

# NONEQUILIBRIUM CHARGE CARRIERS IN THERMAL PLASMA — METAL CONTACT

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The space charge layer on the thermal plasma—metal interface is studied. It is shown that the presence of a volumetric charge gives the formation of nonequilibrium charge carriers. The presence of nonequilibrium charge carriers affects the potential of a dust grain a little if the grain is positive. However, for negative dust grains, the account of nonequilibrium carriers gives the diminution of a potential barrier on the interface by three times.

## 1. Introduction

Thermal plasma at atmospheric pressure consists of a buffer gas (usually air) and an easily ionizable addition agent of alkali-metal atoms with the number density  $n_A \sim 10^{16} \div 10^{23} \text{m}^{-3}$ . At the isothermal Kelvin temperature  $T \sim 1000 \div 3000 \text{K}$ , the easily ionizable addition agent becomes partially ionized by the collisions of gas particles, by forming the low-temperature plasma. Such a plasma, being in equilibrium, can be considered as an ideal gas approximation. Metal electrodes inserted into the low-temperature plasma or condensed phase formation leads to a displacement of the equilibrium state. However, if the inserted perturbations are small, it is still possible to consider the plasma as the ideal or weakly coupled plasma [1–3].

The formation of the charge on grains is accompanied by the exchange of electrons between the condensed phase and the plasma. As a result of this exchange, there is a space charge layer in the plasma that causes the formation of nonequilibrium charge carriers. In a weakly coupled plasma at small potential barriers  $e\phi_s \ll kT$ , this effect can be neglected.

If the charges of grains are large and  $e\phi_s \geq kT$ , the nonequilibrium charge carriers influence exchange processes and the charges of grains. Thus, the plasma should be considered as a strongly coupled plasma.

In the present paper, the formation of nonequilibrium charge carriers in the thermal plasma and their influence on the plasma-metal contact are considered.

## 2. Statement of the Problem

The ionization equilibrium in a thermal plasma is described by the Saha equation

$$\frac{n_e n_i}{n_a} = 2 \frac{g_i}{g_a} \left( \frac{m_e k T}{2\pi \hbar^2} \right)^{3/2} \exp\left(\frac{-I_a}{kT}\right) \equiv K_S, \quad (1)$$

where  $K_S$  is the Saha constant,  $g_i$  and  $g_a$  are the statistical weights of ions and atoms, respectively,  $I_a$  is the ionization potential,  $T$  is the Kelvin temperature,  $m_e$  is the electron mass,  $k$  is the Boltzmann constant, and  $\hbar$  is the Planck constant.

Thus, the requirements of mass conservation and charge conservation should be satisfied,

$$n_i + n_a = n_A, \quad n_e = n_i = n_0, \quad (2)$$

where  $n_A$  is the number density of the addition agent, and  $n_0$  is the unperturbed number density.

In a thermal plasma at atmospheric pressure, the ionization rate due to collisions in any microvolume of the plasma is much more than the diffusion velocity of the gas particles through this volume. Therefore, the number density of charge carriers can be described by the equilibrium distribution law [1]. In particular, in the area of the existence of the electrostatic potential  $\phi$ , the electron and ion number densities are described by the Boltzmann distribution law. If there is the charged interface in such a plasma (a grain or an electrode), then the Boltzmann law corresponds to the neutralization of the drift and diffusion streams of electrons and ions in the surface space charge layer.

