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SIZE KINETIC EFFECTS IN POLYCRYSTALLINE FILMS

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The theoretical and experimental studies of electrophysical properties (conductivity and temperature coefficient of resistance) of metal films with a polycrystalline structure are performed in this paper. It is shown, that the numerical values of these kinetic coefficients in thin conductors are essentially different from the corresponding transport coefficients in thick samples. The reason of this difference lies in the simultaneous realization in thin polycrystalline films both of the internal size effects (relaxation of charge carries at the grain boundaries) and the external size effects (electron scattering at the outer boundaries of the sample). As a result, the kinetic coefficients essentially depend on the internal structure, thickness, and degrees of surface roughness of the conductor. The size effects considered in the review are used to analyze the transport effects in polycrystalline films, and the given asymptotic expressions are used to calculate the electron transport parameters in Al *and* Ni *mono-block films. Shown, that agreement between the experimental and theoretical values of conductivity and temperature coefficient of resistance in each specific case is achieved by using the grain-boundary scattering coefficient as the adjusted parameter. The value of this coefficient is:* 0,37-0,46 *(*Ni *films) and* 0,09-0,3 *(*Al *films) while calculating the conductivity, and* 0,37-0,40 *(*Ni *films) and* 0,15-0,36 *(*Al *films) while calculating the temperature coefficient of resistance.*

Keywords: *POLYCRYSTALLINE FILM, ELECTROPHYSICAL PROPERTIES, KINETIC COEFFICIENTS, INTERNAL AND EXTERNAL SIZE EFFECTS, MAYADAS AND SHAZKES MODELS, ELECTRON TRANSPORT PARAMETERS.*

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1. INTRODUCTION

Constant interest in the study of electron transport in thin metal films is conditioned by both their wide use as an elemental basis of modern microelectronics and measuring techniques and the availability of detailed information about the relaxation of charge carries at the outer and inner boundaries of the conductor [1-3]. The main peculiarity of the electron transport in polycrystalline films in comparison with bulk samples is the interaction of electrons with outer boundaries (external size effect) and inner boundaries (internal size effect) that leads to both the size dependence of the transport coefficients and their dependence on the thin film structure (average grain

size, type of interaction between the charge carries and the grain boundaries (GB), etc.).

A sufficiently correct model of electrical conductivity of polycrystalline film was proposed for the first time by Mayadas and Shatzkes (the MS model) [4, 5]. These authors proposed the model (see Fig. 1) with the average grain size L in the conductor surface and with the grain boundaries perpendicular to the outer surfaces of the sample. Considering that the different electron scattering mechanisms (isotropic phonon and point defect scattering; intergrain boundary and outer surface scattering) are independent, Mayadas and Shatzkes [5] not only obtained the general analytical expression for the conductivity σ of polycrystalline film, but performed its approbation using the experimental results of other authors.

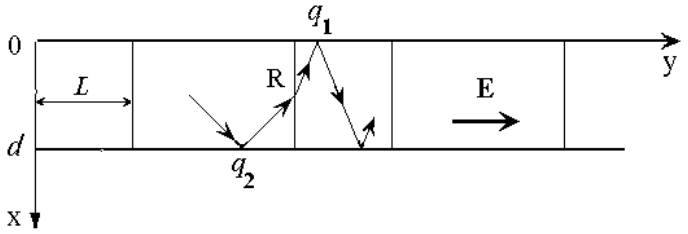


Fig. 1 *– Model of thin polycrystalline film where the possible motion path of a charge carrier is shown by the broken line: d is the film thickness; L is the average grain size in the sample surface; q_j (j = 1, 2) is the probability of specular reflection of charge carriers by j-th outer boundary of the conductor; R is the probability of the diffusion scattering at the inter-grain boundaries;* **E** *is the electrostatic field strength*

Taking the MS model as the basic one, different phenomenological models of the electrophysical properties of polycrystalline film samples were proposed in further theoretical investigations. Thus, in particular, the electrical conductivity of the conductor with mono-block crystallites of the cylindrical shape with the same diameter, generatrices of which are perpendicular to the outer boundaries, is analyzed in [6-8]. The authors of [9-11] have proposed two-dimensional and the authors of [12, 13] have proposed three-dimensional (see detailed information in the monograph [14]) models of the electrical conductivity of thin polycrystalline film under the assumption that the grain size is the same in each of three mutually perpendicular directions, and the interaction of the charge carriers with the grain boundaries is characterized by only one parameter, which determines the probability of the electron passing through the inter-grain boundary. In the sequel, the generalization of three-dimensional model in the case when crystallites have a non-cubic shape and are modeled by three sets of surfaces, which are perpendicular to the three coordinate axes, was performed in [15, 16]. But proposed in [11] numerical analysis showed that the probability values of specular reflection of charge carriers from outer surfaces of the conductor and probabilities of the diffusion electron scattering at the GB, obtained within two- and three-dimensional models, almost coincide with the corresponding results for one-dimensional model in the analysis of the experimental results. This fact is conditioned by the following: intergrain boundaries, which are parallel to the electric current density vector influence weakly the electron flow and, correspondingly, do not change the value of the transport coefficients. Therefore

analyzing the conductivity (resistivity), temperature coefficient of resistance (TCR) and tensosensitivity coefficients (TC) of thin polycrystalline film one can use one-dimensional MS model with sufficient accuracy. We have to note that in the case of ultra-thin films the scattering mechanism of the charge carriers on surface inhomogeneities becomes the dominant one [17, 18].

Mayadas and Shatzkes theory agrees satisfactorily with the experimental investigations of the kinetic properties in one- and multilayer films, it is widely used for the analysis of the electron transport in thin polycrystalline conductors of nanometer scales, and allows to determine the electron transport parameters (see, for example, [19-29]) using the experimental data about the electrophysical properties of thin polycrystalline films.

The aim of the present work is theoretical and experimental investigation of the kinetic coefficients, which characterize the electron transport in polycrystalline films.

