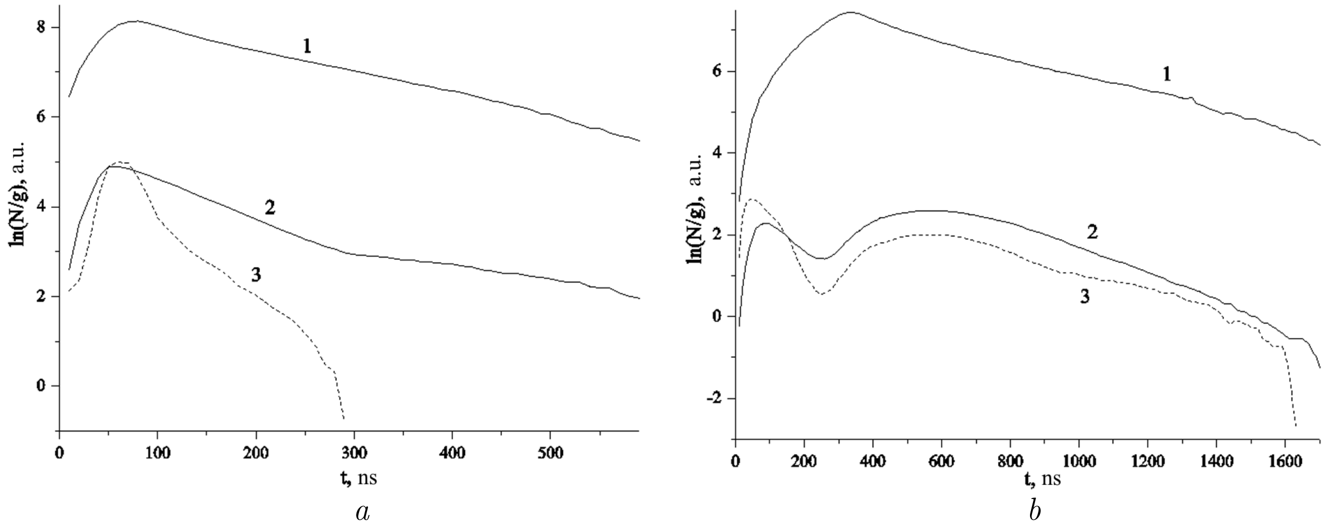


Fig. 1. Population dynamics for excited states of tin atoms and ions with energies of 4.29 (*1*), 5.52 (*2*), and 11.07 eV (*3*) at distances of 1 mm (*a*) and 7 mm (*b*) from the target

T a b l e 1. Parameters for the analysis of time dependences of the radiation intensity of a laser-produced tin plasma at distances of 1 and 7 mm from the target

N	λ, nm	A, 10^8 s^{-1}	Atom, ion	E_L, eV	Term$_L$	E_U, eV	Term$_U$	g
1	303.4	1.51	Sn I	0.21	$5p^2\ ^3P_1$	4.29	$p6s^3P_0$	1
2	242.9	1.77	Sn I	0.43	$5p^2\ ^3P_2$	5.52	$p5d\ ^3D_3$	7
3	328.3	1	Sn II	7.29	$5s5p^2\ ^2D_{3/2}$	11.07	$5s^24f\ ^2F_{5/2}$	6

Using the intensities of spectral lines obtained on the oscillograms, the populations of excited states for atoms and ions were determined by the formula

