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Influence of Vacancies and Pores that Appear during Irradiation in the Surface Metal Layer on Field Emission Current

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Quantum-mechanical problem on electron motion through the model potential barrier of metal-vacuum system with an additional near-surface dipole layer is considered. An analytical generalization of the Fowler-Nordheim formula was done in the case of this potential barrier. The introduction of such a layer is used as a way of taking into account the effect of vacancies and pores that appear during irradiation in the surface metal layer on the field emission current density. Taking into consideration the continuity conditions of wave functions and their derivatives at the interface between two media, general expressions of the transmission coefficient of potential barrier and the field emission current density are obtained. The influence of effective thickness of an additional near-surface dipole layer on value of field emission current is shown. Thus, the method is used to take into account the influence of vacancies and pores that appear during irradiation of metal surface on field emission current density. It was found that field emission current, considering the influence of vacancies and pores, that emerge during irradiation of copper surface with Cu^{2+} ions with energy of 300 keV during the processing time of $5 \cdot 10^3$ s by dose of $1.6 \cdot 10^{21}$ m^{-2}, is 1.3 times less than the value of the Fowler-Nordheim current density. The approximations related to the Fowler-Nordheim formula for current density are given in this paper. The considered approach has more visual mathematical proofs than previous theoretical treatments of the phenomenon of field emission. This research can be a useful basis for studying the transmission of electrons through potential barriers for which there is no an exact analytical theory.

Keywords: Field emission, Potential barrier, Transmission coefficient of the potential barrier, Thickness of the metal surface modification, High-gradient breakdown.

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1. INTRODUCTION

Despite the fact that the theory of field emission under the influence of external electromagnetic fields, written by Fowler and Nordheim in the 20 years of the last century, this topic is relevant [1], in particular, in the task of overcoming high-gradient breakdowns in accelerating structures for receiving charged particles of high energies on modern accelerators. Field emission is one of the stages of occurrence of high-gradient breakdown in accelerating structures.

Experiments on the modeling accelerator structures of the compact linear electron-positron accelerator in the CLIC (Compact Linear Collider) project, carried out at the European Center for Nuclear Research, CERN, showed that when introducing power of a high-frequency electromagnetic field, which provides the electric field strength on the accelerating axis of a value of the order of 100 MV / m, high-vacuum breakdowns arise on the surface of the accelerating structure [2-5]. Since each breakdown leads to a loss of the density of the charged particle beam and to damage of the surface of material, various methods of increasing the resistance to high-gradient breakdown of the accelerating structure are being investigated. One way to eliminate the problem of high-vacuum high-gradient breakdowns is to reduce parasitic currents of field emission. Now CERN studies conditioning of the surface of electrodes to increase the stability of metal surface of accelerating structures to high-gradient breakdowns [2]. Other possible ways to overcome high-gradient breakdowns are implantation of some elements (for example, argon, nitrogen, zirconium ions) into the near-surface layer of the metal [6, 7]; vacuum coating of a more refractory metal film on the surface [7]; desorption of molecules of gases from the metal surface; location of accelerating structure in an external magnetic field.

The effect of electrode surface modification on the field emission current density was experimentally investigated in [7, 8]. Experiments on irradiation of copper surface with ions Ar^{2+}, Zr^{2+}, Cu^{2+} with energies of 300 keV and N^{+} ions with energy of 150 keV and irradiation dose in the range $(1.2, ..., 4) \cdot 10^{21}$ m^{-2} were carried out in [7] to study the influence of ion beam and plasma modification of metal surface of copper samples on the conditions for start and development of a high-voltage breakdown. It was shown experimentally that plasma and ion-beam modification of copper surface leads to an increase in the breakdown voltage from 5 to 35 %, depending on modification method, and reduces the dark current of the anode-cathode system.

In the papers [9, 10], the influence of magnetic field on the field emission current is theoretically investigated. In the paper [9], it was shown a change of the potential barrier under the influence of a magnetic field parallel to the metal surface and provides the ability to reduce the probability of breakdown and increase in the stability of accelerating structures to high-gradient breakdowns by introducing them into a magnetic field system.

