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SURFACE INHOMOGENEITIES IMPACT ON BALLISTIC CHARGE TRANSPORT IN NANOSIZED METAL FILMS

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A model approach is proposed that allows to evaluate the influence of parameters of average amplitude h and correlation length ξ of surface inhomogeneities on the dimensional concentration of electronic states $n(d)$ and specific conductivity $\sigma(d)$ of nanoscale metal films at ballistic charge transport regime. According to Fishman- Calecki, the impact of surface inhomogeneities on the regime of ballistic charge transport in nanoscale films of silver, gold and copper was analyzed. The minimum thickness d_c of the metal film for which the application of proposed approach is valid was carried out within percolation theory.

Keywords: thin metal films, size effect, surface inhomogeneities, ballistic transport.

Introduction.

The study of electron transport phenomena in limited dimensions metal samples, allows you to get information about fundamental properties of matter in a state when the impact of the surface scattering becomes dominant in compare with the volume scattering of conduction electrons. The experimental results of research allow forming nano-sized metal samples with the desired kinetic properties. The problem of quantitative description of dimensional dependences of kinetic coefficients of nanometer-thick metal films is relevant in the production of metal layers with predetermined structure and electrical properties. The quality of agreement of experimental data with the results of theoretical calculations is achieved only with successful choice of model approach when describing kinetic phenomena in a nanosized metal sample. The model should sufficiently take into account possible features of the sample structure and its electronic structure. In most modern approaches of charge transport phenomena description in metal layers, it is carried out for Sondheimer metal. At the same time, it is considered to be homogeneous and isotropic with atomically smooth outer surfaces (model of a plane-parallel layer) or a layer on the surface of which there are inhomogeneities of macroscopic dimensions, the parameters of which can be described with the help of harmonic or power functions. According to modern theoretical approaches [1-13], it is stated that in an electrically continuous film, the structure of which (average linear dimensions of the crystallites) does not change with the increase in the thickness of the metal film, the dependence of the dimensional specific conductivity $\sigma(d)$ is described by a non-linear function. In the interval of thicknesses $(d >> \lambda)$, where λ is electron mean free pass, the

dimensional dependence of the specific conductivity is linear $\sigma(d) \sim d$ and is well described by the quasi-classical approaches of Fuchs-Sondheimer, Mayadas- Schatkes, Tellier-Tose-Pichar, etc. [1,2], who assume film structure as plane-parallel layer on the surface of which there are point defects. In films of smaller thicknesses, the linearity is violated and to describe $\sigma(d)$ it is necessary to take into account the existence of macroscopic inhomogeneities on the surface. The dependence of $\sigma(d)$ is described by the approaches of Namba, Wissman and polycrystalline layer of nonuniform thickness [2]. Further decrease in d of the metal film, realizes the ballistic mode of charge transport, with the majority of current carriers being dispersed on surface inhomogeneities. The dimensional dependence $\sigma(d)$ can be described according to the approach [3-9] by a power dependence, where $\sigma_{res}(d) \sim d^{\alpha}$, where $2{,}1 \le \alpha \le 6$, here $\sigma_{res}(d) = 1/[\rho(d) - \rho_{\infty}]$ is the residual conductivity and ρ_{∞} is film resistivity of infinite thickness $d >> \lambda$.

According to our experimental data, it was shown that freshly deposited [2,10], thermo-stabilized by low-temperature annealing mode, metals films [2], the above picture of $\sigma = \sigma(d)$ description is observed in all cases. In particular, our experimental works show that in the regime of ballistic charge transport in gold, silver and copper films, the value of α can vary in the interval depending on the average of surface inhomogeneities h amplitude (Namba approach [1]): if $h < 2\ nm$ than $1{,}9 < \alpha < 2{,}1$, and if $h = (8-10)\ nm$ than $2{,}8 < \alpha < 3{,}1$ [10].

The purpose of work, to show the influence of macroscopic inhomogeneities of the surface - an isotropic, electrically continuous film of Sommerfield metal on the nature of the dimensional dependence $\sigma = \sigma(d)$ under conditions of ballistic charge transport [5, 11].