However, the presence of the space charge causes a change of collisions between the gas particles, that entails a change of the intensities of ionization and recombination. Hence, there is a displacement of the ionization equilibrium in the superficial space charge region, and the Saha equation (1) can be applied, only if we introduce an additional term describing a change of the ionization equilibrium as a result of the exterior action on plasma [3, 4],

$$\frac{n_e n_i}{n_a} = K_S \exp\left(\frac{-e\varphi_{pl}}{kT}\right), \quad (3)$$

where the bulk plasma potential  $\varphi_{\text{pl}}$  [4–6] characterizes the size of the charge of the whole plasma volume and depends on the potential barrier on the interface. In particular, for a semiinfinite plasma with a potential barrier on the interface  $e\phi_s$ , the bulk plasma potential is defined by the expression [5]

$$\varphi_{\text{pl}} = -2 \frac{kT}{e} \tanh \left( \frac{e\phi_s}{4kT} \right). \quad (4)$$

The total potential  $\phi(r) + \varphi_{\text{pl}} = \varphi(r)$  represents the electrostatic potential with respect to the neutral plasma or the measured potential, since the potential of a probe depends on the bulk plasma potential to a big degree.

The space distributions of charge carriers are described by the expressions

$$n_e = n_q \exp(e\phi/kT), \quad n_i = n_q \exp(-e\phi/kT), \quad (5)$$

where  $n_q = n_0 \exp(-e\varphi_{\text{pl}}/2kT)$  is the quasinonperturbed density.

The problem of definition of the potential barrier on the interface in thermal plasmas was solved in [7] without regard for nonequilibrium charge carriers.

The following streams are considered:

(i) Richardson–Dushman stream of thermionic emission from the grain surface,

$$J_e^T = - \frac{4\pi e m_e k^2 T^2}{(2\pi\hbar)^3} \exp \left( - \frac{W}{kT} \right), \quad (6)$$

where  $J$  is the electric current density in the direction from a grain to plasma,  $W$  is the work function of an electron from a grain to plasma;

(ii) backflow of electrons absorbed by the surface of a grain,

$$J_e^{\text{abs}} = (1/4) e n_{es} v_{Te}, \quad (7)$$

where  $v_{Te} = \sqrt{8kT/\pi m_e}$  is the thermal velocity of electrons, and  $n_{es}$  is the electron number density at the surface of a grain, Eq. (5);

(iii) current density of the surface recombination of ions,

$$J_i^{\text{rec}} = \frac{-e n_{is} v_{Ti}}{4 [1 + g_i/g_a \exp(-E_s^{\text{ion}}/kT)]} \exp(-\varepsilon_a/kT), \quad (8)$$

where  $v_{Ti}$  is the thermal velocity of ions,  $n_{is}$  is the surface number density of ions, Eq. (5), and  $\varepsilon_a$  is the activation energy of the desorption of an atom;

(iv) current density of the surface ionization of atoms,

$$J_a^{\text{ion}} = \frac{e n_{as} v_{Ta}}{4 [1 + g_a/g_i \cdot \exp(E_s^{\text{ion}}/kT)]} \exp \frac{-\varepsilon_i}{kT}, \quad (9)$$

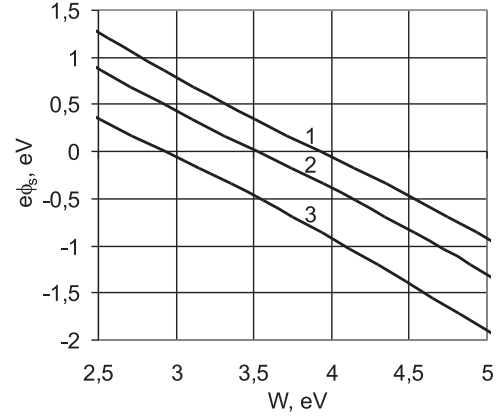


Fig. 1. Dependences of the potential barrier on the work function calculated without regard for nonequilibrium carriers: 1 –  $n_A = 10^{18} \text{m}^{-3}$ , 2 –  $10^{20}$ , 3 –  $10^{23}$

where  $v_{Ta}$  is the thermal velocity of atoms ( $v_{Ti} \approx v_{Ta}$ ),  $n_{as} = n_A - n_{is}$  is the surface number density of atoms, and  $\varepsilon_i$  is the activation energy of the desorption of an ion.