2. CONDUCTIVITY

2.1 General analytical and asymptotic expressions

Conductivity of thin polycrystalline film, the outer boundaries of which scatter charge carries in different ways, can be written as follows [5]:

$$\frac{\sigma}{\sigma_0} \equiv \Phi = f(\alpha) - \frac{3}{\pi k}\int_0^{\pi/2} d\varphi \cos^2\varphi \int_0^1 dz \frac{(z-z^3)(1-E)}{H^2} \cdot \left\{\frac{2-q_1-q_2+(q_1+q_2-2q_1q_2)E}{1-q_1q_2E^2}\right\}, \quad (1)$$

$$E = \exp\left\{-\frac{kH}{z}\right\}, \; H = 1 + \frac{\alpha}{\cos\phi\sqrt{1-z^2}}, \; k = \frac{d}{l}, \quad (2)$$

where σ is the electrical conductivity of polycrystalline film of the thickness d; σ_0 is the conductivity of infinite ($d \to \infty$) metal monocrystalline sample; l is the mean free path of the electrons; q_j (j = 1, 2) is the probability of specular reflection of charge carriers by j-th outer boundary of the conductor with conservation of the energy and the tangential component of quasi-impulse.

Function $f(\alpha)$ in correlation (1) defines the conductivity of bulk ($d \to \infty$) polycrystalline sample and is equal to [4, 5]

$$f(\alpha) = 1 - \frac{3}{2}\alpha + 3\alpha^2 - 3\alpha^3 \ln\left(1+\frac{1}{\alpha}\right) \cong \begin{cases} 1 - \dfrac{3}{2}\alpha + 3\alpha^2, & \alpha << 1, \\ \dfrac{3}{4\alpha} - \dfrac{3}{5\alpha^2}, & \alpha >> 1. \end{cases} \quad (3)$$

In is not difficult to see from formulas (1) and (3) that in the MS model the influence of the sample polycrystallinity is taking into account using only the grain-boundary parameter $\alpha = lR/[L(1-R)]$ (L is the average grain width in the film surface; R is the probability of the diffusion electron scattering at the inter-grain boundaries), which determines the contribution of grain boundaries to the total resistance of thin film. Formula (1) for the conductivity of polycrystalline film obtained by Mayadas and Shatzkes transforms to the Lucas formula [30] (under condition that $q_1 \neq q_2$) or to the Fuchs formula [31] (if the equality $q_1 = q_2 = q$ holds) at $\alpha \to \infty$, i.e., either when the grain

boundaries are absolutely "transparent" for charge carriers ($R \to \infty$) or film is the "single-crystalline" one ($L \to \infty$).

Analytical dimensional dependence of the conductivity of polycrystalline film (1) is rather complicated for its direct comparison with the results of the experimental investigations. Therefore the authors of many theoretical papers (see the review monograph [14]) have proposed approximate relations of the Mayadas-Shatzkes formula, which contain the tabulated functions and have severe restrictions on the variation range of the parameter. General asymptotic expressions of formula (1) without tabulated functions were proposed in [32].

If the sample is rather thick, i.e., inequality $k >> 1$ holds, at the arbitrary values of the grain-boundary parameter α and reflectivity parameters q_j the conductivity of polycrystalline film can be written as [32]:

$$\frac{\sigma}{\sigma_0} = f(\alpha) - \frac{3\left(2 - q_1 - q_2\right)}{16\,k}\left\{1 - \frac{32}{3\pi}\alpha + 12\alpha^2 + \frac{16}{\pi}\left\{5 - \left(4 - 5\alpha^2\right)I\right\}\alpha^3 - 40\alpha^4\right\} \cong$$

$$\cong \begin{cases} 1 - \dfrac{3}{2}\alpha - \dfrac{3\left(2 - q_1 - q_2\right)}{16\,k}\left(1 - \dfrac{32}{3\,\pi}\alpha\right), & \alpha << 1, \\ \dfrac{3}{4\,\alpha}\left\{1 - \dfrac{2 - q_1 - q_2}{4\,k\alpha}\left(1 - \dfrac{512}{105\,\pi\alpha}\right)\right\}, & \alpha >> 1, \end{cases} \tag{4}$$

$$I = \begin{cases} \dfrac{1}{\sqrt{1-\alpha^2}} \ln \dfrac{1 + \sqrt{1-\alpha^2}}{\alpha}, & \alpha \le 1, \\ \dfrac{\arccos\left(\dfrac{1}{\alpha}\right)}{\sqrt{\alpha^2 - 1}}, & \alpha > 1. \end{cases} \tag{5}$$

It follows from (4) that the decrease in the electrical conductivity of thick polycrystalline conductor at $d >> l$ is negligible in comparison with the conductivity of bulk sample. This is connected with the fact that in this case the electrical current is formed by almost all charge carriers nearby the Fermi surface. If there is a strong correlation between the incident and reflected from the surface electron ($q_j = 1$), conductivities of film and bulk samples coincide. Absence of the mentioned correlation between the incident and reflected electron ($q_j = 0$) leads to the effective decrease in the film thickness [33], and, correspondingly, to the decrease in sample conductivity. Terms, which contain the grain-boundary parameter α in formula (4), define the influence of the grain boundaries on the electrical conductivity of thick film. Analysis of the obtained asymptotic expressions shows that in the case when inequality $\alpha \le k$ holds the charge carrier scattering at the GB in comparison with the scattering at the outer boundaries is negligible and can be neglected. In this case the film conductivity is determined by the formula

$$\frac{\sigma}{\sigma_0} = 1 - \frac{3}{16\,k}\left(2 - q_1 - q_2\right), \tag{6}$$

which coincides with the corresponding asymptotic formula for the electrical conductivity coefficient within the Fuchs-Lucas formula [30, 31].

In the case when the opposite inequality $k << 1$ holds, i.e., when the film thickness d is much less than the free path l of charge carriers, exponents, which are present in the integrand of expression (1), can be expanded in powers of k/x, and the following expression for the conductivity can be obtained in the case when inequality $\alpha \leq k$ holds:

$$\frac{\sigma}{\sigma_0} = \frac{3}{4}\frac{(1+q_1)(1+q_2)}{1-q_1q_2}\frac{d}{l}\ln\frac{l}{d}. \qquad (7)$$

In this case the external size effect is the dominant one, and the charge carrier scattering at the GB (internal size effect) is negligible and can be neglected.