In the paper [11], the potential barrier model for two W-Cs systems was considered with a thin layer and a hyperfine layer of cesium. The effect of equalizing Fermi levels of two-metal system in the dipole layer and the

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value of work functions of each component of metal-metal system are taken into account. A similar form of a potential barrier was considered in papers [12, 13], in which the authors described the field emission of electrons from nanoscale objects for the barrier depicted in Fig. 1. In the paper [12], the author has showed the effect of the Coulomb blockade phenomenon on magnitude of the field emission current.

The purpose of this paper is to generalize the Fowler-Nordheim theory of field emission from a metal with an additional near-surface dipole layer. This theory is used to take into account the effect of vacancies and pores on the field emission current density that appears during irradiation of metal surface.

2. BASE MODEL

Fowler and Nordheim obtained the formula for the field emission current density on the basis of finding the transmission probability of the potential barrier *D(W)* as a function of the electron energy *W* at the boundary from the metal to the vacuum. The continuity conditions of the wave functions and their derivatives are used at the interface between the two media. The transmission coefficient is defined as the ratio of the flux density of the wave passing through the barrier to the density of the falling wave flux:

$$D(W) = j_1 / j_0 \,, \tag{1}$$

where j_1, j_0 are the density fluxes of the wave passing through the barrier and the falling wave, respectively.

The field current density *j* is given by:

$$j = e\int_0^\infty D(W)N(W)dW \,, \tag{2}$$

where *N(W)* is the number of electrons incident on a surface of unit area per unit time with a kinetic energy *W* normal to the surface; *e* is the electron charge. The Fowler-Nordheim expression for the current density is written as:

$$\begin{gathered} j_{F-N} = AF^2 \exp\left(-\left(4k/(3F)\right)\chi^{\frac{3}{2}}\right), \\ A = e/(4\pi^2\hbar))\sqrt{\mu}/\left((\mu+\chi)\sqrt{\chi}\right), \end{gathered} \tag{3}$$

where $F = eE$; E is the electric field strength; χ is the work function; μ is the thermodynamic partial potential of an electron; $k = \sqrt{2m}/\hbar$; m is the electron mass.

Let's consider the phenomenological model of the potential barrier of the metal-vacuum system, which is depicted in Fig. 1. In this figure, the region I is the inner region of the metal, the region II is the effective thickness *d* of the near-surface dipole layer, the regions II and III are regions of metal modification, and the IV region is a vacuum. Height *C* of the second and fourth areas, depicted in Fig. 1, is equal to the sum of the work function χ of a metal and the thermodynamic partial potential of an electron μ, $C = \mu + \chi$, *R* is the thickness of metal surface modification, *h* is the distance to the dipole layer, *d* is the effective thickness of an additional near-surface dipole layer, $p = h + d$.

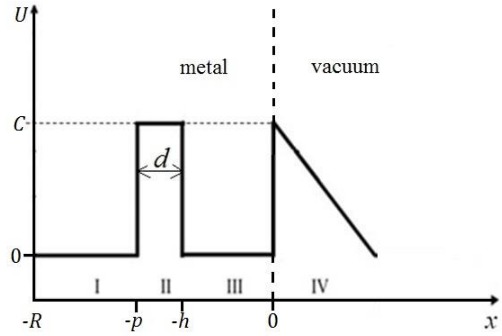

Fig. 1 – Diagram of the potential barrier for escape of electrons from the modified metal surface to vacuum

The wave functions and their partial derivatives must be single-valued and continuous [14]. As a result, authors have obtained a system of equations for finding complex amplitudes:

