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NATURE OF DEVIATIONS FROM THE REGULARITIES OF THREE-PARTICLE RECOMBINATION IN DENSE PLASMA

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An explanation is proposed for the phenomenon of deviations from the functional laws of three-particle recombination in dense electric arc plasma of argon at an initial electron density of ~ 10^{16} cm^{-3}. Experimental data, which are the basis of the work, were obtained by spectral and probe methods during the study of plasma decay of a microsecond pulse electric arc in argon 6 μs at an initial pressure of 70...700 Pa and a discharge current amplitude of 6 kA. Theoretical estimates show that the reason of the observed effects is the self-absorption of resonant spectral lines of argon, which significantly limits the recombination flow of electrons in the energy structure of atoms.

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INTRODUCTION

We will consider the stated topic on the example of electric arc argon plasma of a short-term, microsecond discharge at a pulse pressure close to atmospheric. Plasma whose parameters are sufficient to maintain local thermal equilibrium (LTE) in its volume within the existing boundaries is considered dense.

The factor of boundaries is introduced in this consideration due to the fact that in most practically important applications plasma can be in LTE only if resonant radiation of plasma-forming particles remains trapped in their volume. The determining for this is the requirement that the characteristic path length of resonant radiation quanta just be less than the characteristic size of the plasma. This issue will be discussed in more detail below, in Chap. 2.

Being the cheapest among inert gases, argon has a wide practical application in plasma technologies (see, for example, [1-6]). Therefore, this plasma has become the object of numerous physical studies. The fact that the nature of deviations from the mechanism of three-particle recombination in such plasma has not yet been definitively clarified is all the more surprising, given the basic nature of these processes for any plasma. An illustration can be the materials of the classic monograph [7 (Fig. 6.3)], from which it follows that the regularities of its recombination decay, even at a qualitative level, do not correspond to the collision-radiation model for dense plasma [8].

The illustration cited from the book [7] includes, in turn, the results of numerical calculations by Bates et al. [9], which are compared with experimental data for hydrogen, helium, argon, and xenon. (For convenience, this illustration is reproduced below as Fig. 1). However, the use of these data did not allow the author to adequately explain the reason for the deviations from the regularities of three-particle recombination in dense argon plasma in doctoral thesis [10]. Thus, this experimental result remained for a long time without a theoretical justification. It took many years before this fact found a logical explanation in the report [11].

Before starting the presentation of the main material, it should be emphasized that the problem of deviations from the regularities of three-particle recombination in dense plasma can be considered in a much broader aspect. Indeed, in recent decades, the research direction of recombination processes in dense plasma, connected with taking into account the role of microfields, has been actively developing. At electron densities $N_e > 10^{17}$ cm^{-3}, they become close to intraatomic ones [12-14]. Their role as the field strength increases is reduced first to a significant expansion of the highest levels in the energy structure of the atom and, accordingly, the spectral lines emitted from them, and then to the disappearance of the latter. Thus, the number of levels at which electrons can be captured in step processes of their recombination decreases. As a result, regardless of the increasing frequency of collisions with increasing N_e, the recombination processes slow down significantly – by orders of magnitude. These effects, in turn, are also internally related to the degree of non-ideality of the plasma, but are not uniquely determined by them, as shown in [14].

It should be emphasized once again that in the absolute majority of studies of the microfields effect on the erosion of the highest levels in argon, its role is significant at $N_e > 10^{17}$ cm^{-3}. In the conditions of the experiments of this work, it is observed even at densities of electrons two orders of magnitude lower, which requires an adequate explanation.

In general, according to the conclusions of [14], there is currently no model that would explain all the observed effects of deviations from the mechanism of three-particle recombination in dense plasma.

1. FEATURES OF CHARGED PARTICLES RECOMBINATION IN PLASMA

A necessary condition for the effective elementary act of charged particles recombination is the simultaneous observance of the conservation laws of momentum and energy in the collision process. This condition is easier to fulfill within the framework of the three-particle recombination mechanism. However, in rarefied plasma, the probability of three particles colliding at the same time is small, so this mechanism becomes prevalent only in plasma with a high concentration of charged particles which corresponds to the object of research in this work. It is generally accepted that the recombination coefficient in this case has the form [8]

$$\alpha = 5.4 \cdot 10^{-27} T_e^{-9/2}, \quad (1)$$

and the balance equation for the density of electrons in the decay of a pulsed plasma [7] may be presented as

$$-dN_e/dt = N_e/\tau_D + \alpha N_e^3, \quad (2)$$

where τ_D is the characteristic time of particle diffusion losses. Relations (1) and (2) are classical in physics of low-temperature plasma. However, it has long been known that it is often violated in dense plasma [7] – see Fig. 1. It is easy to conclude that the plasma recombination coefficients for all elements shown in Fig. 1, at relatively high temperatures and, accordingly, high electron densities, do not fall on a straight line *1* corresponding to the theoretical dependence (1).

