EFFECT OF RADIATION TRANSFER ON A DEVIATION OF DENSE ELECTRIC-ARC PLASMA FROM THE EQUILIBRIUM STATE: CRITERION APPROACH

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A principal role of radiation emission processes in deviations of the electric-arc plasma at the atmospheric pressure from a state of local thermodynamic equilibrium has been estimated, by taking the radiation transfer into account. The problem was considered using a cylindrical wall-stabilized electric arc as an example. The solution was obtained in the approximation of local thermodynamic equilibrium with regard for the processes of radiation transfer and radiation losses in plasma. The results of numerical simulation obtained for copper atoms under conditions that correspond to the state of plasma in the atmospheric electric arc between melting copper electrodes confirm the existence of deviations from the equilibrium distribution between the populations at the resonance and ground energy levels.

1. Role of Radiation Emission Processes in a Violation of the Equilibrium State in Plasma

In the overwhelming majority of publications devoted to plasma and gas discharge physics, the arc-discharge plasma is assumed to be in the local thermodynamic equilibrium (LTE) state at pressures $p \geq 1$ atm . From the viewpoint of the terminological classification, it is the so-called thermal plasma [1]. The assumption of LTE allows the mathematical apparatus applied for the determination of plasma properties to be considerably simplified, because, in this case, every property in an elementary volume is a function of only two thermodynamic parameters, for instance, the temperature T and the particle density. In particular, the population numbers for excited levels in plasma-forming particles are determined by the Boltzmann equation

$$n_k^0 = n_i^0 (g_k/g_i) \exp(-\Delta E_{ik}/T), \tag{1}$$

where n_k^0 and n_i^0 are the equilibrium atomic densities in the upper, k, and lower, i, states, respectively; g_k and g_i are the corresponding statistical weights of those levels; and ΔE_{ik} is the excitation energy.

The detailed balancing principle is a key one for an equilibrium system: every elementary process is precisely counterbalanced by the inverse process [2, 3]. For example, the population of excited energy states in an atom that collides with electrons in plasma is coordinated with the inverse process of atomic level deactivation at inelastic collisions of the same particles [4]. In this case, the detailed balancing principle looks like

$$n_k^0 \omega_{ki} = n_i^0 \omega_{ik}, \tag{2}$$

where ω_{ki} and ω_{ik} are the frequencies of the atomic excitation and the deactivation, respectively, between those states. The frequency of excitation events is equal to

$$\omega_{ik} = N_e q_{ik} (kT/m_e)^{1/2} \exp(-\Delta E_{ik}/kT), \tag{3}$$

where N_e is the density of electrons, q_{ik} the average excitation cross-section, k the Boltzmann constant, T the temperature, and m_e the electron mass. The process is possible, if the kinetic energy of an electron exceeds the excitation energy $\Delta E_{ik.}$.

Strictly speaking, the thermodynamic equilibrium is inherent to closed systems only. For the electric-arc plasma, which is an open system at least with respect to radiation emission losses, the validity of the LTE model under ordinary assumptions can be explained by the circumstance that collision-induced processes that give rise to the population of every energy state considerably prevail over the radiative processes resulting in its deactivation. In other words, this assumption is suitable only for a dense plasma.

As a rule, the most difficult task is to hold the conditions for the LTE to be maintained at resonance transitions in plasma-forming particles. Really, from the Saha equation of state, it follows that the ionization maximum for particles of a certain kind and charge is reached at a characteristic temperature $T^* \sim 0.1E_i$, where E_i is the ionization potential for those particles. On the other hand, the energy of resonance level excitation ΔE_{0r} amounts to $(0.7 \div 0.8)E_i$ for most gases and to $(0.4 \div 0.5)E_i$ for most metals used in plasma technologies [5]. Therefore, with regard for the Maxwellian character of the electron distribution in plasma over velocities, there are a relatively insignificant number of electrons at the temperature T^*, the energy of which is sufficient for an atom to be excited onto the resonance level from the ground one (see Eq. (3)). On the contrary, the intensity of resonance lines is the highest, as a rule, because the intensity ε of a radiation line is governed by the population number n_k for the top level in the corresponding spectral transition,

$$\varepsilon = A_{ki} g_k n_k / \lambda_{ki}, \tag{4}$$

where A_{ki} is the probability of the spectral λ_{ki}-transition. Those population numbers, as well as the A_{ki}-values, are the highest, if the resonance transition occurs at the temperature T^*.

As a result of the unbalanced action by those factors, the electron density N_e^*, at which the LTE state is reached in an optically thin plasma (e.g., hydrogen plasma) turns out too high (namely, of about 10^{18} cm^{-3}) to be realized in the majority of practical cases.

However, for an electric arc at the atmospheric pressure, the characteristic path length $\langle l \rangle$ of resonance photons in the maximum of the radiation line, as a rule, turns out much shorter than the typical electric arc radius. For its estimation, we can use the relation

$$\langle l \rangle = \kappa_0^{-1}, \tag{5}$$

where κ_0 is the absorption coefficient at the center of the spectral line. It is determined by the population number of the lower level, n_i, as follows:

$$\kappa_0 = (1 - g_i n_k / g_k n_i) p f_{ik} \lambda_{ki}^2 n_i / \Delta\lambda. \tag{6}$$

