

CONDUCTIVITY AND THE TEMPERATURE COEFFICIENT OF RESISTANCE OF TWO-LAYER POLYCRYSTALLINE FILMS

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Exact and asymptotic expressions for the specific conductivity and the temperature coefficient of resistance (TCR) of two-layer metal polycrystalline films have been derived. A verification of the relations for the TCR has been performed using experimental data for (Cr, Cu, Sc)- and (Ni, Co, Cr)-based two-layer films. The theoretical and experimental results for the TCR are qualitatively consistent.

Introduction

The wide application of two- and multi-layer film systems as a basis for electronic components of contemporary microelectronics and measuring technique invokes a continuous interest to an investigation of their electrophysical properties. The main feature of electron transport in those systems, in comparison with single-layer films, is an interaction of current carriers with the layer interface (LI), which affects substantially the dependences of transport coefficients on layer thicknesses. For the first time this fact has been mentioned in [1], where resistances of two-layer films (2LFs) of Au were measured. Further experimental investigations of various transport parameters, such as resistance [2, 3], TCR [4, 5], longitudinal and transverse piezoelectric factors [6], and so on, revealed their non-monotonous dependences on layer thickness ratio $d_{2,1} = d_2/d_1$, where $d_1 = \text{const}$ and d_2 are the thicknesses of the base and upper layers, respectively.

A theoretical model of the 2LF conductivity has been put forward first in [7], where the simplest case were considered, when the LI does not affect the electron transport. Then, correct boundary conditions for a quasiclassical electron distribution function have been formulated [8, 9], and, making use of them, various transport coefficients have been calculated which characterize transport phenomena in single-crystal 2LFs. Nevertheless, metal multi-layer film

specimens, used in fabrication of microelectronic devices and sensors, possess, as a rule, a polycrystalline structure. Therefore, in addition to the external size effect, the internal size effect is also observed, which can be taken into account in the framework of the Mayadas-Shatzkes (MS) modified model [10]. Such an approach has been proposed in [11, 12]. But the general analytical expressions obtained there for the TCR [11] and polycrystalline 2LF conductivity [12] are rather cumbersome, which impedes their experimental verification, although in [4, 5] a particular case of the Dimmich's theory [11] relation has been considered, provided the absence of electron tunneling through the layer interface, and its verification has been done.

In this work, the exact and asymptotic (in the limit of very thick and very thin metal layers, in comparison with the electron mean free path) expressions for the specific conductivity and TCR of polycrystalline 2LFs have been obtained taking into account temperature variations of layer thicknesses and average crystallite dimensions. Detailed numerical calculations for a conductance and the TCR of a 2LF specimen have been carried out for various electron transport parameters, and a verification of the relations obtained has been done for (Cr, Cu, Sc)- and (Ni, Co, Cr)-based 2LFs, using experimental data.

1. Conductivity

Consider a polycrystalline 2LF with a thickness $d = d_1 + d_2$. The grain boundaries (GBs) form an ensemble of randomly positioned planes, perpendicular to the external surfaces, with the distances between the GBs being described by a Gaussian with a maximum corresponding to the average crystallite dimension L_c . The X -axis is supposed normal to the LI, while the Y - and Z -dimensions of each layer are infinite. Let an external electric field $\mathbf{E} = (0, E_y, 0)$ and a temperature

gradient $\nabla T = (0, \partial T / \partial y, 0)$ to be applied in parallel to the LI.