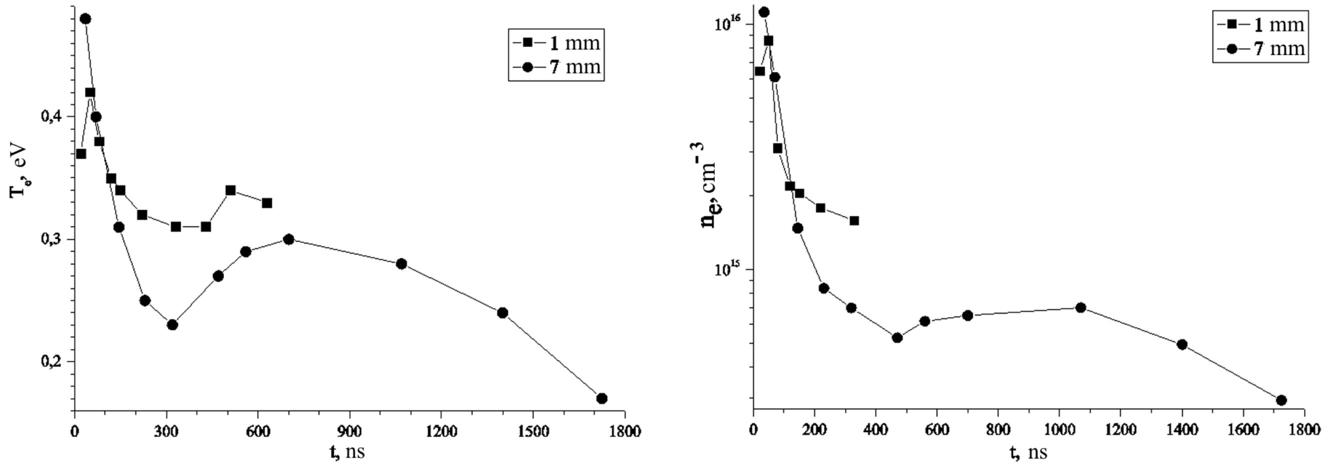
$$\frac{N}{g} = \frac{I\lambda}{Ag},$$

where N is the level population, g the statistical weight, λ the wavelength, I the intensity, and A the transition probability. The temperature of electrons, T_e, in the laser-produced plasma was determined using the graphic method, after having plotted the Boltzmann distribution as the dependence $\ln(N/g)$ versus the level energy. The concentration of electrons, n_e, in the laser-produced tin plasma was calculated from the following relation between the recombination time and the electron concentration, provided that the temperature is known:

$$n_e = (8.75 \times 10^{-27} z^3 t_r T_e^{-9/2})^{-1/2}.$$

Here, z is the charge of a recombining ion, and t_r is the recombination time. The technique for studying the results of measurements of laser-produced plasma parameters was described in work [11] in more details.

In Fig. 1, the variations of populations on the excited states of tin atoms and ions in time are depicted. The data for the analysis of the time dependences of radiation intensity emitted by the laser-produced tin plasma at distances of 1 and 7 mm from the target are quoted in Table 1. At a distance of 1 mm from the target, the populations of excited states grow till the time $t \approx 70$ ns. Then they decrease, and the characters of their reduction are different for excited states with different energies. The main feature is a slowing decrease in the population of upper excited atomic states, which follows after a drastic reduction in the populations of ions observed at $t \approx 250 \div 300$ ns. The variations in the populations of upper and lower excited states can be explained as a result of specific variations of the particle concentrations in plasma, plasma parameters, and a substantial manifestation of three-particle recombination. The character of a variation in the population of upper excited atomic states, which takes place when the population of ionic excited states drastically diminishes (Fig. 1), can reflect an enhancement of the formation of those states at the recombination. In Fig. 1, the reduction in the population of ionic excited states has the most complicated behavior, which demonstrates three different sections of population variation in time. For lower atomic excited states,



a *b*

Fig. 2. Time dependences of the electron temperature (*a*) and density (*b*) in the laser-produced tin plasma

the population variation, after the population having attained the maximum, is linear within the observation period.

At a distance of 7 mm from the target, the maximum in the population of ionic excited states is reached at the observation time $t \approx 50$ ns. For the upper atomic excited states, the character of population change is similar, although the maximum appears at $t \approx 100$ ns. However, their minima coincide, the both being observed at $t \approx 250$ ns. The character of a decrease in the population of atomic excited states is different, depending on whether ions radiate or they do not radiate any more.

At a distance of 7 mm from the target, the population of ionic excited states sharply decreases after $t \approx 1600$ ns. The populations of atomic excited states also diminish: the lower the energy of the lower excited state, the more slowly the population changes. The population of atomic excited state with the lower energy grows till $t \approx 350 \div 400$ ns, reaches a maximum, and, afterward, smoothly diminishes.

After the time $t \approx 300$ ns, the ionic excited states and the upper atomic excited state demonstrate a recurrent growth in the level population. The second maximum is observed at $t \approx 550$ ns. The populations of the upper ionic excited states at distances of 1 and 7 mm from the target reach the corresponding maxima at almost the same time. This means that the propagation velocity of the ionic component at the leading edge changes insignificantly. Therefore, at the 1-mm distance from the target, the concentration profile corresponds to plasma parameters till $t \approx 300$ ns. At the 7-mm distance, the maximum of radiative transitions from the lower excited states is not revealed, when the first maximum for the radiation emission at transitions from the upper atomic excited states and ionic excited states is reached. Therefore, the lower atomic excited states, which are formed as a result of thermal processes, will change their behavior in time, but reflecting the specificity of shock-wave formation. At the same time, the upper atomic excited states reflect the recombination mechanism of their formation, and their radiative transitions reproduce well the temporal dependence of ionic radiation intensity.