Here, p is a numerical coefficient, f the oscillator strength for the corresponding transition, and $\Delta\lambda$ the half-height width of the spectral-line contour. The parenthesized multiplier describes the induced radiation emission, which has to be taken into account, if the top level is occupied (as a rule, in earlier works dealing with radiation transfer [4, 6, 7], this multiplier was neglected). If λ and $\Delta\lambda$ are measured in nanometers and n in cm^{-3} units, the numerical value of p in formula (6) amounts to 8.19×10^{-20} for the Gaussian contour and to 5.64×10^{-20} for the Lorentzian one [8]. The dimensionless quantity f is proportional to the transition probability,

$$A_{ki} = 6.66 \times 10^{13} g_i f_{ik} / \left(g_k \lambda_{ki}^2\right), \tag{7}$$

where A is expressed in terms of s^{-1} units, and λ in nanometers. According to Eq. (4), the path length $\langle l \rangle$ of resonance radiation emission at the characteristic temperature T^* is of the order of 10^{-4} cm in the atmospheric-pressure plasma in hydrogen, helium, nitrogen, and argon, and about 10^{-2} cm for the resonance lines of a copper atom at 324.7 and 327.4 nm emitted in the copper plasma at the copper vapor content of 1% [5]. The self-absorption of radiation effectively reduces the role of the radiative deactivation of excited levels and, respectively, lowers the threshold of the electron density, N_e^*, at which the LTE state is achieved in the optically dense uniform plasma. Numerically, the latter is characterized by the introduction of the effective probability of radiative transition A_{r0}^*, which takes the self-absorption of radiation in plasma into account,

$$A_{r0}^* = A_{r0} \theta(r), \tag{8}$$

where A_{r0} is the probability of the resonance radiative transition, and $\theta(r)$ is the probability for a resonance photon at the point r to go beyond the plasma limits without being absorbed.

The crucial influence of the resonance transition on the equilibrium state in plasma is responsible for a wide application of the so-called two-level model of atom with two energy levels, the ground (1) and excited (2) ones. In the stationary case, the balance of the population at the excited level in this model can be presented in the form

$$n_1 \omega_{12} = n_2 \omega_{21} + n_2 A_{21}^*. \tag{9}$$

Generally speaking, deviations from the equilibrium value are convenient to be described in terms of the relative level population [4],

$$y_k = n_k / n_k^0. \tag{10}$$

Substituting expressions (2) and (10) into Eq. (9), we obtain the basic relation

$$y_1 = y_2 (1 + A_{21}^* / \omega_{21}). \tag{11}$$

Evidently, the criterion of relative equilibrium between two states (i.e. $y_k \approx y_l$) or, in the case of two-level model, $y_2 \approx y_1 \approx 1$ is the inequality

$$A^*_{21}/\omega_{21} \ll 1. \tag{12}$$

In other words, the collision processes that restore the equilibrium state must prevail over the radiative processes that violate it.

Below, we extend the ideology of the two-level model in order to consider the energy transition not only from the resonance onto the ground level, but also onto a metastable one, in such a manner actually examining a three-level system. However, in the framework of the criterion approach, where insignificant deviations from the equilibrium state are dealt with, it is quite a feasible task. The only difference consists in that, in this case, the population number for the lower level in the spectral transition between the resonance and metastable energy levels is to be determined from Boltzmann distribution (1), because no physical reason is contained in the adopted assumptions for the spectral transition to be nonequilibrium. In what follows, we use the indices 0, m, and r, when considering a three-level system, and the numerical indices 1 and 2, when considering a general two-level one.

We cannot assume that the influence of radiative transitions between the resonance and metastable levels is crucial for deviations of the system from the LTE state. However, the corresponding spectral lines are located in the visible spectral range, being well accessible for research purposes. In particular, one of those lines, at 510.5 nm, is among the most popular ones in the optical diagnostics of the copper electric-arc plasma [5, 9].

Criterion approaches aimed at detecting the LTE state in plasma on the basis of a radiation-loss analysis were introduced as long ago as by Griem (see Chapter 6 in book [10]). For the LTE to be maintained to within 10%, the rates of processes in relation (12) must differ by an order of magnitude.

Actually, all that was discussed above concerning the role of the radiation self-absorption was associated with a temperature-uniform plasma. However, in case of an inhomogeneous plasma, the radiation self-absorption can result in the inverse effect. Really, radiation emitted in hotter plasma regions and absorbed at a certain point of observation $r > 0$ is able not only to compensate the radiation energy losses, but also to stimulate the inverse effect: at this point, the population number for the excited level is higher than that for the equilibrium one. The corresponding limit is the population number obtained for the temperature at the arc axis.

A nonequilibrium state in the plasma of a freely supported electric arc between fusible copper electrodes under environmental conditions, which was obtained as a result of the resonance radiation transfer of copper atoms, was experimentally observed in work [9]. Later, a simple model for such a nonequilibrium state was proposed, which consisted in that the population of the resonance level of a copper atom along the electric arc radius was supposed to correspond to the temperature inherent to the arc-axis region (in other words, it corresponded to an overpopulation of the resonance level at the electric arc periphery, where the local temperature is lower than that on the axis) [11]. Its analog at the macro-level is the fireplace effect, i.e., the heating up of objects in a cold room owing to the absorption of emitted thermal radiation [12].

As follows from the aforesaid (see Eqs. (3), (4) and involved text), the non-resonance radiation is not a substantial factor for the effects associated with the radiation transfer. Addressing once more analogies at the macro-level, the radiation emission from higher levels can be confronted with the candle effect: the candle shines, but does not warm.

As was shown in works [11–13], in the framework of this assumption, the effect of arc-channel "enlightenment", i.e. a reduction of its resistance to the electric current, can be obtained. The physical origin of the phenomenon lies in a reduction of the ionization potential for plasma-forming atoms at the electric arc periphery owing to the overpopulation of atomic resonance levels in this region. Therefore, from the viewpoint of further practical applications, it is important that the phenomenon of the radiation transfer in plasma and its influence on a deviation of plasma from the equilibrium state should be taken into account rigorously. This work aimed at developing this direction of researches dealing with the electric-arc plasma.