It was taken into account that the principle of detailed equilibrium should be fulfilled in the equilibrium state on the interface. This means the pair equality of streams (6), (7), and (8), (9). From Eqs. (6) and (7), we obtain

$$\frac{n_q}{\nu_e} = \exp \left( \frac{-W - e\phi_s}{kT} \right), \quad (10)$$

where  $\nu_e = 2 (m_e kT / 2\pi\hbar^2)^{3/2}$  is the efficient density of states of electrons, and  $e\phi_s$  is the potential barrier in the plasma.

The equality of the current densities of the recombination of ions, Eq. (8), and the ionization of atoms, Eq. (9), yields that

$$\frac{n_0^2}{\nu_e n_q} \exp \left( \frac{\varepsilon_a - \varepsilon_i}{kT} \right) = \exp \left( \frac{-W - e\phi_s}{kT} \right). \quad (11)$$

Equation (12) becomes identical to Eq. (11), if we assume that  $-e\varphi_{\text{pl}} = \varepsilon_a - \varepsilon_i$ . As a result, in view of the bulk plasma potential (4), the potential barrier is defined by the expression

$$e\phi_s + kT \tanh \left( \frac{e\phi_s}{4kT} \right) = kT \ln \left( \frac{\nu_e}{n_0} \right) - W. \quad (12)$$

In Fig. 1, the dependences of the potential barrier height on the work function of an electron from metal to plasma are presented for various number densities of the addition agent of potassium ( $I_a = 4.34 \text{eV}$ ) at the

temperature  $T = 2000$  K. The number density of the addition agent  $n_A = 10^{18} \text{m}^{-3}$  is the limiting value for the application of Eq. (11), because the approximation  $n_{as} \approx n_A$  is not valid at lower number densities.

### 3. Formation of Nonequilibrium Charge Carriers

A displacement of the ionization equilibrium means the occurrence of nonequilibrium charge carriers which are not described any more by the Boltzmann distribution law. Accordingly, there are the streams of nonequilibrium carriers which interact with the interface.

The conservation of charge carriers is described by the continuity equations [9–11]

$$\frac{\partial n_e}{\partial t} + \nabla \cdot \mathbf{j}_e = \omega_e, \quad \frac{\partial n_i}{\partial t} + \nabla \cdot \mathbf{j}_i = \omega_i, \quad (13)$$

where  $\mathbf{j}_{e(i)}$  is the stream density of electrons (ions), the source function  $\omega_{e(i)}$  is the intensity of changes of the electron (ion) number density as a result of the ionization-recombination processes.

In any volume of the plasma which does not contain the interface, the ionization degree is defined only by the collision ionization of addition agent atoms and the volumetric recombination:

$$\omega_e = \omega_i = \beta n_e n_a - \gamma n_e n_i = \beta n_e n_A - (\beta + \gamma) n_e n_i = \omega, \quad (14)$$

where  $\beta$  is the volumetric ionization coefficient, and  $\gamma$  is the volumetric recombination coefficient. Here, we have considered that  $n_i + n_a = n_A$ .

Outside of the space charge region, there is the equilibrium state under ionization and recombination:

$$\omega_0 = \beta n_q n_A - (\beta + \gamma) n_q^2 = 0, \quad (15)$$

whence follows  $\beta n_A = (\beta + \gamma) n_q$ .

The existence of the volumetric charge in the space charge layer brings to the disbalance between the intensity of recombination  $\gamma n_e n_i = \gamma n_q^2$  and the ionization rate  $\beta n_e n_a = \beta n_q \exp(e\phi/kT) n_A - \beta n_q^2$  which depends on the potential. Therefore,  $\omega \neq 0$ . Since Eq. (15) is valid outside the space charge layer, we get

$$\omega = \beta n_A n_q \left[ \exp\left(\frac{e\phi}{kT}\right) - 1 \right] \quad (16)$$

or  $\omega \approx \beta n_A n_q e\phi/kT$  for small potentials.