Analysis of asymptotic expression (7) showed that it is more reasonable in the case $q_j << d/l$, i.e., when outer boundaries of the conductor diffusively scatter charge carriers (since $d/l << 1$). Only in this case the expansion of the expression in curly brackets of (1) in powers of k/x is justified. Taking into account this fact and performing the integration in (1) without expansion in series of the mentioned expression we obtain the following result:

$$\frac{\sigma}{\sigma_0} = \frac{3}{4}\frac{(1+q_1)(1+q_2)}{1-q_1q_2}\frac{d}{l}\ln\left\{\frac{2q_1q_2}{1+q_1q_2}+\frac{1-q_1q_2}{1+q_1q_2}\frac{l}{d}\right\}, 1-q_j << d/l \qquad (8)$$

that describes the conductivity of thin film, the outer boundaries of which reflect electrons specularly ("mirror" film by the terminology of [34]).

Presented above asymptotic expressions become "transparent" if they are obtained using the "inefficiency" conception of Pippard [35], which firstly was used for the analysis of the anomalous skin effect. In accordance with this conception all electrons can be conditionally divided into "effective" (which are responsible for the effect) and inefficient. In conditions of strong size effect ($d << l$) the contribution of the electrons to the static size effect depends on the direction of their motion with respect to the conductor boundaries. Charge carries, the velocity direction of which makes an angle $\theta < d/l$ with the conductor boundary, move parallel to the conductor surfaces and are the "effective" ones, i.e., give the main contribution to the effect. Those electrons, which move at large angles $\theta > d/l$ to the film surfaces are scattered by the conductor boundaries and are the inefficient ones.

"Effective" electrons are located in the vicinity of the Fermi surface within a belt about $\Delta p = \theta p_F \cong dp_F/l$ wide (Fig. 2). The area of this belt for the spherical Fermi surface is equal to $\Delta S \cong 2\pi p_F \Delta p \cong 2\pi d p_F^2/l$. Ratio of the number of charge carriers, which are within the mentioned belt $n_{ef} \sim \Delta S$, to the total number of electrons on the Fermi surface is given by the formula

$$\frac{n_{ef}}{n} = \frac{\Delta S}{S_F} \cong \frac{2\pi p_F^2 \frac{d}{l}}{4\pi p_F^2} \cong c\frac{d}{l}, \qquad (9)$$

where c is the numerical factor of the order of 1, and the effective conductivity conditioned by these charge carriers is equal to

$$\sigma_{ef} = \frac{e^2 l}{p_F} n_{ef} \cong \frac{e^2 l}{p_F} nc \frac{d}{l} = \sigma_0 \frac{d}{l} . \qquad (10)$$

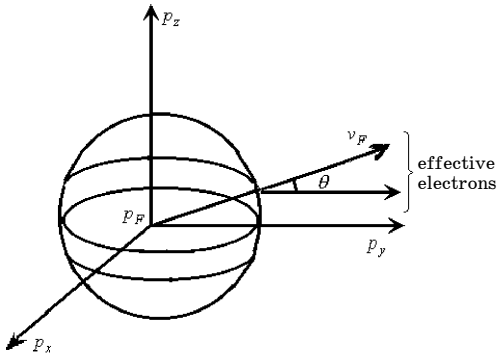


Fig. 2 *– For the calculation of the conductivity of thin film using the "inefficiency" conception of Pippard: $p_\xi(\xi = x, y, z)$ are the components of quasi-impulse; v_F and p_F are the Fermi velocity and impulse, respectively*

Thus, influence of the finiteness of metal layer thickness on its conductivity is reduced to the change of the number of effective electrons on the Fermi surface, which form current in the film and define its conductivity. In a thick film, as it was mentioned above, almost all electrons in the vicinity of the Fermi surface participate in the current formation (in other words, almost all charge carriers are the effective ones), and therefore in this case the change in the conductivity of the thick conductor is negligible in comparison with the bulk sample [31]. Limitations in the sample thickness (thin film, $d << l$) leads to the sharp decrease in a number of effective electrons (their fraction is d/l), and, correspondingly, to the substantial decrease in the conductivity of the thin film in comparison with the bulk sample (we note, leads to the decrease in a number of effective charge carriers responsible for the effect, but not in the total number of electrons). Logarithmic factors in formulas (7) and (8) define the fraction of electrons moving almost parallel to the boundaries of a thin conductor (their free path is limited only by the collisions in the conductor volume). Specular reflection of the electrons by the outer boundaries in the case of both the relatively thick and the thin films with the spherical dispersion law is strictly correlated, and therefore their conductivity will coincide with the conductivity of the bulk metal.

Presented asymptotic expressions (8) and (9) for the conductivity of the thin film allow to obtain the working formulas for the calculation of the electron transport parameters in thin film [3, 36]. But before writing them we should take into account the following circumstance. Comparison of the asymptotic formulas for the conductivity, obtained under conditions that the boundaries of the conductor scatter electrons in the same ($q_1 = q_2 = q$) and in different ($q_1 \neq q_2$) ways, shows that in the last case the interaction of charge carriers with the conductor boundaries can be described by the effective reflectivity parameter written in the form of [34]

$$q_{ef} = \begin{cases} \dfrac{q_1 + q_2}{2}, & k >> 1, \quad (10, \text{a}) \\ 1 - \dfrac{1 - q_1 q_2}{1 + \dfrac{q_1 + q_2}{2}}, & k << 1, \quad (10, \text{b}) \end{cases}$$

But since the asymptotic formula (7) is correct only if the inequality $q_j << d/l$ holds, the product $q_1 q_2$ in formula (10, b) can be neglected being the quantity of the second order of smallness, and the effective reflectivity parameters for thick film (at the arbitrary type of the interaction between the electrons and the boundaries) and thin film (at almost diffusion type of the interaction between the charge carriers and the conductor boundaries since the equality $q_j << d/l < 1$ holds) will coincide, i.e.,

$$q_{ef} = \frac{q_1 + q_2}{2}. \quad (11)$$

Taking into account correlation (11), asymptotic formulas (6) and (7) can be written as

$$\frac{\sigma}{\sigma_0} = \begin{cases} 1 - \dfrac{3l}{8d}\left(1 - q_{ef}\right), & k >> 1, \quad (12, \text{a}) \\ \dfrac{3}{4}\left(1 + 2q_{ef}\right)\dfrac{d}{l}\ln\dfrac{l}{d}, & k << 1. \quad (12, \text{b}) \end{cases}$$

If conductor is rather thick, formula for the resistivity, as it follows from (12, a), can be written in the form convenient for the processing of the experimental results taken from the dimensional dependence of the resistivity

$$\rho d = \rho_0 d + \frac{3}{8}\left(1 - q_{ef}\right)\rho_0 l, \quad (13)$$

where ρ_0 is the resistivity of a bulk ($d \to \infty$) sample with a monocrystalline structure.