Thin metal film model

Let the metal film be considered homogeneous, isotropic and electrically continuous metal plate, which for conduction electrons is a potential well of infinite depth and width d. One of the surfaces of the film has periodic inhomogeneities, which can be described by a harmonic function: $y(x) = h\cos(2\pi x/\xi)$, where h and ξ the amplitude and period of recurrence of the surface inhomogeneities. Let us find the wave functions and eigenvalues of the electron energy for the three-dimensional Schrödinger equation with the appropriate boundary conditions. Note that it is possible to obtain a linear correction to the wave vector of a particle and calculate the change in its energy [11] in the limiting case $k_x = k_y = 0$. The characteristic equation for this case will take the form [11]:

$$tg(d) = \frac{h^2}{2} k_1 ctg(dk_1)\left[k_0 + k_2 tg(dk_0) ctg(dk_2)\right], \quad k_1 = \sqrt{k_0^2 - \left(\frac{2\pi}{d}\right)^2}, \quad k_2 = \sqrt{k_0^2 - 4\left(\frac{2\pi}{d}\right)^2}. \qquad (1)$$

Let us consider two dimensional areas where $k_{0s} \le 2\pi/\xi$ and $k_{0s} \ge 2\pi/\xi$, $k_{0s} = \pi s/d_0, s = 1, 2, 3, 4 \ldots N_c$, where s takes only integer values. From where we get the dimensional condition for the maximum number of states in potential well of width d_0: $N_c = Int(k_F d_0/\pi)$, k_{0s} is the wave vector of particle in a smooth quantum well $(h_0 = 0)$. Let's write down the corrections to the wave vector and particle energy in the corresponding dimensional region:

$$k_{0s} \le 2\pi/\xi,$$

$$\delta k_{0s} = \frac{h^2}{2d} k_{0s} k_{1s} cth(dk_{1s}),\ k_{1s} = \sqrt{\left(\frac{2\pi}{a}\right)^2 - k_{0s}^2}$$

$$E_s = \frac{\hbar^2\pi^2}{2m}\left(\frac{s}{d}\right)^2\left[1+\frac{h^2}{d^2}\sqrt{\left(\frac{2\pi d}{\xi}\right)^2-(s\pi)^2}\,cth\left(\sqrt{\left(\frac{2\pi d}{\xi}\right)^2-(s\pi)^2}\right)\right]$$

$$k_{0s} > 2\pi/\xi, \qquad (2)$$

$$\delta k_{0s} = \frac{h^2}{2d} k_{0s} k_{1s} ctg(dk_{1s}),\ k_{1s} = \sqrt{k_{0s}^2 - \left(\frac{2\pi}{\xi}\right)^2}$$

$$E_s = \frac{\hbar^2\pi^2}{2m}\left(\frac{s}{d}\right)^2\left[1+\frac{h^2}{d^2}\sqrt{(s\pi)^2-\left(\frac{2\pi d}{\xi}\right)^2}\,ctg(\sqrt{(s\pi)^2-\left(\frac{2\pi d}{\xi}\right)^2})\right]$$

The total energy of electron in a metal film, provided that the value of the Fermi energy of the particle is constant, can be written:

$$k_{0s} \le 2\pi/\xi$$

$$E = \frac{\hbar^2}{2m}\left(k_x^2+k_y^2\right)+\frac{\hbar^2\pi^2}{2m}\left(\frac{s}{d}\right)^2\left[1+\frac{h^2}{d^2}\sqrt{\left(\frac{2\pi d}{a}\right)^2-(s\pi)^2}\,cth\left(\sqrt{\left(\frac{2\pi d}{a}\right)^2-(s\pi)^2}\right)\right]$$

$$k_{0s} > 2\pi/\xi \qquad (3)$$

$$E = \frac{\hbar^2}{2m}\left(k_x^2+k_y^2\right)+\frac{\hbar^2\pi^2}{2m}\left(\frac{s}{d}\right)^2\left[1+\frac{h^2}{d^2}\sqrt{(s\pi)^2-\left(\frac{2\pi d}{\xi}\right)^2}\,ctg(\sqrt{(s\pi)^2-\left(\frac{2\pi d}{\xi}\right)^2})\right]$$