The nonzero  $\omega$  causes a change of the ionization degree of plasmas in the space charge layer and the formation of nonequilibrium charge carriers. The

positive potential causes a magnification of the ionization degree, and the negative potential leads to its diminution. This causes the disbalance between the diffusion and drift streams:

$$\begin{aligned} j_e &= b_e n_e^* \nabla \phi - D_e \nabla n_e^* \neq 0, \\ j_i &= -b_i n_i^* \nabla \phi - D_i \nabla n_i^* \neq 0, \end{aligned} \quad (17)$$

where  $b_{e(i)}$  is the electronic (ionic) mobility,  $D_{e(i)}$  is the electronic (ionic) diffusion coefficient, and “\*” means the nonequilibrium character of the number densities of charge carriers.

The nonequilibrium number densities can be presented in the form of a deviation from the equilibrium value  $n_e^* = n_e + \delta n$ ,  $n_i^* = n_i + \delta n$ , and the deviation should be identical for ions and electrons as the volumetric charge  $n_i^* - n_e^* = n_i - n_e = \rho/e$  should be constant.

Streams (17) should be equal because the current through the interface is absent, which is provided by the field of ambipolar diffusion

$$\frac{eE_a}{kT} = -\frac{e(D_e - D_i)}{kT(b_e + b_i)} \nabla \delta n.$$

Whence, taking into account the Einstein’s equation  $b_k T = eD$ , we obtain the current density of nonequilibrium carriers

$$j^* = -2 \frac{D_e D_i}{D_e + D_i} \nabla \delta n, \quad (18)$$

where  $2D_e D_i / (D_e + D_i) = D$  is the coefficient of ambipolar diffusion.

The ambipolar diffusion stream provides the movement of nonequilibrium charge carriers. At a positive field, the nonequilibrium electrons and ions move outwards the space charge layer. If the grain is negatively charged, the field is negative, i.e. it is directed to a grain. In this case, the ionization degree in the space charge layer is decreased, and the stream of nonequilibrium carriers is directed to a grain.

The change of the nonequilibrium carriers number density is determined by a continuity equation

$$\frac{\partial \delta n}{\partial t} + \text{div} j^* = \omega^*, \quad (19)$$

where  $\text{div} j^* = -D \Delta \delta n$ ,  $\omega^* = \beta n_A n_e^* (1 - n_i^*/n_q)$ .

In the stationary case, Eq. (19) is reduced to

$$\lambda_R^2 \Delta \left( \frac{\delta n}{n_q} \right) = \left[ \frac{\left( \frac{\delta n}{n_q} \right)^2}{1 + \left( \frac{E}{E_d} \right)^2} + \frac{\delta n}{n_q} - \frac{\exp \frac{e\phi}{kT} - 1}{1 + \left( \frac{E}{E_d} \right)^2} \right] \times$$

$$\times \left[ 1 + \left( \frac{E}{E_d} \right)^2 \right]. \quad (20)$$

Here, we took into account that  $2 \cosh(e\phi/kT) - 1 = 1 + (r_D e E/kT)^2$ , and the term  $kT/er_D = E_d$  can be considered as a “diffusion” field [12]. At small values of the potential  $e\phi \ll kT$ , we have  $\delta n/n_q \ll 1$ . Therefore, it is possible to neglect the quadratic term in Eq. (13). However, this can be made also at higher potentials, because  $\delta n/n_q \sim 1$  in this case, but  $E/E_d \gg 1$ .

In thermal plasmas at atmospheric pressure, the divergence of the electronic or ionic stream is much less than the rate of ionization and recombination, since the Maxwell time constant describing the diffusion-drift equilibrium stabilization is  $\tau_M = r_D^2/D \sim 10^{-6}$ s, and the lifetime of nonequilibrium carriers is  $\tau_R = (\beta n_A)^{-1} \sim 10^{-9}$ s. The nonequilibrium carriers’ recombination length [13]  $\lambda_R = \sqrt{D/\beta n_A} \sim 0.05 \mu$  is much less than the screening length  $r_D \sim 1 \mu$  which characterizes a change of the stream. Therefore, it is possible to consider the potential and the field as stationary quantities on the recombination length. Then the solution of Eq. (20) is the function

$$\delta n = n_q \frac{\exp \frac{e\phi}{kT} - 1}{1 + \left( \frac{E}{E_d} \right)^2} \left[ 1 + \frac{\lambda_R}{r_D} \exp \left( \frac{-r}{\lambda_R} \sqrt{1 + \left( \frac{E}{E_d} \right)^2} \right) \right]. \quad (21)$$