In Fig. 2, the time dependences of the electron temperature and concentration in the laser-produced tin plasma are depicted. The variation of the electron temperature in time is characterized by two maxima. The first maximum of the electron temperature is observed at the leading edge of the plasma at $t = 30 \div 50$ ns. The corresponding maximum values are 0.42 eV at 1 mm and 0.48 eV at 7 mm from the target. The second maximum is well pronounced at a distance of 7 mm from the target. It is observed within the time interval from 300 to 1800 ns. The maximum temperature value in the second maximum (0.3 eV) is reached at $t \approx 700$ ns. At a distance of 1 mm from the target, the second maximum is less pronounced; it is observed at $t \approx 500$ ns, and its magnitude reaches 0.34 eV.

The total interval of electron temperature change amounts to 0.42–0.3 eV during 650 ns at a distance of 1 mm from the target and to 0.48–0.17 eV during 1800 ns at a distance of 7 mm. According to the adiabatic model, the plasma volume is different at different distances from the target, and the smaller volume corresponds to a higher temperature. The temperature drop at 7 mm from the target after achieving the first maximum is more drastic. The authors of work [12] showed

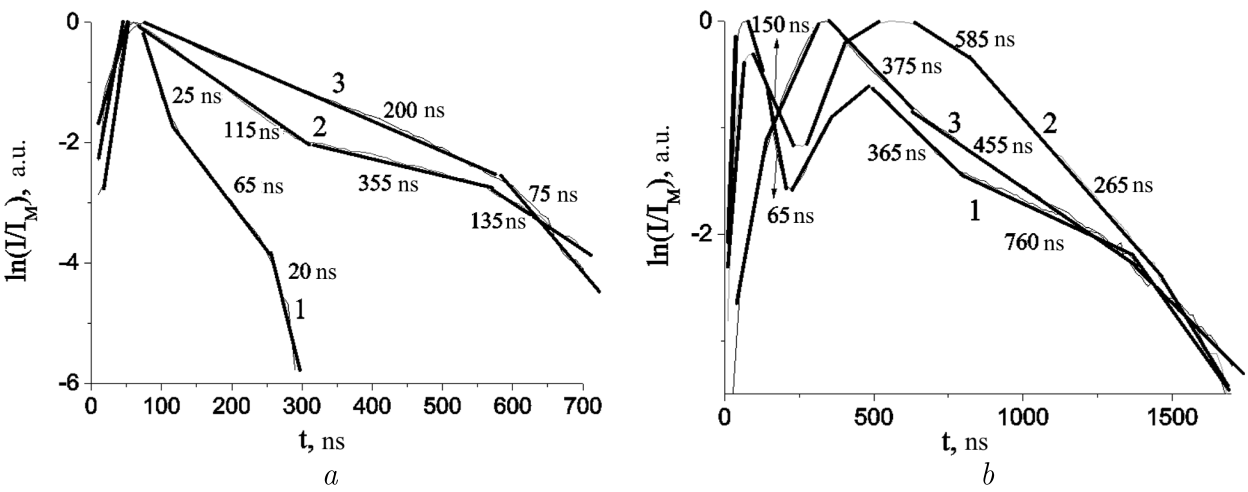


Fig. 3. Time dependences of the logarithms of the normalized intensities for various spectral lines of tin atoms and ions: 328.3 nm Sn II, $E_B = 11.07$ eV (*1*); 242.9 nm Sn I, $E_B = 5.52$ eV (*2*); and 303.4 nm Sn I, $E_B = 4.29$ eV (*3*) at distances of 1 mm (*a*) and 7 mm (*b*) from the target

that, if the distance from the target is small (< 4 mm), the distribution of radiation emitted by the laser torch is characterized by the presence of one maximum, with the second one appearing only at larger distances. In work [13], we demonstrated that such distances from the target are associated with the shock wave formation. Hence, the first maximum in our dependences characterizes the plasma before it has undergone the action of gas-dynamic effects, and the second maximum may probably be a consequence of the shock wave formation.