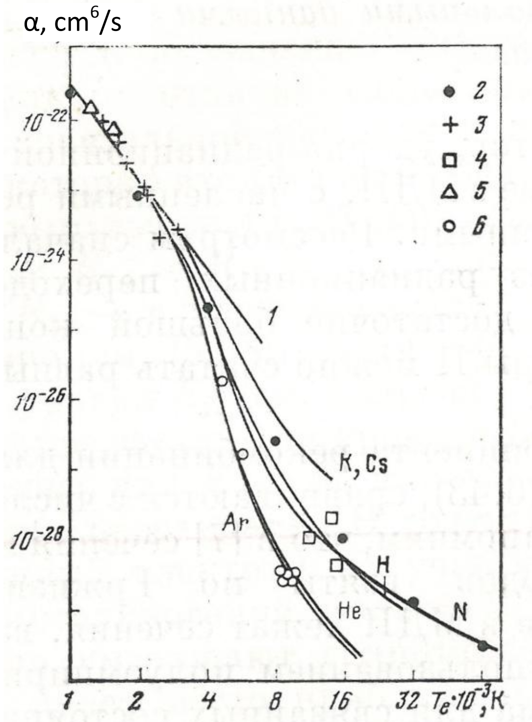

Fig. 1. Triple recombination coefficients depending on T_e for plasma of various elements: 1 – dependence (1); 2 – numerical results of Bates et al.; 3, 4 – two groups of experimental results for hydrogen; 5 – experimental results for helium; 6 – experimental results for xenon and argon

2. DEFINITION OF DENSE PLASMA

As already mentioned, plasma whose parameters are sufficient to maintain LTE in its volume within existing boundaries is considered as dense in this work. To quantify deviations from the LTE, a two-level model of an atom with two energy states – ground and excited – is usually introduced [7]. In the stationary state for plasma consisting of such atoms, the ratio is valid

$$N_1\omega_{12} = N_2\omega_{21} + N_2 A^*_{21}. \quad (3)$$

In Eq. (3), N_1 and N_2 are the concentrations of atoms in the ground and excited states, ω_{12} and ω_{21} are the transition probabilities as a result of collisions between atoms and electrons, A_{21}^* is the effective probability of a radiative transition, which takes into account the self-absorption of radiation in the plasma. The latter contributes to reducing the influence of radiant energy losses with accuracy up to the coefficient entered below:

$$A^*_{21} = A_{21}\theta(r), \quad (4)$$

where $\theta(r)$ is the probability of a photon leaving the point r outside the plasma without absorption. It follows from this that the criterion of the relative balance of two states is inequality

$$A^*_{21} / \omega_{21} << 1. \quad (5)$$

The ratio (5) reflects the fact that the root cause of equilibrium states is chaotic collisions of particles, and radiation is the factor that violates the regularities of equilibrium – according to the Boltzmann distribution – population of excited levels of an atom. Since A^*_{21} is smaller than A_{21}, this formally explains the role of radiation self-absorption as a factor contributing to the establishment of LTE in plasma. In a practical sense, it is thanks to the self-absorption of resonant radiation that it is possible to observe plasma, for example, inert gases, whose state is close to LTE, in electric arcs in laboratory conditions. The same applies to the plasma of electric arcs in a mixture of atmospheric air with metal vapors of low-melting electrode materials.

Just due to the self-absorption of resonant radiation in the plasma of an electric arc, it is characterized by minimal energy losses for the transportation of electricity along its channel. In other words, it is a technological device of high energy efficiency. This property has become one of the determining factors for the active use of electric arc plasma in various industrial applications [15, 16]. One of the oldest among them, as well as the most famous, is electric arc welding.