Accordingly, the density of the stream of nonequilibrium charge carriers is

$$j^* = \frac{D n_q}{r_D} \frac{\exp \frac{e\phi}{kT} - 1}{\sqrt{1 + \left( \frac{E}{E_d} \right)^2}} \exp \left( \frac{-r}{\lambda_R} \sqrt{1 + \left( \frac{E}{E_d} \right)^2} \right). \quad (22)$$

The density of the current of nonequilibrium charge carriers,  $J^* = e j^*$ , is essentially less than the currents given by Eqs. (6) – (9) and therefore cannot essentially influence the balance of the currents. However, a change of the surface electronic and ionic number densities varies the values of these currents.

#### 4. Change of the Surface Potential

Let us consider the balance of currents taking into account the nonequilibrium charge carriers. The balance of electronic currents in this case is represented as

$$n_q \exp \left( \frac{e\phi_s}{kT} \right) + \delta n_s = \nu_e \exp \left( \frac{-W}{kT} \right). \quad (23)$$

At the interface, the number density of nonequilibrium carriers, Eq. (21), becomes

$$\begin{aligned} \delta n_s &= n_q \frac{\exp \left( \frac{e\phi_s}{kT} \right) - 1}{2 \cosh \left( \frac{e\phi_s}{kT} \right) - 1} \left( 1 + \frac{\lambda_R}{r_D} \right) \approx \\ &\approx n_q \frac{\exp \left( \frac{e\phi_s}{kT} \right) - 1}{2 \cosh \left( \frac{e\phi_s}{kT} \right) - 1}. \end{aligned}$$

This allows us to transform Eq. (23) to

$$\exp \left( \frac{-2e\phi_s}{kT} \right) \left[ 2 \cosh \left( \frac{e\phi_s}{kT} \right) - 1 \right] = \frac{n_q}{\nu_e} \exp \left( \frac{W}{kT} \right). \quad (24)$$

The balance of the currents of the ionization of atoms and the recombination of ions gives the expression

$$\begin{aligned} n_q \exp \left( \frac{-e\phi_s}{kT} \right) + \delta n_s &= \\ &= n_{as} \frac{g_i}{g_a} \exp \left( \frac{-I_a + W + \psi_e + \psi_i + (\varepsilon_a - \varepsilon_i)}{kT} \right), \quad (25) \end{aligned}$$

where  $\psi_e$  and  $\psi_i$  correspond to the splitting of the plasma Fermi level to electronic and ionic quasilevels [13, 14] at the interface:  $\psi_e = F_e - F_{pl}$  and  $\psi_i = F_{pl} - F_i$ . For the nonequilibrium number densities, we get

$$n_{es}^* = n_{es} \exp \left( \frac{\psi_e}{kT} \right), \quad n_{is}^* = n_{is} \exp \left( \frac{\psi_i}{kT} \right),$$

where  $n_{es}$  and  $n_{is}$  are, respectively, the equilibrium surface electron and ion number densities.