From (3), the electron quasi-momentum p will take the form:

$$k_{0s} \le 2\pi/\xi$$

$$p^2 = \hbar^2 k_F^2 + \left(\frac{\hbar\pi}{d}\right)^2 s^2\left[1+\frac{h^2}{d^2}b_1\right]\ b_1 = \sqrt{\left(\frac{2\pi d}{\xi}\right)^2-(s\pi)^2}\,cth\left(\sqrt{\left(\frac{2\pi d}{\xi}\right)^2-(s\pi)^2}\right)$$

$$k_{0s} > 2\pi/\xi \qquad 4)$$

$$p^2 = \hbar^2 k_F^2 + \left(\frac{\hbar\pi}{d}\right)^2 s^2\left[1+\frac{h^2}{d^2}b_2\right]\ b_2 = \sqrt{(s\pi)^2-\left(\frac{2\pi d}{\xi}\right)^2}\,ctg(\sqrt{(s\pi)^2-\left(\frac{2\pi d}{\xi}\right)^2})$$

Let's calculate the density of surface states in the phase space, knowing that for a 2D electron gas $dn_{2D} = pdp/(2\pi\hbar^2)$, from where, after differentiating (4), we get the following expressions:

$$k_{0s} \le 2\pi/\xi$$

$$2pdp = -2\left(\frac{\hbar\pi}{d}\right)^2 \frac{n^2}{d}\cdot\left[1+2\frac{h^2}{d^2}B_1\right]dd \text{, where}$$

$$B_1 = A_1 cthA_1\left(1-\frac{1}{A_1}\left(\frac{\pi d}{\xi}\right)^2\left(\frac{1}{A_1}-\frac{1}{shA_1 chA_1}\right)\right);$$

$$A_1 = \sqrt{\left(\frac{2\pi d}{\xi}\right)^2 - (N_c\pi)^2}\ ,\ \xi = \frac{k_F\xi_0}{\pi}.$$

$$k_{0s} > 2\pi/\xi \qquad (5)$$

$$2pdp = -2\left(\frac{\hbar\pi}{d}\right)^2 \frac{n^2}{d}\cdot\left[1+2\frac{h^2}{d^2}B_2\right]dd \text{, where}$$

$$B_2 = A_2 ctgA_2\left(1+\frac{1}{A_2}\left(\frac{\pi d}{\xi}\right)^2\left(\frac{1}{A_2}-\frac{1}{\sin A_2\cos A_2}\right)\right);$$

$$A_2 = \sqrt{(N_c\pi)^2 - \left(\frac{2\pi d}{\xi}\right)^2}\ ,\ \xi = \frac{k_F\xi_0}{\pi}.$$

For convenience and further generalization of the calculation results, we will replace: $k_{0s} \le 2\pi/\xi : B = B_1$ and $k_{0s} > 2\pi/\xi :\ B = B_2$, the change in the density of surface states dn_s can be written as:

$$dn_s = \frac{p}{2\pi\hbar^2}dp = -\frac{2\left(\frac{\hbar\pi}{d}\right)^2\frac{n^2}{d}\left[1+2\frac{h^2}{d^2}B\right]}{4\pi\hbar^2}dd = -\frac{\pi}{2}\frac{n^2}{d^3}\cdot\left[1+2\frac{h^2}{d^2}B\right]dd\ . \qquad (6)$$