Below, we report the results of our calculations, which allow the role of radiation processes in a deviation of the dense low-temperature plasma from the equilibrium state to be estimated in principle. At this stage of consideration, the solution of the problem is obtained in the form of a criterion that the LTE model is applicable, which involves the processes of radiation emission and radiation losses in plasma. Such a formulation makes the problem somewhat simpler, because, when seeking for the solution analogously to what was done in work [10], it allows the consideration to be confined by making the assumption that the plasma is in the equilibrium state.

An objective shortcoming of this approach is an indicative character of the result obtained, which only detects a deviation of plasma from the equilibrium state, but does not allow the magnitude of this deviation to be determined quantitatively.

2. Formulation of the Problem and the Solution Procedure

In contrast to the mass transfer, when the thermodynamic parameters of a substance change insignificantly at lengths of the order of the mean free path for a particle of this substance, the account of the processes of radiation transfer gives rise to considerable difficulties associated with a drastic dependence of the photon mean free path on the photon frequency. Really, expression ([5]) was written down for the center of a spectral line with frequency ν_0. However, for the frequencies beyond the line limits, the local values of absorption factor $\kappa = f(\nu-\nu_0)$ expand over the whole interval $0 < \kappa < \kappa_0$. They correspond to the growth of local $\langle l\rangle$-values in accordance with formula (5). Therefore, a considerable mutual effect between even rather remote elementary plasma volumes and, moreover, the radiation absorption outside of the electric arc channel become feasible, if one takes into account that there are always such $\langle l\rangle$-values, which are comparable with the distance between those volumes (of course, considering the solid angle, in which the mentioned radiation propagates or, in other words, the distance between the mentioned elementary volumes). Generally speaking this self-absorption is the reason that induces the overpopulation at the excited levels of copper atoms located beyond the arc channel in the experiment [9].

It is impossible to introduce the concept of characteristic path length for photons. That is why, the diffusion approximation cannot be applied, whereas differential relations are not sufficient for the mathematical description of radiation transfer processes to be adequate. Therefore, it is necessary that the integral equations, which would take the mutual influence of processes over the whole plasma volume into account, should be engaged.

The dynamics of the population number $n_2(r,\,t)$ at the resonance level in the two-level atomic model is governed by transitions into the ground state and from it. Considering also the processes of excitation radiation transfer, the corresponding equation looks like [4]

$$\frac{\partial n_2(r,\,t)}{\partial t} = -n_2(r,\,t)A_{21} - n_2(r,\,t)\omega_{21}+$$

$$+n_1\omega_{12} + \int\limits_V n_2(r',\,t)A_{21}K\left(|r-r'|\right)dr'. \quad (13)$$

Here, the integral term makes allowance for the radiation transfer, whereas the kernel $K\left(|r-r'|\right)$ corresponds to the probability that the resonance photon emitted from an arbitrary point r' is absorbed in a volume with the coordinate r:

$$K(\rho) = -\frac{1}{4\pi\rho^2}\frac{df(\rho)}{d\rho}, \quad \rho \equiv |r-r'|\,. \quad (14)$$

The multiplier $(4\pi\rho^2)^{-1}$ selects photons that propagate from dr' towards dr, (in other words, it determines the solid angle), and $f(\rho)$ is the probability for a particle to pass the distance ρ and not to be absorbed or scattered,

$$f(\rho) = \int \varepsilon_\nu \exp(-k_\nu\rho)d\nu, \quad (15)$$

where ε_ν is the distribution of photons over the frequencies, normalized to 1, and k_ν is the spectral absorption factor. The ε_ν-distribution is determined by the shape of the radiation-line contour, whereas the radiation line intensity depends on the population at the top level, which results in the appearance of the factor n_2 in the integrand in expression (13). The specific form of $f(\rho)$ depends on the shapes of the absorption and radiation lines, i.e. on k_ν and ε_ν. However, in view of the circumstances mentioned above, this function is not exponential, as it was for the mass transport processes, and falls down considerably more slowly with the growth of ρ [4]. In Eq. (14), the multiplier $df(\rho)/d\rho$ describes the attenuation of a photon beam on its way from point r' to point r and the probability that photons, when having reached dr, are absorbed.

It is worth emphasizing that the formula for $f(\rho)$ expressed in the form (15) is a substantial simplification for the plasma medium, because, actually, it assumes that the spectral absorption factor k_ν is constant. In essence, it is a function of the radial coordinate; it is especially true for a non-uniform electric-arc plasma.

Differentiating in formula (14), we obtain

$$K(\rho) = (4\pi\rho^2)^{-1}\int \varepsilon_\nu k_\nu \exp(-k_\nu\rho)dv. \quad (16)$$

Although, according to the asymptotic behavior of the probability $f(\rho)$, the function $K(\rho)$ falls down slowly, its integral over the infinite volume equals 1. This fact follows from the physical sense of the problem.

In the case of stationary processes, it is convenient to write down Eq. (13) with regard for relation (10) for the reduced population of the excited state. We have

$$y_2(r) = (1+\beta)^{-1}\int\limits_V y_2(r')K(|r-r'|)\,dr' + \beta/(1+\beta). \quad (17)$$

Here, we introduced the notation

$$\beta = \omega_{21}/A_{21} \quad (18)$$

and used the relation between the quantities ω_{12} and ω_{21}, which follows from the detailed balancing principle (Eq. (2)). We also adopted that the value of n_2^0 does not depend on coordinates. The latter assumption is a substantial simplification of the problem under consideration, but, at the same time, it is rather a grounded approximation. Actually, it consists in that the value n_1 is determined not from the relation $n_1 \approx N_a \sim T^{-1}$, where the quantity N_a, in turn, is determined from the following equation for the partial pressure of copper vapor,

$$[N_a + (1 + x_{\mathrm{Cu}})N_e]kT = x_{\mathrm{Cu}}p, \quad (19)$$

where p is the atmospheric pressure, and x_{Cu} the content of copper vapor in the plasma-forming mixture, but from a formula $n_1 \sim n_2^0 \exp(\Delta E_{12}/T) = \mathrm{const}$ which corresponds to the Boltzmann equation. However, its validity for our approximate calculations stems from the mean-value theorem for definite integrals.