This yields that

$$\psi_e = kT \ln \left( 1 + \frac{\delta n_s}{n_e} \right), \quad \psi_i = kT \ln \left( 1 + \frac{\delta n_s}{n_i} \right).$$

Accordingly, the nonequilibrium ion number density in Eq. (25) is

$$n_q \exp \left( \frac{-e\phi_s}{kT} \right) + \delta n_s = n_q \exp \left( \frac{-e\phi_s + \psi_i}{kT} \right).$$

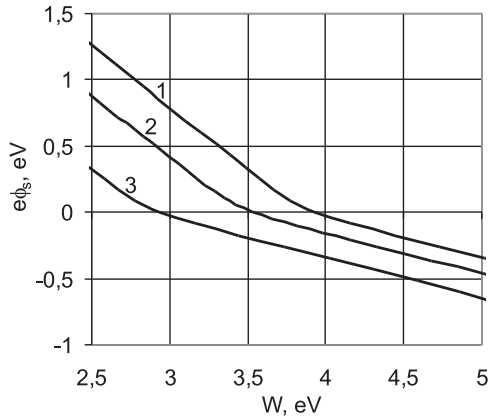


Fig. 2. Dependences of the surface potential barrier on the work function calculated with regard for nonequilibrium carriers: 1 –  $n_A = 10^{18} \text{m}^{-3}$ , 2 –  $10^{20}$ , 3 –  $10^{23}$

Then, in view of the relations  $\varepsilon_a - \varepsilon_i = -e\varphi_{pl}$  and  $n_{a0}K_S = n_q^2 \exp(e\varphi_{pl}/kT)$ , Eq. (25) yields that

$$n_q \exp\left(\frac{-e\phi_s}{kT}\right) = \frac{n_q^2 n_{as}}{\nu_e n_{a0}} \exp\left(\frac{W + \psi_e}{kT}\right) = \frac{n_q^2 n_{as}}{\nu_e n_{a0}} \exp\left(\frac{W}{kT}\right) \left[1 + \frac{\delta n_s}{n_q} \exp\left(\frac{-e\phi_s}{kT}\right)\right].$$

Whence we obtain

$$\exp\left(\frac{-2e\phi_s}{kT}\right) \left[2 \cosh\left(\frac{e\phi_s}{kT}\right) - 1\right] = \frac{n_q n_{as}}{\nu_e n_{a0}} \exp\left(\frac{W}{kT}\right). \quad (26)$$

Equation (26) corresponds to (24) if  $n_{as}/n_{a0} \sim 1$ , which is valid for the number densities of an addition agent  $n_A \geq 10^{18} \text{m}^{-3}$ . At smaller number densities, it is necessary to consider a change of the chemical potential of atoms that can be presented as follows [4]:

$$\psi_a = \mu_{a0} - \mu_{as} = kT \ln\left(\frac{n_{a0}}{n_{as}}\right) = kT \ln\left(\frac{n_A - n_q}{n_A - n_q \exp(-e\phi_s/kT) - \delta n_s}\right).$$

In Fig. 2, we present the dependences similar to those in Fig. 1, but with regard for nonequilibrium charge carriers.

Equation (24) corresponds to (10) for the positive potential barriers, when  $e\phi_s > kT$ , because, in this case,

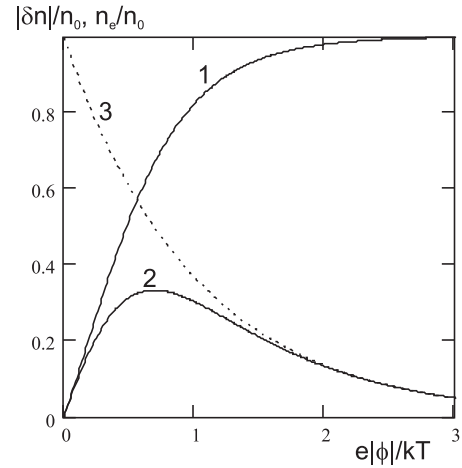


Fig. 3. Number density of nonequilibrium charge carriers versus the potential: 1 – at a pinch of ionization ( $\phi > 0$ ,  $\delta n > 0$ ); 2 – at a decrease in ionization ( $\phi < 0$ ,  $\delta n < 0$ ); 3 – change of the electron number density at  $\phi < 0$

$\delta n \sim n_q$  and  $n_{es} \gg n_q$ . That is, the presence of nonequilibrium charge carriers affects the potential barrier height which is defined by preferentially electronic streams a little. However, at negative potentials, the distinction is large. The difference in the dependences of the positive and negative surface potentials on the work function is defined by that the ionization degree change in the space charge layer occurs differently at the positive and negative surfaces.