It is known that the concentration of electrons in a massive sample is $n_0 = k_F^3/(3\pi^2)$, taking into account that, according Pauli principle, 2 electrons can be in one level with different spins, we get:

$$2dn_s = -2\frac{k_F^2}{2\pi d_0}\frac{n^2}{\left(\frac{k_F d_0}{\pi}\right)^2}\left[1+2B\frac{\left(\frac{k_F h_0}{\pi}\right)^2}{\left(\frac{k_F d_0}{\pi}\right)^2}\right]dd_0 = -2\frac{3k_F^3}{2\pi^2 3}\frac{1}{\left(\frac{k_F d_0}{\pi}\right)}\frac{n^2}{\left(\frac{k_F d_0}{\pi}\right)^2}\left[1+2B\frac{\left(\frac{k_F h_0}{\pi}\right)^2}{\left(\frac{k_F d_0}{\pi}\right)^2}\right]dd \quad (7)$$

As the thickness of the film d_0 increases, the number and density of electronic states will also increase $N_c = Int(k_F d_0/\pi)$, then for a certain thickness of the metal film we must sum up all the allowed electronic states formed in the film of thickness d_0. After the appropriate replacement of $d = k_F d_0/\pi$ and $h = k_F h_0/\pi$:

$$dn_s = -\frac{3n_0}{2}\frac{1}{d}\frac{\sum_{n=1}^{N_c} n^2}{d^2}\left[1+2B\left(\frac{h}{d}\right)^2\right]dd \tag{8}$$

Expression (8) allows us to describe the size-concentration dependence of the allowed states of the particle in the phase space in the interval of thicknesses for which the number of electronic states remains constant. According to (8), the number of states decreases according to the hyperbolic law with increasing d. Consider those values for which the density of states is maximal and equal to N_c. AS shown on fig. 1, when the dimensional condition is fulfilled, $N_c/d=1$, $k_z=k_F$, N_c reaches its maximum value.

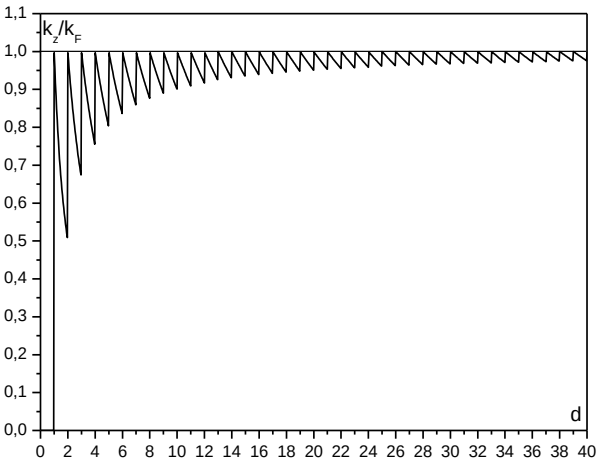

Fig. 1. Dependence of k_z/k_F – the consolidated component of the electron quasi-momentum on the consolidated thickness $d=k_F d_0/\pi$ of the metal film

At points $N_c/d=1$, we have $dn_{s_0}=\frac{3n_0}{2}\frac{N_c}{d}\left[1+2B\left(\frac{\kappa'}{d}\right)^2\right]dd$, because $dn(d)=dn_{S_0}/dd$, then:

$$n(\kappa)=\frac{3n_0N_c}{2d}\left[1+2B\left(\frac{\kappa'}{d}\right)^2\right]-\frac{3n_0\sum_{n=1}^{N_c} n^2}{2d^3}\left[1+2B\left(\frac{\kappa'}{d}\right)^2\right]. \tag{9}$$

After mathematical transformations, expression (9) will take the form:

$$\frac{n(\kappa)}{n_0}=\frac{3N_c}{2d}\left(1-\frac{N_c(N_c+1)(2N_c+1)}{6N_cd^2}\right)\left[1+2B\left(\frac{h}{d}\right)^2\right]. \tag{10}$$

When $h=0$ (which corresponds to an atomically smooth surface), expression (10) goes to the asymptote:

$$\frac{n(\kappa)}{n_0} = \frac{3N_c}{2d}\left(1 - \frac{N_c(N_c+1)(2N_c+1)}{6N_c d^2}\right). \quad (11)$$

The result (11) is similar to the expression by Trivedi and Ashcroft [5] for a plane-parallel metal film.