To find an analytical solution for the equation of radiation transfer is a complicated problem. In those rare cases where analytical solutions were managed to be obtained, they turned out rather cumbersome [6]. Therefore, numerical methods play an important role, while solving such problems.

L.M. Biberman [7] proposed a simple approximate method for the solution of Eq. (17). Namely, the quantity that characterizes a local violation of the equilibrium state is assumed to vary weakly enough, even if the dependence of $n_2(r)$ on the radial coordinate is strong. It is an additional approximation, but enables the function $y_2(r')$, by assigning it the value

$$y_2(r') = y_2(r) \quad (20)$$

to be taken outside the integral sign. In this case, Eq. (17) becomes an algebraic one, which can be used to obtain the following approximate results (we denote them, by using the tilde sign):

$$\tilde{y}_2(r) = \frac{\beta(r)}{\theta(r) + \beta(r)}, \quad (21a)$$

$$\tilde{n}_2(r) = n_1\omega_{12}/(\omega_{21} + A_{21}\theta(r)), \quad (21b)$$

$$\theta(r) = 1 - \int\limits_V K(|r-r'|)\,dV'. \quad (21c)$$

The latter of those expressions describes the probability for a photon starting from point r to escape, not being absorbed, beyond the plasma boundaries (this quantity was introduced in relation (8)). Here, the integration is carried out over the whole arc volume, and its result indicates which part of radiation was absorbed in this volume. Hence, the influence of finite optical-density values is taken into account through the effective probability of spontaneous radiation A_{21}^*. A reduction of this quantity gives rise, naturally, to an increase of the reciprocal quantity $\tau_{\mathrm{eff}} = (A_{21}^*)^{-1}$, which is called the effective lifetime of the excited level. Therefore, this approximation is often referred to as the method of effective lifetime.

This method has been used successfully (see work [4]) to obtain solutions for a number of problems dealing with a temperature-uniform plasma, for which expressions (15) and, respectively, (16) are valid. In such a version, $0 \le \theta(r) < 1$, which completely satisfies the requirements imposed onto the variable describing the probability.

Let the coordinate dependence of the absorption factor κ_ν be associated only with the number of absorbing atoms, provided that the contours of spectral lines do not change, i.e. $\kappa_\nu = an_1(r)$, where a is a proportionality coefficient. Then the optical thickness can be introduced into consideration for its application in Eq. (13),

$$\int\limits_{\mathrm{r}}^{\mathrm{r}'} k_\nu(\mathrm{r}'')dr'' = t' - t, \quad dt = an_1(\bar{r})d\bar{r} \quad (22)$$

as a new coordinate system, in which the method of effective lifetime remains valid. However, this problem does not allow one to reveal specific effects in plasma, which would stem from the discrepancy between the characteristic path lengths of resonance radiation, which was mentioned above.

In contrast, we seek for the solution of problem (13) in the case of inhomogeneous media. The general form of the solution, Eqs. (21), remains correct at that. However, in expression (21c), we will consider such a kernel $K(r', r)$ that would include the integration of the self-absorption along every beam:

$$K(r', r) = \frac{1}{4\pi}\int\limits_0^\infty \frac{k_\nu(r')\varepsilon_\nu(r')}{|\mathrm{r}-\mathrm{r}'|^2}\exp[-\int\limits_{\mathrm{r}}^{\mathrm{r}'} k_\nu(\mathrm{r}'')dl]d\nu. \quad (23)$$

Here, the exponent includes the contour integral along the beam that connects the r- and r'-points. We also assume that the plasma is contained in a long cylindrical volume with radius R.

Considering the dependence of the line absorption factor in the integral kernel (23) on the temperature, which is a function of coordinates, the quantity $\theta(r)$ does not possess, in contrast to work [4], a simple probabilistic meaning. Now, it can acquire both positive and negative values [6]. In our case, this parameter makes allowance for the influence of two probabilistic processes, which exert opposite effects on the energy level population in a local plasma volume, namely, a reduction of the population owing to radiation losses from this volume and a growth of the population owing to the absorption of radiation from other plasma regions, if they have higher temperatures. Nevertheless, the quantity $A_{21}\theta(r)$ allows one to obtain an approximate value for the photon flux divergence at a point.

It follows from Eq. (21a) that, for the quantity $y_2(r)$ to be close to 1, the inequality

$$|\theta(r)/\beta(r)| \ll 1 \qquad (24)$$

must be satisfied, i.e. it is an LTE criterion, which is intrinsically consistent with assumption (20). In accordance with the results of work [6], this LTE condition gives rise to a correct result, if it is obeyed in the whole volume occupied by plasma. In view of relations (8) and (18), criterion (24) is a generalization of condition (12) for the LTE to take place in plasma, which considers only radiation losses. The generalization can be considered equivalent as far as the contents of the quantity $\theta(r)$ can be regarded equivalent in the cases of radiation losses and transfer.