$$\begin{aligned} & A_1 e^{-ik_1 p} + B_1 e^{ik_1 p} = A_2 e^{\beta p} + B_2 e^{-\beta p}, \\ & ik_1\left(A_1 e^{-ik_1 p} - B_1 e^{ik_1 p}\right) = \beta\left(-A_2 e^{\beta p} + B_2 e^{-\beta p}\right), \\ & A_2 e^{\beta h} + B_2 e^{-\beta h} = A_3 e^{-ik_1 h} + B_3 e^{ik_1 h}, \\ & \beta\left(-A_2 e^{\beta h} + B_2 e^{-\beta h}\right) = ik_1\left(A_3 e^{-ik_1 h} - B_3 e^{ik_1 h}\right), \\ & A_3 + B_3 = \sqrt{\varphi/F}\, e^{-\pi i/2}(k^2F)^{1/6} H_{1/3}^{(2)}(y), \\ & ik_1\left(A_3 - B_3\right) = \left(L\cdot H_{1/3}^{(2)}(y) - M\cdot\left(dH_{1/3}^{(2)}(y)\big/dQ\right)\right), \\ & L = \left((\varphi/F)^{-1/2}/2\right)(k^2F)^{1/6} e^{\pi i/2}, \\ & M = (\varphi/F)k\sqrt{F}(k^2F)^{1/6} e^{3\pi i/2}, \end{aligned} \tag{4}$$

where A_1, B_1, A_2, B_2, A_3, B_3 are the complex amplitudes of the wave functions; $k_1 = \sqrt{2mW}/\hbar$; $\beta = \sqrt{2m(C-W)}/\hbar$; $\varphi = C - W$; $y = e^{-3\pi i/2}Q$; $Q = 2k\sqrt{F}(\varphi/F)^{\frac{3}{2}}/3$.

The transmission coefficient of the potential barrier *D*(*W*) in accordance with (1) and taking into account the absence of sources of particles can be found by the formula:

$$D(W) = 1 - |A_1|^2 / |B_1|^2 \,. \tag{5}$$

In the general case, the coefficient *D*(*W*) depends on parameters *W, F,* μ, χ, *d, h* and its analysis is done numerically. If $d \to 0$, the expression *D*(*W*) takes the form found by Fowler and Nordheim:

$$D_{F-N} = 4\left(\sqrt{(W\varphi)}/C\right)\exp\left(-4k\varphi^{\frac{3}{2}}/3F\right). \tag{6}$$

The transmission coefficient *D(W)* under conditions

$$k_1 d << 1 \,,\ \beta d << 1 \,,\ k_1 h << 1 \,,\ \beta h << 1, \tag{7}$$

can be simplified to the following analytical form:

$$D(W) = D_{F-N}(1-d/d_0), \quad (8)$$

$$d_0 = (F/\varphi + 2\sqrt{2m\varphi}/\hbar)^{-1}. \quad (9)$$

In this approximation, the expression for $D(W)$ does not depend on the distance h to the dipole layer. Notice, under conditions (7), it is correct at $d << d_0(W)$ for all W. At the value $F = 5 \cdot 10^9$ eV/m for the copper characteristics:

$$C = 12 \text{ eV}, \ \mu = 7.5 \text{ eV}, \ \chi = 4.5 \text{ eV}, \ W = \mu, \quad (10)$$

it is found that $d_0(\mu) = 0.5 \cdot 10^{-10}$ m^{-1}.

Fig. 2 shows dependences of D on d at aforementioned parameters. The solid line corresponds to the equations (6), and the line depicted by dots corresponds to (8). The curve that is shown by dashed-dotted line in Fig. 2 describes the numerical calculations of D at $h = 10^{-11}$ m. From Fig. 2 it is seen that the graph of the analytical formula (8) under the condition $d << d_0(W)$ lies below the line of dependence of D on d calculated by the numerical method. The tunneling probability D on d of the potential barrier, calculated numerically, is 1.3 times smaller in relation to the value D_{F-N} at the value of the electric field strength $E = 5 \cdot 10^9$ V/m, $d = 10^{-11}$ m.