With the decrease in the average grain size the probability of the charge carrier scattering at the GB increases, and the conductivity of thin conductor can be written in the form of [32]

$$\frac{\sigma}{\sigma_0} = \frac{3}{4}\frac{\left(1 + q_1\right)\left(1 + q_2\right)}{1 - q_1 q_2}\frac{d}{l}\ln\frac{l}{d} - \frac{4}{\pi}\alpha, \qquad d / l < \alpha < 1, \quad (14)$$

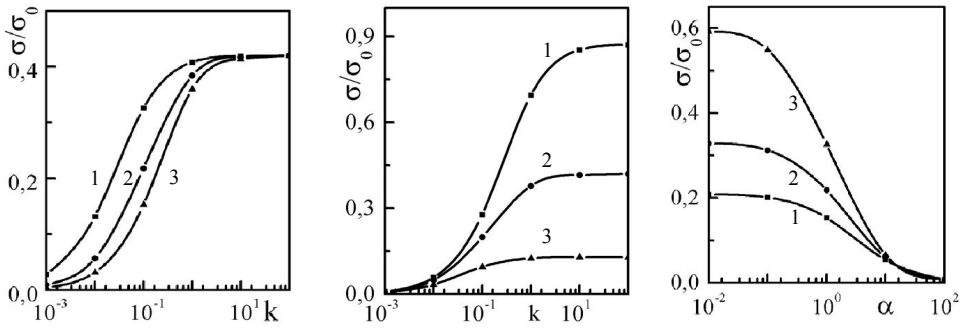
where the term $4\alpha/\pi$ defines the fraction of electrons scattered by the inter-grain boundaries, and the internal size effect becomes "commensurable" with the external size effect.

With the further growth of the parameter α in such a way that $\alpha >> 1/k$, the electron scattering at the inter-grain boundaries is the main mechanism of electron relaxation. Charge carrier scattering at the outer boundaries can be neglected in comparison with their scattering at the GB, and, correspon-

dingly, conductor will be effectively "thick". In other words, in this case the external size effect is negligible in comparison with the internal one, and the electrical conductivity coefficient of "thin" sample is given by formula

$$\frac{\sigma}{\sigma_0} = \frac{3}{4}\frac{(1+q_1)(1+q_2)}{1-q_1q_2}\frac{d}{l}\ln\frac{l}{d\,\alpha}, \qquad 1 < \alpha << l/d\,. \tag{15}$$

In Fig. 3 we present the set of lines obtained by the numerical calculation using correlation (1), which illustrate the dependence of the conductivity σ/σ_0 on the normalized film thickness k and the grain-boundary parameter α.



a b c

Fig. 3 *– Dependence of the conductivity of polycrystalline film on the normalized (to the electron free path) film thickness k (a, b) and the grain-boundary parameter α at the following parameter values: a) α = 1: 1 – q_j = 0,8, 2 – q_j = 0,4, 3 – q_j = 0,0; b) q_j = 0,1: 1 – α = 0,1, 2 – α = 1, 3 – α = 5; c) k = 0,1: 1 – q_j = 0,8, 2 – q_j = 0,4, 3 – q_j = 0,0*

2.2 Approbation of asymptotic correlation for the conductivity of polycrystalline films with a mono-block structure

Conductivity of polycrystalline film with a mono-block structure is described by expression (4) where it is necessary to take into account correlation (11)

$$\frac{\sigma}{\sigma_0} = 1 - \frac{3}{2}\alpha - \frac{3(1-q_{ef})}{8}\frac{l}{d}\left(1 - \frac{32}{3\pi}\alpha\right), \ \alpha << 1. \tag{16}$$

Dimensional dependences of the conductivity for Al and Ni films were experimentally obtained to approbate correlation (16). Films were obtained by the electron-beam (Ni) and thermal (Al) evaporation method in vacuum of the order of 10^{-3}-10^{-4} Pa on the substrate at the temperature of T_s = 300 K. Glass or glass ceramic polished plates and thin carbon films were used as the substrates while investigating the conductivity of the samples and performing the electron-microscopic investigations, respectively. The condensation rate of metal layers was 0,2-0,5 nm/s and 1,5-3 nm/s for Ni and Al films, respectively. To stabilize the electrophysical properties and for the structure recrystallization, film samples were annealed during three cycles by the scheme "heating ⇔ cooling" with constant rate in the temperature range from 300 to 680-700 K. Universal digital voltmeters were used to determine

the resistance by two- or four-point scheme. Temperature control with an accuracy of 1 K was realized using a chromel-alumel thermocouple connected with digital voltmeter. Sufficiency of this stabilization method is observed in Fig. 4. It is easier to see that the heating and cooling curves are overlapped after the first thermostabilization cycle.