In order to calculate the numerical value of the integral in expression (21c), it is convenient to pass to the local spherical coordinate system connected with the point of observation r,

$$I = \frac{1}{4\pi}\int\limits_0^\infty \iiint\limits_V \frac{k_\nu(r')\varepsilon_\nu(r')}{|\mathrm{r}-\mathrm{r}'|^2}\exp[-\int\limits_{\mathrm{r}}^{\mathrm{r}'} k_\nu(\mathrm{r}'')dl]dV'd\nu =$$

$$= \frac{1}{4\pi}\int\limits_0^\infty\int\limits_0^{2\pi}\int\limits_0^{\pi}\int\limits_0^{R(\varphi)} \frac{k_\nu(\rho)\varepsilon_\nu(\rho)}{\rho^2}\times$$

$$\times\exp[-\int\limits_0^\rho k_\nu(t)dt]\rho^2\sin(\theta)\,d\rho\,d\theta\,d\varphi\,d\nu, \qquad (25)$$

where $I = \int\limits_V K(r',r)dV'$, $\rho = |\mathbf{r}-\mathbf{r}'|$, and $0 \le \rho < \infty$. On the basis of the symmetry of the problem, let us expand the limits of integration over the frequency and make the change of the variable $\omega = (\nu-\nu_0)/\Delta\nu(r)$, so that

$$I = \frac{1}{\pi}\int\limits_{-\infty}^\infty\int\limits_0^{\pi}\int\limits_0^{\pi/2}\int\limits_0^{R(\varphi)} k_\nu(\rho)\varepsilon_\nu(\rho)\times$$

$$\times\exp[-\int\limits_0^\rho k_\nu(t)dt]\sin(\theta)\,d\rho\,d\theta\,d\varphi\,d\omega. \qquad (26)$$

Consecutively changing the variables, namely, $\rho = \rho'/\sin(\theta)$ and $\rho' = \rho''/\sin(\theta)$, this integral is transformed to the form

$$I = \frac{1}{\pi}\int\limits_{-\infty}^\infty\int\limits_0^{\pi}\int\limits_0^{\pi/2}\int\limits_0^{r_0(\varphi)} k_\nu(\rho)\varepsilon_\nu(\rho)\times$$

$$\times\frac{\exp[-\int_0^{\rho''} k_\nu(t)dt/\sin^2(\theta)]}{\sin(\theta)}\,d\rho''\,d\theta\,d\varphi\,d\omega, \qquad (27)$$

where the function $r_0(\varphi)$ is determined from the relation $R^2 = r^2 + r_0^2(\varphi) - 2rr_0(\varphi)\cos(\pi-\varphi)$, and R is the arc radius. In essence, the quantity $r_0(\varphi)$ is a projection of a radius-vector, which connects the point r with a point on the internal surface of the cylinder, onto the polar plane ($\theta = \pi/2$). In view of the integral representation for the modified Bessel function of the third kind of imaginary order (the Macdonald function),

$$\int\limits_0^{\pi/2}\frac{\exp[-z/\sin^2(\theta)]}{\sin(\theta)}\,d\theta = \frac{1}{2}\exp\left(-\frac{z}{2}\right)K_0\left(\frac{z}{2}\right),$$

we ultimately obtain

$$I = \frac{1}{2\pi}\int\limits_{-\infty}^\infty\int\limits_0^{\pi}\int\limits_0^{r_0(\varphi)} k_\nu(\rho)\varepsilon_\nu(\rho)\times$$

$$\times\exp\left(-\frac{Z}{2}\right)K_0\left(\frac{Z}{2}\right)\,d\rho''\,d\varphi\,d\omega, \qquad (28)$$

where

$$Z = \int\limits_0^{\rho''} k_\nu(t)dt. \qquad (29)$$

With regard for the divergence of the Macdonald function in a vicinity of its zero argument, the difference of the function $K_0\,(z)$ and it asymptote expression $K_0(t) \approx -\ln\left(\frac{t}{2}\right) - \frac{\gamma}{2}$, where γ is the Euler constant, were integrated. As the last integral has analytic expression, the value I from Eq. (28) is calculated as a sum of the analytic expression and the numerical value of difference of these integrals. The trapezium method was applied to integrate with a uniform mesh.

3. Spectral Characteristics of Plasma

The list of spectral lines emitted by a copper atom from both resonance levels, $E_k = 3.79$ and 3.82 eV, and their spectroscopic parameters are presented in the table. Some of those lines have the ground level as the lower level of a spectral transition, whereas the others the metastable levels $E_i = 1.39$ and 1.64 eV. To estimate the role of the radiation transfer effects in a deviation of plasma from the LTE state, the researches were carried out for a pair of such spectral lines at 327.3 and 510.5 nm. In the qualitative aspect, the results of our researches can be extended to include other groups of resonance lines, because the oscillator strengths for them do not differ considerably from one another.

The temperature in an electric arc changes in a wide interval, ranging from the plasma one to the temperature of the internal surface of a stabilizing wall or to the environmental one, if the arc is supported in atmospheric air. Accordingly, the local radiative (4) and absorptive (6) properties of plasma are substantially different along the arc radius. As a result, the different mechanisms of spectral line broadening prevail in each of those cases. Two main groups of spectral line contours are typical of those mechanisms [8]: the Gaussian contour (it corresponds to the Doppler effect),

$$\varepsilon_\nu(r) = \varepsilon_0^{\rm D}(r)p\exp[-(\Delta\nu/\Delta\nu^{\rm D}(r))^2], \quad (30a)$$

$$\kappa_\nu(r) = \kappa_0^{\rm D}(r)\exp[-(\Delta\nu/\Delta\nu^{\rm D}(r))^2], \quad (30b)$$

Resonance spectral lines of a copper atom and their spectroscopic parameters

Line, nm	E_k, eV	g_k	E_i, eV	g_i	$\Delta\lambda_s^*$, nm (at $N_e = 10^{17}$ cm^{-3}) [15]	f
324.7	3.82	4	0	2		0.430
510.5	3.82	4	1.39	6	0.021	0.0051
570.0	3.82	4	1.64	4	0.026	0.0011
327.3	3.79	2	0	2		0.220
578.2	3.79	2	1.64	4	0.027	0.0042

and the dispersion (or Lorentzian) one,

$$\varepsilon_\nu(r) = \varepsilon_0^{\rm L}(r)p[1 + (\Delta\nu/\Delta\nu^{\rm L}(r))^2]^{-1}, \quad (31a)$$

$$\kappa_\nu(r) = \kappa_0^{\rm L}(r)[1 + (\Delta\nu/\Delta\nu^{\rm L}(r))^2]^{-1}, \quad (31b)$$

where $\Delta\nu = \nu - \nu_0$; $\varepsilon_0^{L,D}(r)$ and $k_0^{L,D}(r)$ are the emissive capacity and the absorption factor, respectively, at the line center; and $\Delta\nu^{L,D}(r)$ are the widths of the radiation and absorption lines for the Lorentzian and Gaussian contours, respectively. The values of coefficient p in formulas (30a) and (31a) are determined from the condition of their normalization to value (4), when being integrated over the frequency. The corresponding values obtained insignificantly differ from each other for different contours. For this reason, the p-values in expressions for $k_0^{L,D}(r)$ are also different, in accordance with formula (6).