From the analysis of the electron concentration n_e varying in time, it comes up that, at a distance of 1 mm from the target, the maximum is observed at $t \approx 50$ ns and amounts to 8.6×10^{15} cm^{-3}; at a distance of 7 mm from the target, the first maximum is observed at $t \approx 40$ ns and amounts to 1×10^{16} cm^{-3}; then the concentration of electrons smoothly decreases to a value of 5.3×10^{14} cm^{-3}, which is observed at $t \approx 456$ ns. The second maximum is observed at $t = 1070$ ns, with the corresponding concentration of electrons $n_e = 7.1 \times 10^{14}$ cm^{-3}; afterward, the concentration falls down to 2.9×10^{14} cm^{-3} at $t = 1725$ ns [14]. The second maximum reflects gas-dynamic processes in plasma. The front part of the laser torch interacts with a surrounding gas, so that its velocity slows down. The second maximum can be produced owing to the emergence of turbulence, at which the kinetic energy of atoms and ions transforms into the excitation one. From the analysis of those results, it follows that the electron concentration and temperature decrease as the distance from the target grows. However, if the power introduced into the target increases, the mentioned parameters also increase.

From the time dependence of the spectral line intensity for an ion with the charge z, it is possible to determine the recombination time for ions with the charge $z + 1$. Namely, the recombination time equals the tangent of the slope angle for the straight line obtained while plotting the logarithm of the intensity I versus the time t,

$$t_r = \frac{\Delta t}{\Delta \ln \dfrac{I}{I_M}},$$

where I_M is the intensity maximum value. The peculiarities in the variation of the intensity in time give an additional information concerning the course of physical processes in the laser-produced plasma. The technique of this analysis was described in work [15]. From Fig. 3, one can see that, at a distance of 1 mm from the target and within the time interval of 100–200 ns, the recombination time grows to $t = 65$ ns, and, if $t > 250$ ns, amounts to 20 ns.

The recombination time changes several times in a jump-like manner during the observation period. At a distance of 7 mm from the target, the glow of ions and atoms is observed up to 1500 ns. The recombination time was 150 ns for single-charged and 65 ns for two-charged tin ions at this distance and at $t = 100 \div 250$ ns. If the observation time was $t = 500 \div 700$ ns, the recombination time was 585 ns for single-charged ions and 365 ns for two-charged ones [16].

The time of ion recombination depends on the temperature and the concentration of electrons, being expressed by the following relation [15]:

$$t_r \sim T_e^{9/2}/n_e^2.$$

By plotting the logarithmic dependences of the temperature and the concentration of electrons, as well as the population of the level that corresponds to the excited state of this ion, one can see that the temperature and the population also change in a jump-like manner at $t = 100 \div 120$ ns and 200–220 ns. In the time interval between those values, the temperature and the population change linearly. For the electron concentration, the jump in the linear dependence is observed at $t = 120$ ns.

Thus, the peculiarities in a change of the recombination time are related to variations of the electron temperature and the ion concentration. They may be a result of gas-dynamic processes which govern the motion and the expansion of a plasma torch. At jump points, the modifications in the characteristic stages of plasma torch expansion are possible. In the logarithmic dependence of the population variation at the upper excited states of tin atoms on the time, one can observe a linear section with a jump at a time of about 300 ns. The specific variation of the concentration profile in time for atoms and ions is explained by the fact that ions are located in the center of the plasma cloud, whereas atoms at its periphery. Therefore, the corresponding specificities in the motion and the dynamics of plasma expansion manifest themselves differently. As for the central part of the plasma, the manifestation of a specificity of the concentration profile, which was formed at the plasma emergence, should be more probable. At the same time, for atoms in the external plasma layer, the processes of interaction with the surrounding gas play a more important role.

If thermal processes took place, the intensity variation would manifest itself identically for all excited states with all energies, which can be well observed at a distance of 7 mm from the target.

The recombination time of single-charged ions at the trailing edge amounts to 115 ns at $t < 250$ ns, 355 ns at $t < 550$ ns, and 135 ns at $t > 550$ ns. For $t > 300$ ns, the variation of the electron temperature has a considerable influence, because the concentrations of electrons and ions change much more slowly.