According to the approach of Fishman and Calecki [6-9], in the case when the concentration of electrons in a nanoscale sample is $N_c >> 1$, the expression for the conductivity σ of a metal film in the regime of the quantum-ballistic size effect takes the form:

$$\sigma \sim d^2\left(1 - \frac{6}{\left(3n\pi^5\right)^{1/3}}\frac{1}{d}\right) \sim d^{\alpha}, \quad (12)$$

here α is the ballistic charge transport index, varying in the range from 2,1 ($N_c >> 1$) to 6 ($N_c \approx 1$).

Dependences α on h for different ξ and α on for different h are shown in fig. 2 and fig. 3. The change in the coefficient α of parameters of surface inhomogeneities indicates changes in the conditions of ballistic charge transport in a nanoscale metal film. In particular, in fig. 2, we observe that when the value h of and is not constant, the indicator α increases, the reverse trend of the course is manifested when the value ξ remains constant with h increases (fig. 3).

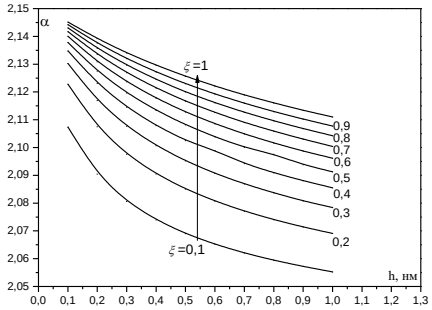

Fig. 2. Dependences of α on average amplitude of surface inhomogeneities h at different values of the lateral length ξ.

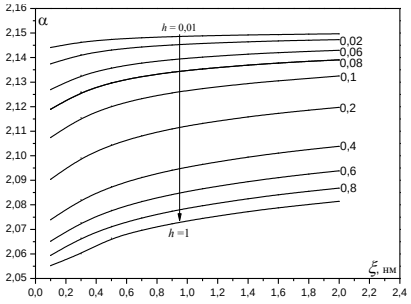

Fig. 3. Dependences of α on lateral length ξ at different values of the summed average amplitude of surface inhomogeneities h.

This approach allows us to evaluate the magnitude of change in the exponent from h and ξ, however, we predict that the influence of the parameters of the surface morphology changes the exponent α in a fairly narrow range of values. It is no coincidence that already existing models of quantum charge transport [3-9] use single-crystal $CoSi_2$ films with metallic nature of charge transport as a test object [3-4, 6, 12], which makes it possible to test these approaches [3-4 , 6, 12] on a single-crystal film with metallic nature of charge transport.

Nanoscale, single-crystal $CoSi_2$ films with metallic charge transport, the electron mean free path length reaches $\lambda \sim 100\,nm$, the surface specularity coefficient is $p \sim 90\,\%$ (the specularity coefficient p between film and silicon substrate surface is $\sim 100\,\%$), and the average amplitude of surface inhomogeneities within Soffer model [1] is $h \approx 0{,}04\,nm$. The $CoSi_2$ films can be considered homogeneous, continuous (without breaks), the structure of which meets the requirements of the plane-parallel layer model [1].

When metal film is deposited on a dielectric substrate, in particular films with a bulk thickness of 1-2 nm, the processes of coagulation of metal condensation nuclei on the surface of the substrate due to the action of surface tension forces, as well as the physical, chemical and temperature conditions of the surface of the substrate, significantly change growth conditions of a real film in comparison with plane-parallel metal film model. As mentioned above, the parameter α for gold, silver and copper films, freshly deposited at T = 78 K [10], varies in range $1{,}9 < \alpha < 3{,}1$. The use of models of quantum charge transport makes it possible to evaluate experimental data in a fairly narrow range of ξ and h values. It is possible to quantitatively describe the experimentally observed dimensional behavior by establishing the interval of electrical continuity thicknesses of the studied nano-sized, polycrystalline metal films using the percolation approach [13-14]. In the vicinity of the percolation transition, the correlation length in the mode of layer-by-layer formation of a metal film is given by power-law function [14]:

$$\xi \sim (d - d_c)^{-\gamma}, \tag{13}$$

where γ is the percolation index, d is the metal films mass thickness, d_c is metal film mass thickness at which metal islands form the first ohmic conductive channel. The dimensional behavior of the dynamic conductivity σ_ω and the dielectric constant ε of metal film in the vicinity of percolation, differs significantly from the dimensional behavior of electrically continuous or dispersed metal films. In particular, during the transition from the dispersed to the solid state film, the dielectric constant ε changes its sign from positive to negative with values that correspond to transition to the continues state of metallic condensate [13].