Returning to Eq. (4), the value $\theta(r)$ is determined mainly by the ratio of the characteristic size of the plasma volume, or if we are talking specifically about the arc, its radius R and the length of the free path of radiation [7, 17]

$$\langle l \rangle = \kappa_0^{-1}, \quad (6)$$

where the absorption coefficient κ_0 in the center of the spectral line is defined as:

$$\kappa_0 = p f_{ik} \lambda_{ik}^2 N_k / \Delta\lambda. \quad (7)$$

Here p is a numerical coefficient, f is the oscillator strength for an optical transition with a wavelength λ, N_k is the density of emitting particles in the lower state of the optical transition, $\Delta\lambda$ is the full width at half maximum of the spectral line contour.

3. OBJECT OF STUDY

The analysis of this work is based on experimental results [18, 19]. A modified version of the electric shock tube (EST) used for studies of dense plasma. This device was considered promising in the 1970s for obtaining high-speed flows of thermal plasma behind the front of a shock wave generated by a pulsed discharge [20]. However, the obtained flow was, in particular, turbulent and lost the advantages of thermal origin [21, 22].

EST is shown schematically in Fig. 2. It is created on the basis of a glass tube with an inner diameter of 1.6 cm, which includes ring electrodes E_1 – E_2 made of covar. The width of each of them is 1.5 cm; they are separated by a 5 cm long segment of glass tube. They formed discharge gaps for generating a pulsed discharge between electrodes. These electrodes were embedded in the tube in such a way that their inner surfaces coincided with the surface of the glass.

In the experiments described below, in contrast to the traditional ones where EST was applied, the main emphasis is placed on plasma studies in discharge section $E_1 - E_2$, in the middle plane of mirror symmetry between electrodes (and not in the so-called section of plasma expansion beyond the discharge gap). This, in particular, allows us to study the effect of expansion on the properties of non-moving plasma in the plane of its mirror symmetry.

Steel pistons $P_1$and P_2 are placed in the internal volume of the EST, which can be brought close to the discharge gap with the help of a magnet. In this case, the decay of the plasma was not accompanied by its expansion. Thus, a comparative version of the experiment on the effect of plasma expansion on its decay was implemented.

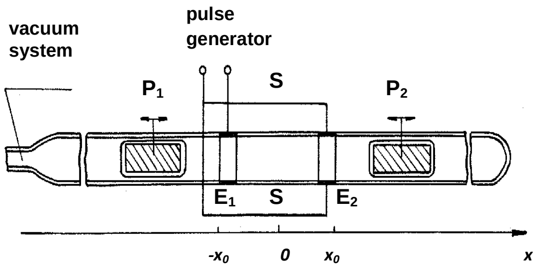

Fig. 2. EST to study plasma decay and system of coordinates: E_1, E_2 – ring electrodes; S – a system of return current conductors symmetrical around the discharge gap; P_1, P_2 – steel pistons every confined in a glass jacked

The decay of Ar plasma was studied at the initial pressure p_0 from 70 to 700 Pa. The plasma was obtained using a pulse current generator (PCG) based on a capacitor bank with a capacity of 6 μF, which formed a current pulse with amplitude of 6 kA and duration of 6 μs in the form of a half-period of a sine wave. The mentioned generator was specially designed to power supply the EST [23].

In the conditions of kiloampere pulsed currents, the symmetry of the plasma in the discharge gap could be distorted due to the magnetic interaction of the discharge current and the return current in the outer conductor. To eliminate this effect, a system of return current conductors *S* symmetrical around the discharge gap was used (see Fig. 2).

4. DIAGNOSTICS OF PLASMA

Spectral analysis showed that the nature of the plasma radiation spectrum changes depending on the initial argon pressure. At a pressure of 70 Pa, the spectrum was dominated by the linear emission of ArI atoms and ArII ions; the level of the continuum in radiation was negligible. And, on the contrary, at a pressure of 700 Pa, the spectral lines were practically indistinguishable against the background of the high intensity of the continuum.

The technique of the relative intensities of the ArII 362.2 and ArI 360.6 nm spectral lines provided the highest accuracy of spectroscopic temperature measurements. At the same time, the degree of deviation from the ionization equilibrium of the concentration of particles in a higher charge state, i.e. argon ion, was determined by the method of similarity in relation to known calculated data for helium plasma [24]. This allows us to extend this technique of temperature measurements to the region of partial LTE (PLTE) existence of argon plasma. It ensured high measurement accuracy: a temperature change of only 0.1 eV corresponds to a change in the ratio of line intensities by almost 2 orders of magnitude.