In the practical spectroscopy, such a scale of radiation wavelengths is usually used, in which the following relation is valid for the line width:

$$\Delta\lambda = (c/\nu^2)\Delta\nu. \quad (32)$$

The Doppler contour is governed by the thermal motion of atoms. Its width is

$$\Delta\lambda^{\rm D} = 7.16 \times 10^{-7}\lambda_0\sqrt{T/\mu}, \quad (33)$$

where μ is the atomic weight, and T is the Kelvin temperature. For $\mu \approx 64$ and $\lambda_0 = 500$ nm, the value of $\Delta\lambda^D$ is about 0.77×10^{-3} nm at $T = 300$ K and about 3.7×10^{-3} nm at $T = 7000$ K.

The Lorentzian contour of a spectral line describes both its natural broadening and the broadening induced by collision processes. The former is associated with a finite lifetimes τ_k and τ_i of the atomic energy states of the transition, at which the spectral line is emitted. For every of the lines emitted from the resonance levels, this time, τ_r, is determined by the sum of probabilities of spontaneous transitions from the given resonance level onto the ground and metastable (including the case where there are several of them) levels,

$$\tau_r^{-1} = \tau_{r0}^{-1} + \tau_{rm1}^{-1} + \tau_{rm2}^{-1} + \ldots = A_{r0} + A_{rm1} + A_{rm2+\ldots}. \quad (34)$$

The corresponding line broadening reads

$$\Delta\lambda_n^L = \lambda_0^2/(2\pi\tau_r c) = (A_{r0} + A_{rm1} + A_{rm2+\ldots})\lambda_0^2/(2\pi c). \quad (35)$$

For the spectral lines at 510.5 and 324.7 nm, which are emitted from the same resonance level of a copper atom with $E_r = 3.82$ eV (see the Table), $\Delta\lambda_n^L$ amounts to 0.5×10^{-5} nm, which is smaller by one or two orders of magnitude than the Doppler width.

The broadening of spectral lines owing to collision processes is associated with a violation of the line monochromaticity as a result of collisions of the emitting particles with the neutral or charged plasma component. In the former case [8],

$$\Delta\lambda_g^L = 6.6 \times 10^5 \sigma^2 p \lambda_0^2 (\mu T)^{-1/2}, \tag{36}$$

where σ^2 is the effective cross-section of collisions (cm^2), p the pressure (mm Hg), and λ_0 is expressed in nanometers. At the pressure $p = 1$ atm, $\sigma = 5 \times 10^{-8}$ cm^2, $\mu \approx 30$, and $T = 1000$ K, the $\Delta\lambda_g^L$-value is about 2×10^{-3} nm. It is much larger than the natural width and is comparable with the maximal Doppler width.

At last, the Stark mechanism [14] gives the largest contribution to the line broadening owing to collision processes, if it is related to the density of particles that are responsible for collisions. In this case, we selected the data given by R. Konjevic and N. Konjevic [15] for the broadening parameters of those resonance lines for a copper atom, the lower level of which is metastable (see table). Earlier [15], we have carefully analyzed the publications that included this parameter for another spectral line, at 515.3 nm, which is widely applied in the copper plasma diagnostics. On the basis of the results obtained while comparing accessible data (the relevant values differ from one another by an order of magnitude) with our own experimental results, we chose the data of work [15]. Here, we also use them, but for the 510.5-nm line, as such that were obtained in the framework of the same methodological basis. They are normalized to the electron density in plasma $N_e = 10^{17}$ cm^{-3}, so that the real effect can be evaluated according to the expression

$$\Delta\lambda_s^L = 10^{-17} N_e \Delta\lambda_s^*, \tag{37}$$

where the quantity N_e is expressed in cm^{-3}, and $\Delta\lambda$ in nanometers. The other data were taken from handbook [17]. It is worth emphasizing that the deeper the energy levels are located in the atomic structure, the more they are screened from the influence of external electric fields, and the less the Stark broadening effect for them. For the lines emitted from the upper excited states of a copper atom, the values of broadening parameter $\Delta\lambda_s^*$ are larger by an order of magnitude [5]. On the contrary, for the resonance lines corresponding to the transitions onto the ground state, the Doppler mechanism of line broadening turns out sufficient, as a rule.

The result of the mutual action by several dispersion mechanisms of line broadening is characterized by a Lorentzian contour, the total width of which is determined by a sum of individual terms,

$$\Delta\lambda^L = \Delta\lambda_n^L + \Delta\lambda_g^L + \Delta\lambda_s^L. \tag{38}$$

The result of the combined action by line broadening mechanisms, one of which is described by the Gaussian contour and the other by the Lorentzian one, is determined by the convolution operation [8, 14]. It is the so-called Voigt contour, which has a special conventional notation, H:

$$H(a,u) = \frac{a}{\pi} \int_{-\infty}^{+\infty} \frac{\exp(-y^2)}{a^2 + (u-y)^2} dy, \tag{39a}$$

$$a = (\ln 2)^{1/2} \Delta\nu^{\mathrm{L}} / \Delta\nu^{\mathrm{D}}, \tag{39b}$$

$$u = 2(\ln 2)^{1/2} (\nu - \nu_0) / \Delta\nu^{\mathrm{D}}. \tag{39c}$$

Its features have been described in sufficient details in the literature (e.g., see p. 15 in [8]). The most important of them consists in that the central part of the contour (around its maximum) mainly corresponds to the Doppler contour, while its "wings" to the Lorentzian one. In all numerical calculations, the integrated influence of spectral line broadening was taken into account in accordance with the last relation.