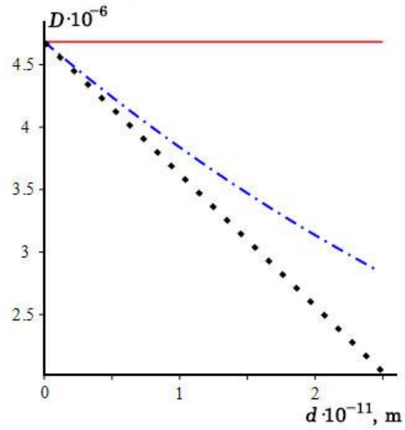

Fig. 2 – Dependences on d of the transmission coefficient of the potential barrier

The current density, which is found by the expression (2), taking into account (5), at

$$N(W) = (m/(2\pi^2\hbar^3))(\mu - W), \quad (11)$$

in general case depends on parameters F, μ, χ, d, h. Under condition $d << d_0(\mu)$, the expression for the field emission current density is obtained as

$$j = j_{F-N}(1 - d/d_0(\mu)). \quad (12)$$

The dependence of the current density (12) on the electric field strength at parameters (10) and the value $d = 10^{-11}$ m is shown in Fig. 3 (dashed line) in the logarithmic coordinates of Fowler-Nordheim. The solid line describes the Fowler-Nordheim current density j_{F-N}. As can be seen from Fig. 3, similarly to the transmission coefficient $D(W)$ the current density j of the field emission decreases by 1.3 times relative to the value j_{F-N} at the value of the electric field strength $E = 10^{10}$ V/m.

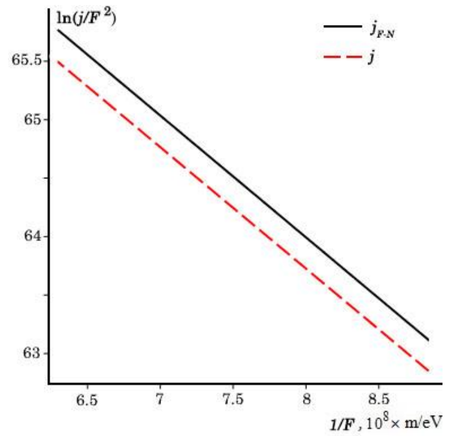

Fig. 3 – The dependences of the field emission current density from the metal surface

Fig. 4 shows the graphs of the dependence of $j_{F\text{-}N}$ on the work function χ for various electric field strengths E, where $j_0 = 10^7$ A/m^2, $E_0 = 10^9$ V/m. From graphs it can be concluded that a change of 10 % in the value of the work function of the metal changes the magnitude of the current density j_{F-N} by 7 times.

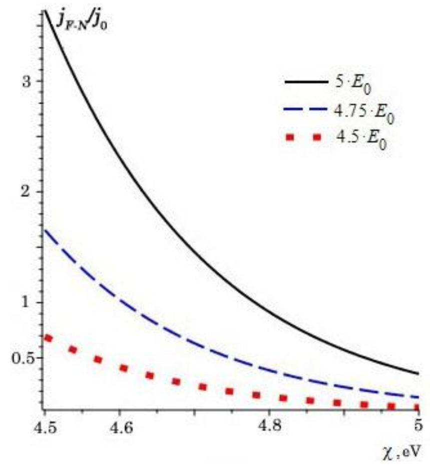

Fig. 4 – The dependence of the current density on the metal work function at different electric field strengths

3. RESULTS AND DISCUSSION

Introduction of region II shown in Fig. 1 is considered as a way of taking into account the influence on the field emission current density of vacancies and pores that appear during irradiation of the metal surface. Assume that the effective thickness d of the dipole layer is defined from the sum of the volumes of all generated point defects and emptiness of the modified layer. For example, one of the methods for the formation of defects is the bombardment of the surface of the copper structure by high-energy beams of copper ions. The value of the effective thickness d of the dipole layer can be estimated using the formula:

$$d = nRV_0, \quad (13)$$

where n is the concentration of generated defects, V_0 is an average volume of a point defect. The thickness of the surface modification R is assumed to be equal to projected range of charged particle in the metal.

One can define the distance to the dipole layer h and the thickness d in the following way. The irradiation dose with Cu^{2+} ions with an energy of 300 keV during the whole processing time $t = 5 \cdot 10^3$ s of the copper surface in [7] is equal to $1.6 \cdot 10^{21}$ m^{-2}. The effective range R of Cu ions with an energy of 300 keV in copper is $8.62 \cdot 10^{-8}$ m according to results of calculations of the SRIM program [15] or using the Bethe-Bloch formula.