In order to calculate the current density

$$\mathbf{J} = \frac{2e}{\hbar^3} \sum_{i=1}^2 \int dx \int d^3 p \mathbf{v}_i f_i(|x|, \mathbf{p}), \quad (1)$$

one should resolve a Boltzmann kinetic equation for an electron distribution function $f_i(x, \mathbf{p})$ in each layer of the 2LF:

$$f_i(x, \mathbf{p}) = f_0(\varepsilon_i) - \frac{\partial f_0}{\partial \varepsilon_i} \Psi_i(x, \mathbf{p}), \quad i = 1, 2, \quad (2)$$

which, in the τ -approximation for the collision integral, reads:

$$v_{xi} \frac{\partial \Psi_i}{\partial x} + \frac{\Psi_i}{\tau_i} = \varepsilon v_{yi} \left\{ \mathbf{E} + \frac{1}{e} \frac{\varepsilon_i - \mu}{T} \frac{dT}{dy} \right\} \equiv g_i. \quad (3)$$

Here, e , x , \mathbf{p} , v_{xi} and ε_i are the charge, coordinate, quasimomentum, speed, and energy of a current carrier, respectively, μ is a chemical potential, $f_0(\varepsilon_i)$ is a Fermi distribution function, and τ_i is an effective relaxation time of electrons, which can be found, in the MS theory [10], from

$$\frac{1}{\tau_i} = \frac{1}{\tau_{0i}} \left\{ 1 + \alpha_i \frac{p_{Fi}}{|p_{yi}|} \right\}, \quad (4)$$

where τ_{0i} is a characteristics time of current carrier relaxation with respect to their collisions in the specimen bulk, p_{Fi} is an electron quasimomentum on the Fermi surface, p_{yi} is an electron quasimomentum component normal to the GBs, $\alpha_i = \frac{\lambda_i}{L_i} \frac{R_i}{1-R_i}$ is a GB parameter of the problem, λ_i is a mean free path of current carriers in the specimen bulk, R_i is a probability of electron scattering by GBs.

A general solution of the kinetic equation (3) can be found by the Lagrange multiplier method. It has the form:

$$\Psi_i(x, \mathbf{p}) = F_i e^{-\frac{x-x_s}{v_{xi}\tau_i}} + \frac{1}{v_{xi}} \int_{x_s}^x dx' g_i(\mathbf{p}) e^{-\frac{x-x'}{v_{xi}\tau_i}}, \quad (5)$$

where x_s is a coordinate of the electron scattering on either an external surface ($x_s = -d_1, d_2$) or a metal LI ($x_s = 0$).

For the sake of simplicity, a dispersion law for electrons in each of two metal layers is considered quadratic and isotropic. In this case, there is no renormalization for the chemical potential of current carriers due to their interaction with the LI and external

surfaces [8,13], so the boundary conditions for the function $\Psi_i(x, \mathbf{p})$ [equation (5)], which make it possible to determine F_i , are as follows [7-9,14]:

$$\Psi_i^{s_i}(s_j d_i, \mathbf{p}) = q_i \Psi_i^{s_j}(s_j d_i, \mathbf{p}'), \quad (6)$$

$$\Psi_i^{s_j}(0, \mathbf{p}) = P_{ij} \Psi_i^{s_i}(0, \mathbf{p}') + Q_{ji} \Psi_j^{s_j}(0, \mathbf{p}''). \quad (7)$$

Here, q_i is a specularity parameter of Fuchs [14], being equal to a probability of electron scattering with a conservation of energy and a momentum component, which is tangential to the i -th external surface of the 2L specimen, P_{ij} is a probability of the specular reflection of current carriers from the interface between the i -th and j -th metal layers, Q_{ji} is a probability of the electron transmission from the i -th into j -th layer without scattering, $P_{ij} + Q_{ij} \leq 1$. Quasimomenta \mathbf{p} , \mathbf{p}' , and \mathbf{p}'' are interconnected by conversation conditions for energy and the quasimomentum component tangential with respect to the external surfaces and the LI, $s_i = \text{sign } v_{xi}$ and indicates the sign of a speed component v_{xi} of current carriers that is normal to the external surfaces and the LI.