4. Simulation of Properties of an Electric Arc

For calculations, we used a temperature profile obtained from the solution, in the one-dimensional approximation, of the equation of energy balance for a cylindrical wall-stabilized arc (the Elenbaas–Heller equation) [18, 19],

$$\frac{1}{r}\frac{d}{dr}\left(r\frac{dS}{dr}\right) + \sigma E^2 = 0, \quad S = \int_0^T \lambda(T) dT, \tag{40}$$

where r is the radial coordinate, $\sigma(T)$ the electroconductivity coefficient, E the electric field strength, S the heat potential, and $\lambda(T)$ the heat conductivity. The corresponding boundary conditions are

$$ds/dr_{(r=0)} = 0; \qquad S_{(r=rw)} = S_w, \tag{41}$$

where the parameter S_w characterizes the temperature T_w of a cooling quasiwall. Its introduction allows the arc to be simulated assuming the one-dimensional axial symmetry of the problem [20].

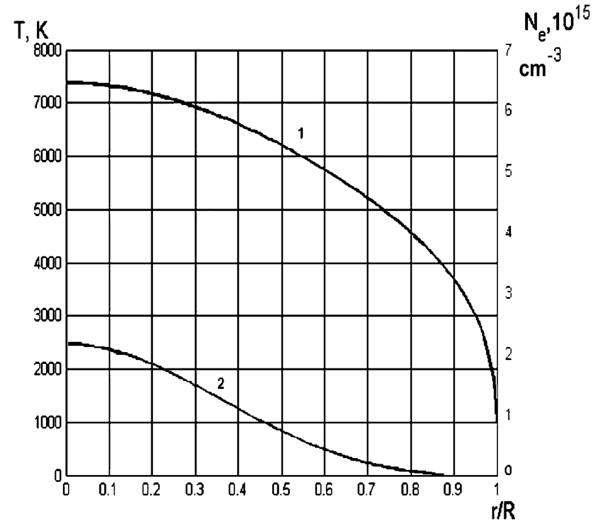

Fig. 1. Radial distributions of the temperature (curve *1*) and the electron density (curve *2*) in an electric arc with the copper content $x_{\mathrm{Cu}} = 1\%$

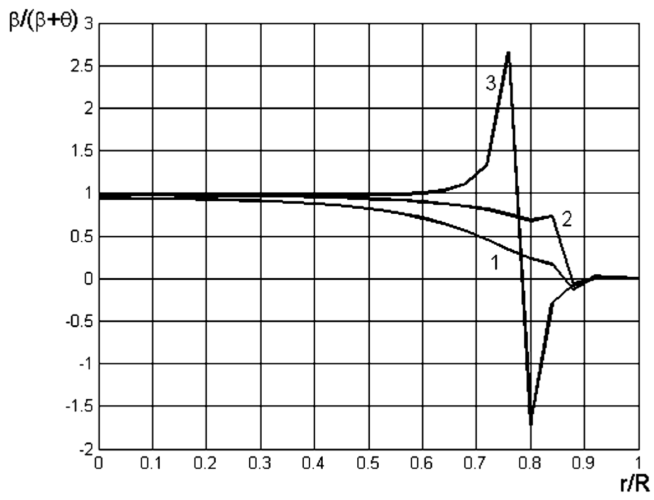

Fig. 2. Criterion estimations of conditions for a deviation of the plasma from the equilibrium state owing to the radiation transfer for the 327.3-nm resonance line in electric arcs with various copper contents $x_{\mathrm{Cu}} = 0.1$ (*1*), 1 (*2*), and 10% (*3*)

The numerical solution of the nonlinear boundary-value problem (Eqs. (40) and (41)) is determined using the parameter continuation method [21]. At every step of calculations, a linearized differential equation of the second order is solved by reducing the boundary-value problem to a sequence of Cauchy problems. The latter, it turn, are solved using the Dormand–Prince method [22] to the fifth order of accuracy. The electric current and the absorbing quasiwall radius are supposed to be known. The analytical solution obtained for the quasichannel model of electric arc [11, 18] was used as the first approximation.

The electric and heat conductivities of a copper–nitrogen mixture are approximated in accordance with the results of works [11, 23] as follows:

$$\sigma = \sigma_0 S^{n_\sigma}, \quad \lambda = \lambda_w \left(T/T_w\right)^{n_\lambda} . \tag{42}$$

At $x_{\mathrm{Cu}} = 1\%$, the approximation coefficients are $\sigma_0 = 5.9 \times 10^{-8}$ $(\mathrm{W{\cdot}m})^{k\sigma}/(\Omega \cdot \mathrm{m})$, $n_\sigma = 17/7$, $n_\lambda = 5/2$, and $\lambda_w = 0.066$ W/(m · K) at $T_w = 1000$ K.

In view of the estimation character of our calculations, the plasma state was described by the Saha equation for the first ionization level,

$$\frac{N_e^2}{N_a} = 2\left(\frac{2\pi m_e kT}{h^2}\right)^{3/2} \frac{\Sigma_i}{\Sigma_a} \exp\left(-\frac{E_i}{kT}\right), \tag{43}$$

where Σ_a and Σ_i are the partition functions for an atom and an ion, respectively; and $E_i = 7.73$ eV is the ionization potential for a copper atom. This equation should be supplemented by an equation for the partial pressure of copper vapor ([19]), which takes into account that the air component is actually adopted to be inert.