Fig. 5 shows the dependence of the sum of the electronic and nuclear components of the stopping power of an incident ion Cu^{2+} in copper based on the results of the SRIM calculations. Inequalities in the maximum of the graph can be explained by the agreement of the dependency program for small and high velocities in the range of intermediate energy of ions when passing through the sample material.

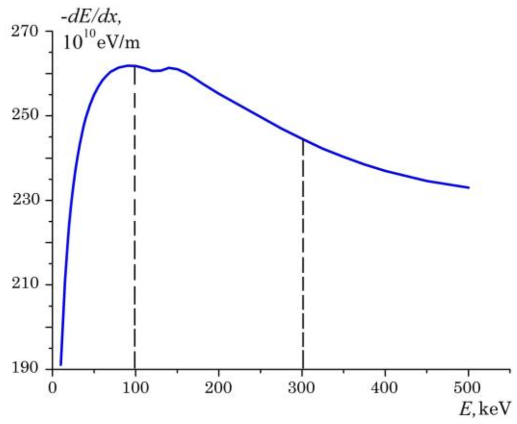

Fig. 5 – Graph of the dependence of the full stopping power of Cu^{2+} ions on energy in copper

Fig. 5 shows that the largest energy release of the Cu^{2+} ion is at energies near the value of 100 keV. With the particle energy in the range from 300 keV to 100 keV, the frictional forces acting upon the ion passing through the substance are inversely proportional to the energy, and in the interval from 100 keV to 0 they are directly proportional. Thus, the particle at the beginning of the motion will lose its energy by interacting with the target electrons, and at the end of its run in the substance it will be stopped as a result of nuclear collisions with atoms.

Generation of vacancies reduces the density of the sample by increasing the volume at an almost constant mass [16]. Fig. 6 represents the distribution of generated vacancies that depends on the depth of ion transmission in copper during irradiation of the copper sample surface by Cu^{2+} ions with an initial energy of 300 keV, which is based on the results of the SRIM calculations. The area under the line of dependence is proportional to the total number of vacancies that appear during irradiation of the metal surface by ions. The condition of equality of areas $S_1 = S_2$ is used to find the parameter h. By the results of calculations, $h = 5.8 \cdot 10^{-8}$ m. The equality $S_1 = S_2$ means that the number of vacancies in a layer depth from 0 to h is equal to the number of vacancies in the range from h to R. That is, vacancies are produced unevenly in the material during the irradiation process. The total number of vacancies N_v without taking into account the process of recombination with interstitial atoms is equal to $4.9 \cdot 10^{20}$ at a radiation dose of $1.6 \cdot 10^{21}$ m^{-2}. One can assume that the vacancy radius is $r = 0.5 \cdot 10^{-10}$ m, then the volume V_0 of one vacancy is $5.24 \cdot 10^{-31}$ m^3. The volume of the modified layer can be defined as $V = S \cdot R = 8.62 \cdot 10^{-12}$ m^3, where S is the surface area of the copper sample, which is equal to 10^{-4} m^2 [7]. Thus, the volume of generated vacancies is equal to $N_v \cdot V_0 = 2.6 \cdot 10^{-10}$ m^3, and it is represented phenomeno-logically as a dipole layer located at a distance of $h = 5.8 \cdot 10^{-8}$ m from the modified metal surface with a cross-sectional area S and a thickness d. The effective thickness d of the dipole layer is estimated, neglecting the presence of the recombination process of vacancies and interstitial atoms, as $d = N_v \cdot V_0 / S = 2.6 \cdot 10^{-6}$ m.