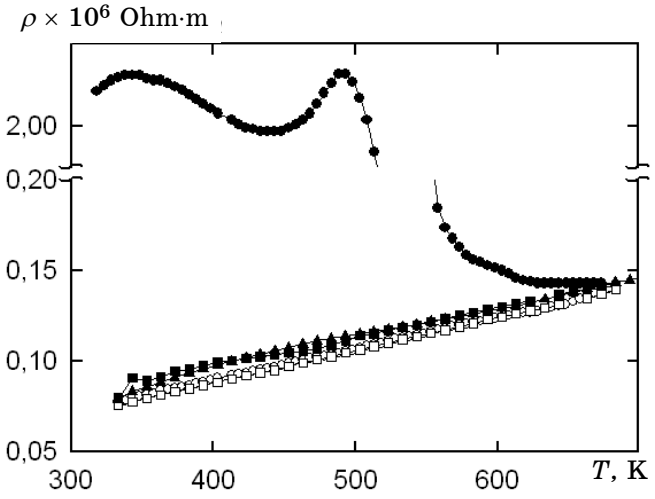


Fig. 4 *– Temperature dependence of the resistivity of Al film of the thickness d = 82 nm: ●, ○ – I cycle; ■, □ – II cycle; ▲, Δ – III cycle (dark – heating, light – cooling)*

Thickness of film samples was determined by the interferometric method (device MII-4) with an accuracy to 10%. The stainless steel masks were used to provide the iteration of the film length (a_1) and width (a_2) that allowed to calculate the resistivity using correlation $\rho = R_f a_2 d a_1^{-1}$ (R_f is the resistance).

Electron-diffraction investigations showed that Al and Ni films have the fcc-structure, and the lattice parameters for Al and Ni films are equal to a = (0,406-0,407) nm and a = (0,352-0,353) nm, respectively, that corresponds to the values a_0 = 0,406 nm (Al) and a_0 = 0,3524 nm (Ni) for bulk samples [37].

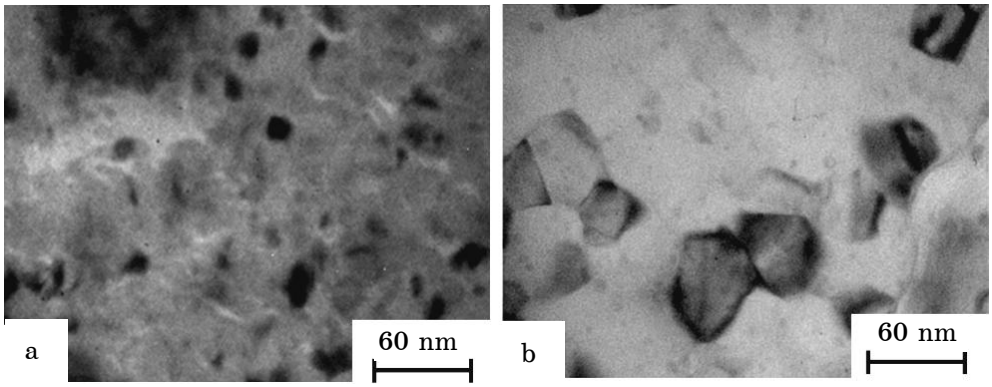


Fig. 5 *– Crystal structure of Ni (a) and Al (b) films of the thickness of 20 nm and 33 nm, respectively*

Data processing of electron microscopic investigation (Fig. 5) allowed to determine the average grain size and linear nature of its dependence on the thickness, i.e., $L \approx kd$, k is the coefficient of proportionality. For Ni films, which were thermally treated in the temperature range of 300-700 K, $k \approx 1$ that follows from both the cited publications (see, for example, [32, 38]) and our results. For Al films the coefficient k is almost equal to 1 (Fig. 6).

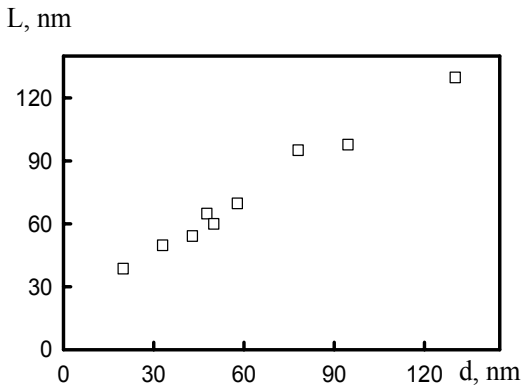


Fig. 6 *– Dependence of the average grain size on the* Al *film thickness. The annealing temperature is* 700 *K*

Analysis of the obtained results showed that Ni and Al films at the mentioned conditions of preparation and thermal treatment are the coarse-grain and mono-block in thickness and, correspondingly, inequality $\alpha << 1$ holds for these films. Therefore correlation (16) was used for the calculation of the conductivity of film samples. Here, due to the fitting of the grain-boundary scattering parameter, which in this case was as an adjusting one, the agreement between the experimental and calculated values of the conductivity was reached. Calculation of the parameter α was performed under condition that $l = const$. Note, that instead of the value σ_0 we used $\sigma_\infty = \lim_{d \to \infty} \sigma_0$, which is the conductivity of a bulk sample with the same type of defects and defect concentration as in the film. In our case this value was determined by the reconstruction of the experimental dependence of σ on d in coordinates of σ on d^{-1}. For Ni and Al films the value of σ_∞ is $7{,}7 \cdot 10^6$ $(\text{Ohm} \cdot \text{m})^{-1}$ and $1{,}47 \cdot 10^7$ $(\text{Ohm} \cdot \text{m})^{-1}$, respectively. We have taken the value of $l(1 - q_{ef})$ from papers [32] (Ni) and [39] (Al); it is equal to 33 nm and 80 nm for Ni and Al, respectively.

In Fig. 7 we present dimensional dependences of the conductivity for Al and Ni films. Agreement of the experimental and calculated values of the conductivity is observed for the case when the value of the grain-boundary scattering coefficient R varies in the range of thickness from 0,37 to 0,46 for Ni and from 0,09 to 0,3 for Al films. Comparing the obtained numerical values for the coefficient R and the known data (see, for example, [22, 40]) it is possible to see their qualitative and quantitative agreement.