In Fig. 3, the modulus of the number density of nonequilibrium carriers versus the potential is presented for  $r = 0$ . It is seen that  $\delta n$  tends to  $n_q$  at the positive potential, which compensates a diminution of the ion number density in the space charge layer. The electron number density in the space charge layer is decreased ( $\delta n < 0$ ) at the negative potential, and the ionization degree is decreased as well. However, a decrease in ionization (magnification of  $-\delta n$ ) cannot be more than the electron number density, i.e.  $-\delta n \leq n_e = n_q \exp(-e|\phi|/kT)$ . Therefore, the large negative potential causes a diminution of the number of nonequilibrium charge carriers.

As a result, relation (24) can be approximated for small potential barriers  $e\phi_s \ll kT$  by the expression

$$e\phi_s = \frac{4}{9} \left[ kT \ln\left(\frac{\nu_e}{n_0}\right) - W \right], \quad |e\phi_s| \ll kT. \quad (27)$$

In the case  $W_{pl} < W$ , we get

$$e\phi_s = \frac{1}{3} \left[ kT \ln\left(\frac{\nu_e}{n_q}\right) - W \right], \quad \phi_s \ll 0; \quad (28)$$

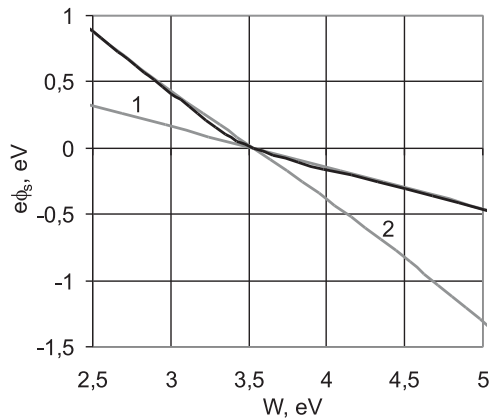


Fig. 4. Surface potential barrier versus the work function calculated with regard for nonequilibrium carriers: 1 — approximation by Eq. (28), 2 — approximation by Eq. (29)

For  $W_{pl} > W$ , relation (24) can be approximated by the expression

$$e\phi_s = kT \ln \left( \frac{\nu_e}{n_q} \right) - W, \quad \phi_s \gg 0. \quad (29)$$

Both last dependences are presented in Fig. 4 for the plasma containing potassium:  $n_A = 10^{20} \text{m}^{-3}$  at temperature  $T = 2000 \text{K}$ .

## 5. Conclusion

It has been demonstrated that the existence of nonequilibrium charge carriers essentially affects the potential of the interface if the work function of an electron from the metal exceeds the work function of an electron from the plasma: the account of nonequilibrium charge carriers gives the decrease of the potential by three times at a negative charge of dust grains. This occurs because the initial ionization degree in the low-temperature plasma is small. Therefore, there can be a large increase of the ionization degree up to the full ionization of the gas at a positive charge of grains. But the ionization degree decrease is restricted by the ionization-free state of the gas at a negative charge of grains.

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## НЕРІВНОВАЖНІ НОСІЇ ЗАРЯДУ У КОНТАКТІ ТЕРМІЧНА ПЛАЗМА — МЕТАЛ

В.І. Вишняков

Резюме

Досліджено шар просторового заряду на межі поділу термічна плазма — метал. Показано, що наявність об'ємного заряду приводить до утворення нерівноважних носіїв заряду. Наявність останнього мало відбивається на величині потенціалу порошинки, якщо ця порошинка заряджена позитивно. Але для негативно заряджених частинок врахування нерівноважних носіїв заряду дає триразове зменшення потенціального бар'єра на межі поділу фаз.