Conclusions

1. The impact of surface inhomogeneities parameters on percolation exponent was established, which confirms the change in the conditions of current transition in nanoscale metal film in the regime of the ballistic dimensional effect.

2. A quantum-mechanical description of ballistic charge transition in nanoscale gold, silver and copper films showed that the presence of surface inhomogeneities on nanoscale metal film, reduces concentration of conduction electrons, which leads to increase in the Fishman-Calecki parameter $\alpha > 2{,}1$ and decrease in the electrical conductivity $\sigma(d)$ of polycrystalline metal films.

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ВПЛИВ ПОВЕРХНЕВИХ НЕОДНОРІДНОСТЕЙ НА БАЛІСТИЧНЕ ПЕРЕНЕСЕННЯ ЗАРЯДУ В НАНОРОЗМІРНИХ МЕТАЛЕВИХ ПЛІВКАХ

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Проблема кількісного опису розмірних залежностей кінетичних коефіцієнтів металевих плівок нанометрової товщини актуальна при виготовленні нонорозмірних, металевих шарів з наперед заданими структурою та електричними параметрами. Якість узгодження експериментальних даних з результатами теоретичних розрахунків досягається лише при коректному виборі модельного підходу при описі явищ перенесення заряду в нанорозмірному металевому зразку. Модельний підхід повинен в достатньому обсязі,

враховувати особливості поверхневої будови плівки металу та його електронної будови. В більшості сучасних модельних підходів при описі явищ перенесення заряду в металевих шарах проводять для плівки Зоммерфельдівського металу. При цьому вважається, що плівка металу є однорідною та ізотропною з атомно-гладкими зовнішніми поверхнями (модель плоскопаралельного шару) або шаром, на поверхні якого існують неоднорідності макроскопічних розмірів, параметри яких можна описати з допомогою гармонічних чи степеневих функцій.

Квазікласичне перенесення заряду описується моделями класичного розмірного ефекту, оскільки зонна енергетична структура металевої плівки залишається аналогічною зонній енергетичні структурі масивного металевого зразку. Коли довжина вільного пробігу електрона λ стає більшою від товщини плівки металу d, квазікласичне перенесення заряду переходить в режим балістичного електронного транспорту. Особливістю цього перенесення заряду, є переважаючий вплив поверхневого розсіювання. Про особливості впливу макроскопічних поверхневих неоднорідностей на балістичне перенесення заряду в нанорозмірних плівках металі існує мало інформації. В режимі квазікласичного та класичного перенесення заряду залишкова провість лінійно зростає і з ростом товщини плівки металу $\sigma_{res} \sim d$, в той час як в режимі балістичного перенесення заряду $\sigma_{res} \sim d^{\alpha}$, де α змінюється в інтервалі $2{,}1 < \alpha < 6$.

Запропоновано модельний підхід, що дозволяє оцінити вплив параметрів середньої амплітуди h та кореляційної довжини ξ поверхневих неоднорідностей на розмірну концентрацію електронних станів $n(d)$ та питомої провідності $\sigma(d)$ нанорозмірних плівок металів. Згідно Фішмана-Цалецького [6-9], проаналізовано вплив поверхневих неоднорідностей на режим балістичного перенесення заряду в нанорозмірних плівках срібла, золота та міді. Мінімальна товщина плівки металу d_c для якої застосування підходу можливе, здійснене в рамках перколяційного підходу.

Ключові слова: тонка плівка металу, поверхневі неоднорідності, перенесення заряду в нанорозмірній плівці металу, балістичне перенесення заряду.

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