The relative radial distribution of the electron density $N_e(r)$ in the discharge was determined by the spatial distribution of the radiation intensity of the argon continuum J_c in accordance with the relation [18]

$$J_c \sim N_e^2. \quad (8)$$

The result of measurements determined by formula (8) practically does not depend on temperature. Absolutization of these measurements was carried out by broadening the spectral lines of 504.4 nm ArI and 480.8 nm ArII according to the known parameters of their broadening. These lines contours were recorded using a Fabry-Perot interferometer with a time resolution no worse than 1 μs [25].

Control measurements in the plasma volume, as well as special studies of near-wall plasma properties, were carried out by probes. Here, the probe measurements are carried out in the discharge gap, which is powered by a high-voltage PCG. Its turning is accompanied by a sharp change of the space potential. Therefore, to measure plasma parameters, double probes with galvanic separation from the measuring circuit using a highly sensitive Rogovsky belt were used.

The main methodological difficulty of using the probe technique is that for a dense plasma it is impossible to produce a probe with a size smaller than the length of the free path of charged particles. Accordingly, classical processing of measurement results according to the Langmuir method is excluded. However, probe measurements processed in the diffusion approximation in combination with the results of spectral studies provided valuable information about the characteristics of dense plasma, including the role of wall effects in plasma decay. The elements of technique of probe measurements and the obtained results are presented in detail in a separate publication [26].

5. EXPERIMENTAL RESULTS

The behavior of the particles temperature in the pulse plasma at p_0 = 70 Pa is shown in Fig. 3. In the process of decay (after t = 5 μs), the temperature in the modes of expansion and non-moving plasma remains almost unchanged at the level of 1.3 eV within 5 % (with no discussing here the difference in both modes of its decay). The intensities of the spectral lines themselves during the time interval indicated in the figure changed by two orders of magnitude, which, in fact, limited the time range of measurements. The reliability of these results during the specified period was controlled by the standard method of the relative intensity of ten spectral lines of ArII in the energy range of the upper levels of the corresponding spectral transitions of 19.2...25 eV, the accuracy of which, however, was ± 15 %. Comparing the intensities of the ArII 446.1 and 463.7 nm spectral lines most confidently recorded at the late stages of plasma decay, in which the upper levels of the spectral transitions are significantly different from each other (respectively, 19.2 and 21.1 eV), we can conclude, that the temperature does not change significantly up to 70 µs.

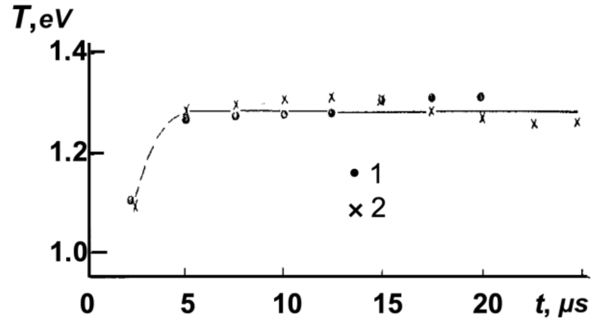

Fig. 3. Results of temperature measurements in the argon plasma in the EST at p_0 = 70 Pa under conditions of expansion (1) and without it (2)

A similar situation was observed at higher argon pressures, with the only difference being that the *T* level slightly decreases (respectively, 1.2 and 1.1 eV at pressures of 280 and 700 Pa). However, temperature measurements were only possible based on the relative intensity of the continuum that prevailed in the radiation spectrum; their accuracy did not exceed ±20 %.

In this type of measurements, in the case of temperature-inhomogeneous plasma, the error in the values of the intensities of the spectral lines ArII and ArI may increase due to their disproportionate illumination along the radius. In our case, the radial profiles of the radiation intensities of these spectral lines turned out to be identical. This allows us to draw a conclusion about the constancy of temperature along the radius even without performing the Abel inversion procedure.

An example of the obtained results of $N_e(t)$ measurements in the decay is presented in Fig. 4. They are given in coordinates, which already at a qualitative level allow us to draw the most important conclusions regarding the behavior of the plasma during the decay process. When the pressure changed from 70 to 700 Pa, the initial value N_e^{max} varied from $2 \cdot 10^{16}$ to $6 \cdot 10^{16}$ cm^{-3}. This corresponds to the stage of plasma existence in a strongly ionized state with the degree of ionization of argon atoms ω ~ 1...0.2. A comparison, on the one hand, of N_e values determined from the experiment, and on the other hand, calculated from the Saha equation using experimentally determined temperatures and concentrations of the heavy component at a known initial pressure of argon, indicates their closeness and, accordingly, the LTE in the plasma. A significant decrease in N_e in the process of plasma decay at an almost constant temperature value corresponds, in particular, to the transition to the state of PLTE for the ionic component [24].