For the numerical integration of the internal integral in the contour integral (29), as well as for the determination of the Voigt contour for spectral lines (39a), the trapezium method was applied. The functional dependence of the plasma temperature on the radius was determined for the points of integration by carrying out the linear interpolation of the temperature profile, which was found as a solution of the boundary-value problem (40), (41).

5. Results of Calculations

Numerical calculations were carried out for an atmospheric electric arc burning between melting copper electrodes. The discharge current was 30 A. The radius of a cooling quasiwall was $R_w = 3$ mm, and its temperature was $T_w = 1000$ K. In Fig. 1, the calculated temperature profile $T(r)$ in this arc and the corresponding distribution of the electron density $N_e(r)$ obtained by solving the equation of energy balance (40) for a wall-stabilized cylindrical arc in the one-dimensional approximation are shown.

The criterion estimations of conditions for the plasma to deviate from the equilibrium state in such an arc as a result of the radiation transfer for the 327.3-nm resonance line, the lower transition level for which is the ground level of copper atoms, are depicted in Fig. 2. The figure demonstrates (see curve *2*) that the parameter $\beta(r)/[\theta(r) + \beta(r)]$ for this line reveals a drastic evolution along the arc radius. In particular, it is close to 1 in a vicinity of the arc axis, which testifies that the radiation effects, if they take place here, maintain the LTE state in plasma. On the contrary, in the peripheral region of the arc, this parameter is negative, being almost equal to zero. This fact testifies that, in this region, the radiation influence represented by a term in the denominator becomes prevailing. This circumstance indicates that a considerable absorption of resonance spectral lines should be expected here, which results in a plasma deviation from the LTE state [24, 25].

To specify the character of the transition between the above-mentioned regions (for $r/R = 0.8 \div 0.9$), we varied the copper content in this arc. Curves *1* and *3* in Fig. 2 correspond to $x_{\rm Cu} = 0.1$ and 10%, respectively. In the former case, a reduction of the electron density at the arc axis, $N_e(r = 0)$, to 0.7×10^{15} cm^{-3} is observed. In the latter one, one can observe the electron density growth to 6.8×10^{15} cm^{-3}. (In this version, the problem is not absolutely self-consistent, because the temperature profile in the arc, presented in Fig. 1, was considered constant.) Accordingly, the populations at the excited levels of copper atoms change. The character of the transition between those regions at $x_{\rm Cu} = 10\%$ testifies that, in the region near $r/R \approx 0.8$, the effects of radiation losses and radiation absorption are compensated. The transition is associated with the vanishing of the denominator of the parameter $\beta(r)/[\theta(r)+\beta(r)]$ or with the transition "plus infinity–minus infinity" of this parameter itself.

On the contrary, for the 510.5-nm resonance line of copper atoms, for which the lower level of the spectral transition is metastable, the parameter $\beta(r)/[\theta(r)+\beta(r)]$ changes smoothly from 0.8 at $r/R = 0$ to zero at $r/R = 0.9$. Therefore, the result of calculations for this line does not demonstrate that there exists a region, in which the spectral line is substantially absorbed, although this effect was observed experimentally [5, 9]. This fact is evidently related to the simplifications adopted at the statement of the problem concerning the constant population of a resonance level along the arc radius, which ultimately results in a partial loss of the criterion approach sensitivity in this version. The cancellation of this approximation should expectedly bring about an improvement of the criterion approach sensitivity, but would make the problem considerably more complicated.

The results obtained give an opportunity to carry out the detailed calculations of the nonequilibrium plasma parameters considering the radiation transfer processes and including the kinetics of population of the metastable and resonance levels into the model concerned.

6. Conclusions

In this work, the preliminary calculations have been carried out, which allow the role of radiation emission processes in the deviation of plasma from the equilibrium state to be estimated in principle. Since the calculations are rather complicated, the solution of the problem was obtained as a criterion for the LTE-model applicability, by considering the role of the processes of radiation transfer and radiation losses in plasma. Such a formulation simplifies the problem to some extent, because it allows the consideration to be confined to the model, in which the equilibrium state of plasma is assumed.

The results obtained undoubtedly testify that the plasma is in a nonequilibrium state in a large part of the discharge channel volume in the atmospheric electric arc between fusible copper electrodes. The character of this nonequilibrium behavior corresponds to the overpopulation of resonance levels in plasma-forming atoms at the arc channel periphery owing to the absorption of resonance radiation emitted from the hot arc region located near the arc axis.

The results obtained evidence the ineligibility of the widely applied assumption concerning the presence of the LTE state in plasma produced in electric arcs between melting electrodes.

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ВПЛИВ ПЕРЕНЕСЕННЯ ВИПРОМІНЮВАННЯ НА ВІДХИЛЕННЯ ВІД РІВНОВАЖНОГО СТАНУ ЩІЛЬНОЇ ЕЛЕКТРОДУГОВОЇ ПЛАЗМИ: КРИТЕРІАЛЬНИЙ ПІДХІД

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Р е з ю м е

Принципово оцінено роль процесів випромінювання з урахуванням його перенесення щодо відхилення від рівноважного стану щільної електродугової плазми атмосферного тиску. Задачу розглянуто на прикладі електричної дуги циліндричної форми, стабілізованої стінкою. Розв'язок отримано у варіанті критерію застосовності припущення локальної термодинамічної рівноваги з урахуванням ролі процесів перенесення випромінювання у плазмі та його втрат. Дані числового моделювання доводять наявність ефектів відхилення від рівноважного розподілу заселення між резонансними та основним енергетичними рівнями атомів міді в умовах, що моделюють стан плазми в атмосферній електричній дузі між плавкими мідними електродами.