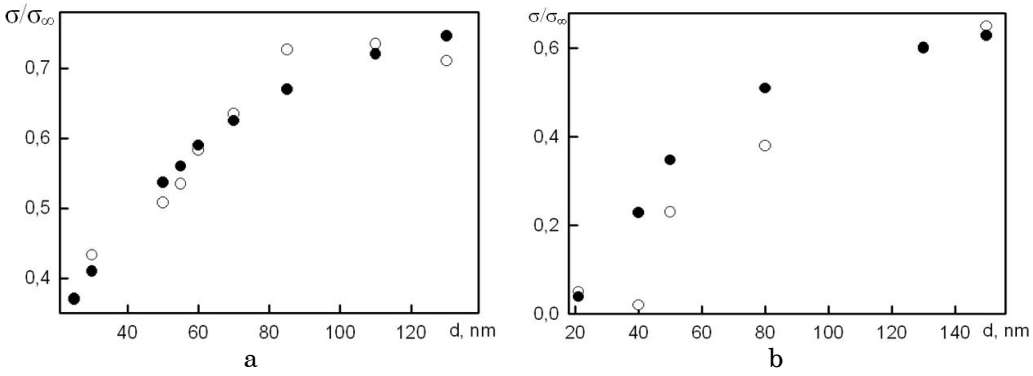


Fig. 7 *– Dependences of the conductivity on the thickness for* Ni *(a) and* Al *(b) films: ○ – experiment, ● – calculation*

3. TEMPERATURE COEFFICIENT OF RESISTANCE

3.1 Theoretical analysis of size effects in the temperature coefficient of resistance

Temperature change of the resistance R_f of polycrystalline films in conditions of the external and internal size effects is defined by the dependence of the free path l of charge carriers on the temperature, on the one hand, and by the temperature dependence of the metal layer thickness d and the average grain size L, on the other hand.

By definition the temperature coefficient of resistance β is equal to [14]

$$\beta = \frac{d \ln R_f}{d T}, \; R_f = \frac{a_1}{a_2 \, d \, \sigma}, \tag{17}$$

where a_1 and a_2 are the film length and width, respectively; σ is the film conductivity determined by expression (1).

In most of theoretical papers (see the review monographs [14, 34]) while analytically calculating the TCR it is considered that the temperature coefficient of resistance β (which is directly determined from the experimental investigations) is equal to the temperature coefficient of resistivity β_ρ, i.e., $\beta = \beta_\rho$. But this equality usually holds for monocrystalline and most of polycrystalline samples since the coefficient of thermal expansion β_T of geometric sizes of the sample is small: $\beta_T < (10^{-2}\text{-}10^{-3})\beta_\rho$ [34]. If condition $\alpha >> 1$ holds for a film sample, the temperature coefficient of thickness expansion for metal layer and crystallites can be commensurable with the temperature coefficient of resistivity, i.e., $\beta_T \sim \beta_\rho$ [34]. Therefore the TCR of polycrystalline film was determined as the change of the total resistance R_f with the temperature (17), and henceforth the effects of thermal expansion of the metal layer thickness d and the average grain size L will be taken into account.

Substituting expression (1) into correlation (17), we obtain general analytical expression for the TCR with regard to the temperature dependence of geometric sizes of the sample, coefficients q_j and R, and the temperature change of the average grain size L:

$$\beta = \beta_0 - (\chi_d - \chi_l)\frac{\partial \ln \Phi}{\partial \ln k} - \left(\chi_l - \chi_L - \frac{1}{1-R}\chi_R\right)\frac{\partial \ln \Phi}{\partial \ln \alpha} - \chi_q \frac{\partial \ln \Phi}{\partial \ln q}, \quad (18)$$

where $\chi_l = \frac{d \ln l}{dT}$, $\chi_L = \frac{d \ln L}{dT}$, $\chi_R = \frac{d \ln R}{dT}$, $\chi_q = \frac{d \ln q}{dT}$ are the coefficients, which determine the temperature change of the electron free path, average grain size, scattering coefficient of the charge carriers at the GB and Fuchs reflectivity coefficient, respectively, and the value of Φ is determined by (1).

Change in the plate thickness with temperature with regard to the substrate properties, which is deposited by the film, is defined by relation [14]

$$\chi = \chi_d \left\{1 + \frac{2\mu}{1-\mu}\left(1 - \frac{\chi_S}{\chi_d}\right)\right\}, \quad (19)$$

where $\chi_d = d\ln d/dT$ is the temperature change of the metal layer thickness; μ is the Poisson coefficient of a film conductor; χ_S is the temperature coefficient of expansion of the substrate material.

Temperature coefficient of resistivity of an infinite sample can be determined as follows [41]:

$$\beta_0 = -\frac{d \ln \sigma_0}{dT} - \chi_d, \quad (20)$$

where it is taken into account that the coefficients defining the temperature change in the film width and length coincide due to the fact that the film sizes along y and z axes are infinitely large with respect to the film thickness.

In the low-temperature range the reflection coefficients of electrons from the outer and inner boundaries do not practically depend on the temperature [19]. In the intermediate-temperature range the grain boundary evolution conditions temperature dependence of the coefficient R, but its changes are negligible. Note, that in [42] the influence of the temperature coefficients of the electron-transport parameters on the TCR value is discussed. Neglecting the temperature dependence of the coefficients q and R and substituting formula (1) into correlation (18) for the temperature coefficient of resistance we obtain the following expression at arbitrary relation between the film thickness and the electron free path and at arbitrary values of the reflectivity parameters q_j and the grain-boundary parameter α [42]:

$$\frac{\beta}{\beta_0} = 2 - \left(1 + \frac{2\chi}{\beta_0}\right)\frac{J}{\Phi} - \left(1 + \frac{\chi}{\beta_0} + \frac{\chi_L}{\beta_0}\right)\frac{J_\alpha}{\Phi} + \frac{2\chi}{\beta_0}, \quad (21)$$

where

$$J = f(\alpha) - \left\langle \frac{kH\,E}{z}\left\{\frac{G}{(1-E)} - \Theta\right\}\right\rangle, \quad (22)$$

$$J_\alpha = f^*(\alpha) + \left\langle \frac{kE}{z}(H-1)\left\{\frac{G}{(1-E)} - \Theta - \frac{z}{kEH}2G\right\}\right\rangle, \quad (23)$$

$$\Theta = \frac{2q_1q_2\left(2-q_1-q_2\right)E+\left(q_1+q_2-2q_1q_2\right)\left(1+q_1q_2E^2\right)}{1-q_1q_2E^2}, \tag{24}$$

$$f^*(\alpha) = \frac{3}{2}\alpha - \frac{3\alpha^2\left(2+3\alpha\right)}{1+\alpha} + 9\alpha^3\ln\left(1+\frac{1}{\alpha}\right) \cong \begin{cases} \dfrac{3}{2}\alpha - 6\alpha^2, & \alpha << 1, \\ \dfrac{3}{4\alpha} - \dfrac{6}{5\alpha^2}, & \alpha >> 1. \end{cases} \tag{25}$$

Correlation (21) determines the exact (within the given model) value of the temperature coefficient of resistance of thin polycrystalline film, the outer boundaries of which scatter electrons in different way. Integrals in formula (21) cannot be expressed in elementary functions, and therefore further theoretical analysis of the TCR is possible by numerical calculation only. However, for large and small values of the parameters k and α it is possible to obtain rather simple analytical expressions for the TCR, which allow to compare theory with experiment.