Thus, the application of the model proposed above for the diagnostics of processes in the recombining argon plasma of an electric arc is quite adequate to the physical conditions of this plasma existence.

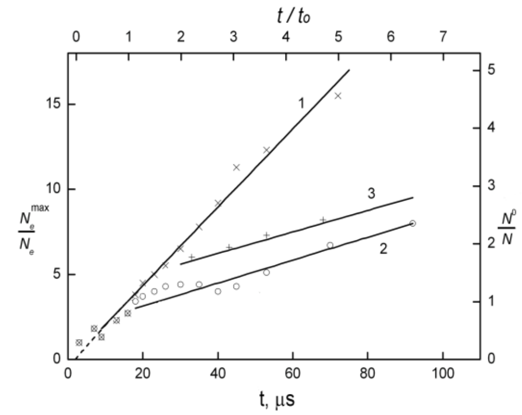

Fig. 4. Plot of the ratios (N_e^{max}/N_e) for density of electrons in expanding (1) and motionless plasma (2) as well as (N^0/N) for density of atoms in expending plasma as a function of time. p_0 = 280 Pa, $N_e^{max} = 2.1 \cdot 10^{16}$ cm^{-3}

6. INTERPRETATION OF THE RESULTS OBTAINED

From a qualitative viewpoint, the picture of plasma decay at an almost constant temperature – including in the conditions of its expansion – can be imagined in such a way that the temperature stabilizer is the energy of resonant radiation, which is almost completely enclosed in the conditions of a dense plasma of an electric arc in its volume. Indeed, the length of the free radiation path of resonance spectral lines ArI 104.82 and 106.66 nm, which is determined similarly [24], is $\langle l \rangle = 8.6 \cdot 10^{-4}$ and $4.2 \cdot 10^{-3}$ cm, respectively (see also Eqs. (6) and (7)). Energy excitation of resonance levels of argon atom is E_r = 11.83 and 11.62 eV, respectively. Since the energy of resonance transitions is 9 – 11 times higher than the thermal energy of plasma particles, it is sufficient to maintain the temperature of plasma particles at an almost constant level during dissipation in collisional processes to maintain the temperature of plasma particles at an almost constant level. At the same time, radiation losses from the arc channel are practically excluded.

The following analysis of the role of collision processes also includes the participation of metastable levels of E_m = 11.55 and 11.72 eV in them [27].

The balance equation for the density of electrons in the decay of dense pulsed plasma in the approximation of the three-particle recombination mechanism was presented above (2) as well as the recombination coefficient in it (1). Let's bring it to the form that corresponds to the graphic interpretation presented in Fig. 4:

$$- N_e^{-1}\, dN_e/dt = \tau_D^{-1} + \alpha N_e^2 = \alpha' N_e, \qquad (9)$$

where $\alpha' = \alpha N_e$. The straightness of the graph in the coordinates of the graph in Fig. 4 clearly shows that α' = const, which means that the recombination mechanism formally corresponds to the two-particle mechanism. The value τ_D^{-1} from equation (9) corresponds to the intercept of the ordinate axis in Fig. 4, which is nearly zero; therefore, the characteristic diffusion time τ_D exceeds the duration of observation of the decay process.

Thus, when observing the decay of argon plasma, there is a somewhat unexpected formal correspondence of the process to the two-particle mechanism of plasma recombination. At the same time, the character presented in Fig. 4 dependence is internally consistent with the results of temperature measurements (see Fig. 3): only minor changes in T are due to recombination heating of particles under conditions of significant optical thickness of the plasma in resonance lines with relatively small losses due to diffusion and heat conduction. Accordingly, the recombination coefficient α' = const and the data in Fig. 4 are approximated by a straight line.

The expression for the recombination coefficient follows from work [8] taking into account the ratio $\alpha' = \alpha N_e$ (see Eq. (9)) and has the form of a power dependence on temperature:

$$\alpha' = 5{,}4\cdot 10^{-27} T_e^{-9/2} N_e^{*} \qquad (10)$$

(α' – in cm^3/s, $T_e = T$ – in eV). Using the data of Fig. 4 it is not difficult to determine that it is $\alpha' = 10^{-11}$ cm^3/s; this at $T_e = 1.3$ eV corresponds to the value $N_e^* = 2\cdot 10^{15}$ cm^{-3}, i.e. the lower value of the N_e measurement range in the experiment.