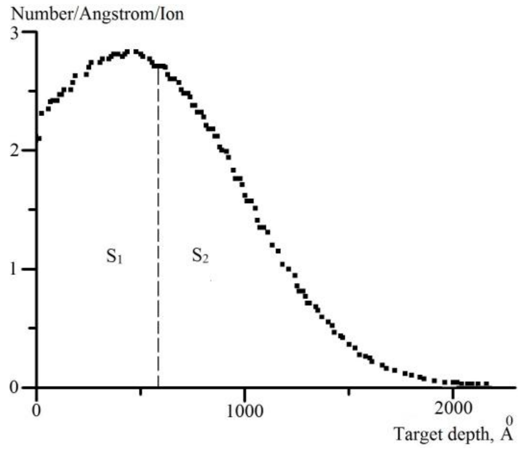

Fig. 6 – Distribution of vacancies that appeared by irradiation with Cu^{2+} ions of a copper surface with an initial energy of 300 keV based on the results of calculations of the program SRIM

Taking into account the influence of defect recombination process [17-19] will reduce the effective thickness d of the dipole layer by several orders, leading to the values of the parameters d and h, which were defined earlier.

4. CONCLUSIONS

The model of the potential barrier of metal with an additional near-surface dipole layer is proposed. The introduction of such a layer is used as a way of taking into account the effect of vacancies and pores that appear during irradiation in the surface metal layer on the field emission current density. An analytical generalization of the Fowler-Nordheim formula was done in the case of the potential barrier shown in Fig. 1. When $d << d_0(\mu)$, the analytical expressions (8) and (12) are obtained using the Fowler-Nordheim method for the transmission coefficient of the potential barrier and the field emission current density, taking into account the additional near-surface dipole metal layer, respectively. It is shown that at $d = 10^{-11}$ m, the field emission current (12) decreases by 1.3 times relative to the value j_{F-N} for the considered barrier at the value of the electric field strength $E = 10^{10}$ V/m.

From the analysis of the dependence of j_{F-N} (3) on the work function of the metal at various electric field strengths E, it follows that a change of 10 % in the value of the metal work function changes the magnitude of the current density j_{F-N} by 7 times. Formula (3) does not take into account the effects of impurities, cracks, vacancies, pores, inclusions, surface irregularities of the metal surface on the value of j_{F-N}. The experiments on the modification of the near-surface metal layer show a weak dependence of the field emission current density on the metal work function [5, 7].

Using the SRIM code, the distribution of vacancies is shown as a function of transmission depth of Cu^{2+} ions into the volume of the copper sample to find the effective thickness d of an additional near-surface dipole layer and the distance h to this dipole layer.

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Вплив вакансій і пор, що виникають при опроміненні в поверхневому шарі металу, на струм польової емісії

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Розв’язана квантово-механічна задача про рух електрона через модельний потенціальний бар’єр системи метал-вакуум з введенням додаткового приповерхневого дипольного шару. Проведено аналітичне узагальнення формули Фаулера-Нордгейма для розглянутого потенціального бар'єру. Введення додаткового дипольного шару використовується як спосіб врахування впливу вакансій і пор, що виникають при опроміненні у поверхневому шарі металу на густину струму польової емісії. Враховуючи умови неперервності хвильових функцій і їх похідних на межі розділу двох середовищ, одержано загальні вирази для коефіцієнта прозорості потенціального бар’єру та густини струму польової емісії. Показано вплив ефективної товщини додаткового приповерхневого дипольного шару на величину польового емісійного струму. Таким чином, цей метод застосовується для врахування впливу на густину струму польової емісії вакансій і пор, утворених опроміненням поверхні металу. З’ясовано, що густина струму польової емісії, з врахуванням впливу утворених вакансій і пор опроміненням поверхні металу іонами Cu^{2+} з енергією 300 кеВ протягом часу обробки $5 \cdot 10^3$ с поверхні міді дозою $1.6 \cdot 10^{21}$ м$^{-2}$, менша у 1.3 рази відносно значення густини струму Фаулера-Нордгейма. Наведено наближення, пов'язані з одержанням формули Фаулера-Нордгейма для густини струму. Розглянутий підхід має більш наочні математичні вирази, ніж попередні теоретичні дослідження явища польової емісії. Ця робота може бути корисною основою для дослідження проходження електронів через потенціальні бар'єри, для яких ще не існує точної аналітичної теорії.

Ключові слова: Польова емісія, Потенціальний бар'єр, Коефіцієнт прозорості потенціального бар'єру, Товщина модифікації поверхні металу, Високоградієнтний пробій.