If polycrystalline metal layer is rather thick, i.e., $k >> 1$, for arbitrary values of the parameters q_j and α the TCR of a thick sample is defined by formula (21) where the asymptotic value of the function Φ is determined by expression (4), and the asymptotic values of the functions J and J_α can be written as follows:

$$J = f(\alpha), \tag{26}$$

$$J_\alpha = f^*(\alpha) - \frac{2\left(2-q_1-q_2\right)\alpha}{\pi\, k}\left\{1 - \frac{9\pi}{4}\alpha - 30\alpha^2 + 15\pi\alpha^3 + 6\alpha^2\left(3-5\alpha^2\right)I + \frac{3\alpha^2\left(1-\alpha^2 I\right)}{2\left(1-\alpha^2\right)}\right\}, \tag{27}$$

where the value of I is defined by the formula (5).

Obtained asymptotic formula (21) for arbitrary values of q_j and α in the case when $d >> l$ can be significantly simplified for polycrystalline samples with coarse-grain ($\alpha << 1$) and fine-grain ($\alpha >> 1$) structures

$$\frac{\beta}{\beta_0} = 1 - \frac{3}{2}\alpha - \frac{3\left(2-q_1-q_2\right)}{16\,k}\left(1-\frac{12}{\pi}\alpha\right) - \left\{\frac{3}{2}\alpha + \frac{3\left(2-q_1-q_2\right)}{8\,k}\left(1-\frac{9}{\pi}\alpha\right)\right\}\frac{\chi}{\beta_0} - \frac{3}{2}\alpha\left\{1 - \frac{3\left(2-q_1-q_2\right)}{4\pi\,k}\left(1-\frac{13}{2}\alpha\right)\right\}\frac{\chi_L}{\beta_0}, \qquad \alpha << 1, \tag{28}$$

$$\frac{\beta}{\beta_0} = \frac{4}{5\alpha} - \frac{3\left(2-q_1-q_2\right)}{32\,k\alpha^2}\left(1-\frac{3}{2\alpha}\right) - \left\{1 - \frac{4}{5\alpha} - \frac{3\left(2-q_1-q_2\right)}{8\,k\alpha}\left(1-\frac{1}{\alpha}\right)\right\}\frac{\chi}{\beta_0} - \left\{1 - \frac{4}{5\alpha} - \frac{2-q_1-q_2}{8\,k\alpha}\left(1-\frac{3}{2\alpha}\right)\right\}\frac{\chi_L}{\beta_0}, \qquad \alpha >> 1. \tag{29}$$

It follows from asymptotic correlations (28) and (29) that consideration of the thermal expansion of the metal layer thickness and the average grain width decreases the value of the temperature coefficient of total resistance of polycrystalline film samples.

For thin films ($d >> l$) with coarse-grain structure ($\alpha \le d/l$) asymptotic formulas for the temperature coefficient of resistance with different types of interaction between charge carriers and boundaries of thin conductor can be written as follows:

$$\frac{\beta}{\beta_0} = \begin{cases} \dfrac{1}{\ln\dfrac{1}{k}}\left(1+\dfrac{2\chi}{\beta_0}\right)-\dfrac{2\chi}{\beta_0}, & q_j < d/l, \\ \dfrac{1}{1+\dfrac{2q_1q_2}{1-q_1q_2}\dfrac{d}{l}}\ln^{-1}\left(\dfrac{2q_1q_2}{1+q_1q_2}+\dfrac{1-q_1q_2}{1+q_1q_2}\dfrac{l}{d}\right)\left(1+\dfrac{\chi_d}{\beta_0}\right), & 1-q_j < d/l. \end{cases} \tag{30}$$

With the increase in the grain-boundary parameter α the temperature coefficient of resistance can be estimated by the order of magnitude using the following correlations ($q_j << d/l$):

$$\frac{\beta}{\beta_0} \cong \begin{cases} \dfrac{1}{\ln\dfrac{1}{k}-\dfrac{4}{\pi}\alpha}\left[1+\dfrac{2\chi}{\beta_0}-\dfrac{4}{\pi}\alpha\left(1+\dfrac{\chi}{\beta_0}+\dfrac{\chi_L}{\beta_0}\right)\right]-\dfrac{2\chi}{\beta_0}, & \alpha > k, \\ -\dfrac{1}{\ln\dfrac{1}{\alpha k}}\left[\dfrac{3}{4\alpha^2}\left(1+\dfrac{\chi}{\beta_0}+\dfrac{\chi_L}{\beta_0}\right)+\dfrac{\chi_L}{\beta_0}-\dfrac{\chi}{\beta_0}\right]-\dfrac{2\chi}{\beta_0}, & \alpha << \dfrac{1}{k}. \end{cases} \tag{31}$$