7. THE ROLE OF RADIATION SELF-ABSORPTION IN THE DECAY OF DENSE PLASMA

The obtained set of experimental results can be explained by significant self-absorption of resonant radiation. Under these conditions, on the one hand, considerable recombination heating of plasma particles takes place, and on the other hand, the speed of only collisional transitions from resonance levels is insufficient to close the flow of recombining electrons to the ground level in the energy structure of the atom. Indeed, the recombination *flow* of electrons should be $d\Phi_e/dt = \alpha' N_e^2 = 10^{19}$ cm^{-3}/s at $N_e = 10^{15}$ cm^{-3} according to (9) and (10). However, the limiting value of the resulting flow of electrons in collisional processes to the ground level from the resonant level, the population of which N_r is in equilibrium with the electron continuum, is only $d\Phi_g/dt = N_r\omega_{rg} \sim 10^{18}$ cm^{-3}/s, where ω_{rg} is the frequency of acts of collisional deactivation of resonant levels (see [7, 17]) using the effective cross section of these processes from publication [6]), which are an order of magnitude smaller. And only at $N_e = 10^{16}$ cm^{-3} they become equal. Thus, in the investigated range of N_e values, the recombination flow of electrons in the energy structure of argon atoms is limited by collisional transitions from the resonance level, which outwardly corresponds to the two-particle recombination mechanism.

CONCLUSIONS

Effects of deviations from the mechanism of three-particle recombination in dense plasma, observed in experiments for a long time, found their theoretical explanation in this work. It is based on the special role of radiation in a dense low-temperature plasma, and in this particular case, the self-absorption of resonant radiation.

On the one hand, it is thanks to the self-absorption of resonant radiation that it is possible to observe plasma, for example, inert gases, whose state is close to LTE, in electric arcs in laboratory conditions. The same applies to the plasma of electric arcs in a mixture of atmospheric air with metal vapors of low-melting electrode materials.

On the other hand, as it is clearly visible from the results of this work, the self-absorption of radiation limits the possibilities for closing the recombination flow of electrons to the ground level in the energy structure of atoms, which leads to deviations from the regularities of the three-particle recombination mechanism usual for a dense plasma.

Returning to the general nature of the special role of radiation in dense low-temperature plasma, it should be emphasized that just due to the self-absorption of resonant radiation in the plasma of an electric arc, it is characterized by minimal energy losses for the transportation of electricity along its channel. In other words, it is a technological device of high energy efficiency. This property has become one of the determining factors for the active use of electric arc plasma in various industrial applications.

Another distinct feature of the role of the self-absorption of resonant radiation in the plasma is a special mechanism of deviation in LTE in a dense plasma of electric arcs in a mixture of atmospheric air with a metal vapor of low-melting electrodes. It arises in conditions when the characteristic length of free path of resonant radiation is close to the radius of the arc channel and is characterized by overpopulation of resonance levels of atoms of plasma-forming particles on the periphery of the arc [15, 16].

In modern technologies of using dense plasma for waste processing or gasification of carbon-containing raw materials, plasma radiation is an important gasifying agent, so it is important to use the resource capabilities of plasma torches used for this purpose in the most rational way.

In summary, problems related to the effects of (self)absorption of radiation in non-uniform plasma require continued systematic research.

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ПРИРОДА ВІДХИЛЕНЬ ВІД ЗАКОНОМІРНОСТЕЙ ТРЬОХЧАСТИНКОВОЇ РЕКОМБІНАЦІЇ В ЩІЛЬНІЙ ПЛАЗМІ

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Запропоновано пояснення явища відхилень від закономірностей трьохчастинкової рекомбінації в щільній плазмі аргону при початковій концентрації електронів ~ 10^{16} см$^{-3}$. Експериментальні дані, які покладені в основу роботи, отримані спектральним та зондовим методами при дослідженні розпаду плазми імпульсної електричної дуги в аргоні тривалістю 6 мкс при початковому тиску 70...700 Па та амплітуді розрядного струму 6 кА. Теоретичні оцінки показують, що причиною спостережуваних ефектів є самопоглинання резонансних спектральних ліній аргону, що істотно обмежує рекомбінаційний потік електронів у енергетичній структурі атомів.