At a distance of 1 mm from the target, the emission intensity at transitions from the lower excited states changes more rapidly than that at transitions from the upper excited states after a time of 300 ns. This fact testifies to the recombination processes that run rather slowly at the trailing edge of the plasma and lead to a reduction in the intensity decrease rate for atoms formed as a result of those processes. The velocity of motion at the trailing edge of the plasma will be much lower than that at the leading one.

The minimum value of electron temperature at a distance of 7 mm from the target was observed at the time $t = 320$ ns, and the corresponding minimum for the electron concentration at $t = 470$ ns. A spatial redistribution of particles is observed, as well as a change of the motion character from directed to chaotic, which is accompanied by the energy release owing to the recombination processes and the mixing.

The fall in the population of ionic states slows down, when the concentration of electrons is maximal. At the time $t = 700$ ns, there appears a second maximum for the electron temperature; almost the same takes place for the electron concentration as well. At a distance of 7 mm from the target and when the minimum electron temperature takes place at $t \approx 320$ ns, the maximum population of lower excited states is observed (at $t \approx 335$ ns). At the same time, when the concentration of electrons is growing (till the time $t = 1200$ ns), the temperature of electrons, on the contrary, falls down. It is probable that, owing to the shock wave formation, the temperature at the leading edge is maximal, and the electron concentration increases according to the energy release by recombination processes and the changes in the character of motion. Particles with high kinetic energies undergo collisions at their motion, so that their ionization becomes possible.

The maximum population of the lower atomic excited state is observed at the time $t = 75$ ns at a distance of 1 mm from the target and at $t = 335$ ns at a distance of 7 mm from the target. When the plasma expands in the shock wave regime, the level population does not coincide with the spatial distributions of the electron temperature and concentration.

4. Conclusions

Emission spectroscopy of a laser-produced tin plasma torch allowed the dynamics of population of the excited states of tin atoms and ions to be studied. It is found that the variations in the population of upper and lower excited states can be explained by specific features in the variation of the particle concentration in the plasma and the changes of plasma parameters. The total interval of electron temperature variation at a distance of 1 mm from the target amounted to 0.42–0.3 eV during the time period of 650 ns. At a distance of 7 mm from the target, this interval equals 0.48–0.17 eV, and the time period was 1800 ns.

At a distance of 1 mm from the target, the maximum electron concentration $n_e = 8.6 \times 10^{15}$ cm^{-3} was observed at 49 ns. At the same time, at a distance of 7 mm from the target, two maxima were observed: at 40 ns ($n_e = 1{\times}10^{16}$ cm^{-3}) and 1070 ns ($n_e = 7.1{\times}10^{14}$ cm^{-3}). If a shock wave is formed, the temperature at the leading edge of the plasma is in its maximum, and the concentration of electrons increases owing to the energy release at recombination processes and the change in the character of laser torch expansion.

The times of the ion recombination at distances of 1 and 7 mm from the target are determined. At a distance of 1 mm from the target and within the observation time interval of 100–200 ns, the recombination time for double-charged ions grows from 30 to 65 ns; at the observation time longer than 250 ns, the recombination time amounts to 20 ns. For single-charged ions, the corresponding recombination times amount to 115 and 355 ns, respectively. The recombination time demonstrates several jump-like changes during the observation period. At points that correspond to jumps, changes in the characteristic stages of plasma torch expansion are possible. The recombination processes, which run rather slowly at the trailing edge of the plasma, reduce the intensity decrease rate for radiation emitted by atoms that are formed as a result of those processes. At a distance of 7 mm from the target, the recombination times are 150 ns for single-charged ions and 65 ns for two-charged ones, provided that the times of observation are 100–250 ns. For the times of observation within the interval 500–700 ns, the recombination time is 585 ns for single-charged ions and 365 ns for double-charged ones.

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ДИНАМІКА НИЗЬКОЕНЕРГЕТИЧНОЇ ЛАЗЕРНОЇ ПЛАЗМИ ОЛОВА

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Р е з ю м е

Вивчення початкових етапів розширення ерозійної лазерної плазми олова проведено методом емісійної спектроскопії з високим часовим розділенням. Досліджено часові залежності заселеності збуджених станів атомів та іонів олова. Проведено оцінку часів рекомбінації іонів, встановлено залежності від часу для температури і концентрації електронів лазерної плазми на відстанях 1 і 7 мм від мішені.