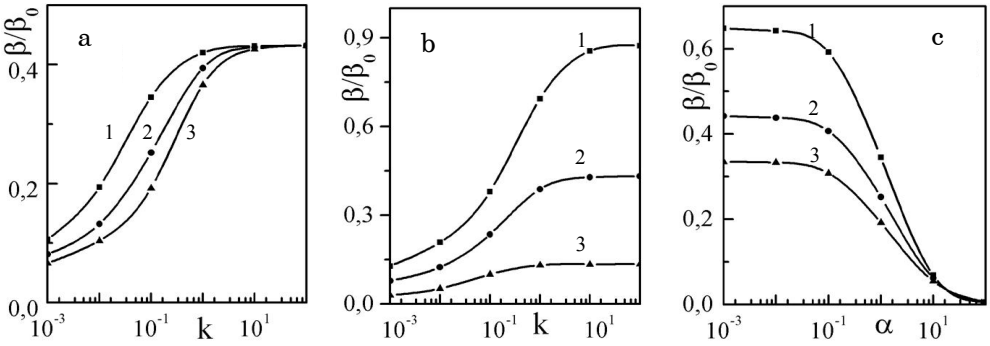


Fig. 8 *– Dependence of the temperature coefficient of resistance of a thin film with polycrystalline structure on the normalized (to the electron free path) film thickness k (a, b) and the grain-boundary parameter α (c) at the following values of the parameters* $\chi/\beta_0 = \chi_L/\beta_0 = 10^{-3}$*: a)* $\alpha = 1$*: 1 –* $q_j = 0{,}8$*, 2 –* $q_j = 0{,}4$*, 3 –* $q_j = 0{,}0$*; b)* $q_j = 0{,}3$*: 1 –* $\alpha = 0{,}1$*, 2 –* $\alpha = 1$*, 3 –* $\alpha = 5$*; c)* $k = 0{,}1$*: 1 –* $q_j = 0{,}8$*, 2 –* $q_j = 0{,}4$*, 3 –* $q_j = 0{,}0$

It follows from formulas (30) and (31) that in the presence of a conductor with large roughness on the outer boundaries the TCR does not depend on the reflectivity parameter [36], and in the case of its fine-grain structure the TCR value is negative since the electron scattering at the GB is the main mechanism of electron relaxation in this case.

In Fig. 8 we present the series of curves obtained by the numerical calculation using exact formula (31), which illustrate the dependence of the temperature coefficient of resistance of thin polycrystalline film on the normalized film thickness k and the grain-boundary parameter α.

3.2 Experimental analysis of size effects in the temperature coefficient of resistance

To approbate proposed correlations we obtained the experimental dependence of the TCR on the thickness for Ni and Al films (Fig. 9), the production method of which was described in detail in the previous subsection.

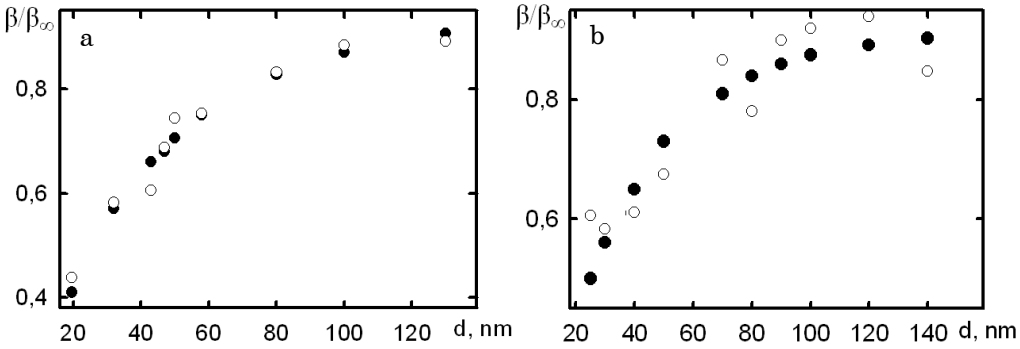


Fig. 9 *– Dependences of the temperature coefficient of resistance on the thickness for* Al *(a) and* Ni *(b) films:* ○ *– experiment,* ● *– calculation*

To process the experimental results of the dimensional dependence of the TCR one can use correlation (28) (investigated film samples partly satisfy the requirement $\alpha << 1$). Since in the considered case the values of χ_d and χ_L are of the order of 10^{-6} K^{-1} and $\beta_0 \sim 10^{-3}$ K^{-1}, two last terms in correlation (28) can be neglected, and taking into account formula (11) for the TCR of polycrystalline film we can write

$$\frac{\beta}{\beta_0} = 1 - \frac{3}{2}\alpha - \frac{3\left(1 - q_{ef}\right)}{8k}\left(1 - \frac{12}{\pi}\alpha\right). \qquad (32)$$

As in the case of the conductivity while calculating the TCR it is reasonable to use the value $\beta_\infty = \lim_{d\to\infty}\beta$ (which is the TCR of a bulk sample with the same type of defects and defect concentration as in the film) instead of β_0. For Ni and Al films the value of β_∞ is equal to $3{,}94\cdot10^{-3}$ K^{-1} and $2{,}17\cdot10^{-3}$ K^{-1}, respectively. In Fig. 9 we present the experimental and calculated dependences of the TCR for Al (a) and Ni (b) films. Agreement of the experimental and theoretical data in each specific case was achieved by the fitting of the values of the grain-boundary scattering coefficient. The values of this coef-

ficient for Ni films varied in the range of 0,37-0,40 and for Al films – in the range of 0,15-0,36. In spite of the fact that this data correlates with that obtained for the conductivity, it was possible to gain more agreement between the theoretical and experimental values of the TCR while fitting the average free path value in each point. For Al film in the thickness range of 20-140 nm $l(1 - q_{ef})$ = 80-130 nm, for Ni films in the thickness range of 25-150 nm $l(1 - q_{ef})$ = 30-80 nm. The authors of [43] analyzed the change of the average free path with the change of the average grain size. It is evident that in film samples with smaller value of the average grain size the grain boundaries impose more restrictions to the total value of the average free path than in coarse-grain films.

4. CONCLUSIONS

Based on the correlations of the Mayadas and Shatzkes theory we present the numerical calculations of the dependences of the conductivity and the TCR of metal films on the normalized thickness and the grain-boundary scattering coefficient at different values of the parameters, which characterize surface and grain-boundary electron relaxation. It is shown that strengthening of the grain-boundary and surface scattering causes changes of the mentioned coefficients.

Investigations of the dimensional dependences of the conductivity and the TCR for single-layer mono-block Ni and Al films are performed. Agreement between the experimental and calculated data of the kinetic coefficients is achieved when the grain-boundary scattering coefficient is used as the adjusting parameter. It is shown that while calculating the conductivity the value of R is 0,37-0,46 (Ni film) and 0,09-0,30 (Al film), and while calculating the TCR it is equal to 0,37-0,40 (Ni film) and 0,15-0,36 (Al film).

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