Substituting functions $\Psi_i(x, \mathbf{p})$ in the form (5) into the boundary conditions (6), (7), a set of linear algebraic equations is obtained for F_i . Knowing the electron distribution functions $\Psi_i(x, \mathbf{p})$ in each layer of the 2LF, one can evaluate the current density \mathbf{J} (1). From a comparison of this result with the relation [15]

$$\mathbf{J} = \sigma \mathbf{E}, \quad (8)$$

the following expression for a specific conductivity σ of a polycrystalline 2LF is obtained:

$$\sigma = \frac{1}{d} \sum_{i=1}^2 d_i \sigma_{0i} \Phi_i. \quad (9)$$

Here, σ_{0i} is a specific conductivity of a specimen with single-crystalline structure and without LI, and Φ_i is a function which makes allowance for an influence of layer dimensions on the 2LF conductivity. The latter equals

$$\Phi_i = f(\alpha_i) - \langle G_i \rangle, \quad (10)$$

where

$$\begin{aligned} G_i = & \frac{1}{\Delta} \{ (2 - q_i - P_{ij} + E_i(q_i + P_{ij} - 2q_i P_{ij})) \times \\ & \times (1 - q_j P_{ji} E_j^2) - q_j Q_{ij} Q_{ji} E_j^2 (1 - E_i + 2q_i E_i) - \\ & - Q_{ji} \tau_{j,i} (1 - E_j) (1 + q_i E_i) (1 + q_j E_j) \}, \quad (11) \\ \Delta = & 1 - q_i P_{ij} E_i^2 - q_j P_{ji} E_j^2 - \end{aligned}$$

$$-q_i q_j (Q_{ij} Q_{ji} - P_{ij} P_{ji}) E_i^2 E_j^2, \tag{12}$$

$$E_i = \exp \left\{ -\frac{k_i H_i}{x} \right\}, \quad H_i = 1 + \frac{\alpha_i}{\cos \varphi \sqrt{1-x^2}},$$

$$k_i = \frac{d_i}{\lambda_i}, \quad \tau_{j,i} = \frac{\tau_{0j} H_i}{\tau_{0i} H_j} \equiv \tau_{0j,i} H_{i,j}, \tag{13}$$

(...) =

$$= \frac{3}{\pi k_i} \int_0^{\pi/2} d\varphi \cos^2 \varphi \int_0^1 dx \frac{(x-x^3)(1-E_i)}{H_i^2(x,\varphi)} \left\{ \dots \right\} \tag{14}$$

If $P_{ij} = P_{ji} = P$ and $Q_{ij} = Q_{ji} = Q$, functions G_i coincide with the relevant relations from [14].

The function $f(\alpha_i)$ describes the conductivity of infinitely thick specimens with polycrystalline structure and, in the MS model [10], is as follows:

$$f(\alpha_i) = 1 - \frac{3}{2} \alpha_i + 3\alpha_i^2 - 3\alpha_i^3 \ln \left(1 + \frac{1}{\alpha_i} \right) \cong \begin{cases} 1 - \frac{3}{2} \alpha_i + 3\alpha_i^2, & \alpha_i \ll 1, \\ \frac{3}{4\alpha_i} - \frac{3}{5\alpha_i^2}, & \alpha_i \gg 1. \end{cases} \tag{15}$$

If the LI in the 2LF is opaque for current carriers ($Q_{ij} = Q_{ji} = 0$), then the function Φ_i in each layer is independent of parameters of the adjacent layer, and probabilities P_{ij} and P_{ji} are identical to the Fuchs' specularly parameters [14]. In this instance, if relevant parameters characterizing polycrystalline metal layers coincide ($q_i = q_j$, $P_{ij} = P_{ji}$, $\alpha_i = \alpha_j$, $d_i = d_j$, $\lambda_i = \lambda_j$), the 2L polycrystalline specimen can be regarded as a single polycrystalline metal layer, whose conductivity is described by the generalized formula of Mayadas and Shatzkes [10] in the case where the external surfaces of a thin layer scatter electrons differently with effective specularly parameters $q_{e\pi} = q_i$, P_{ij} [16]. If the current carrier scattering is absent both on the LI ($P_{ij} + Q_{ij} = 1$) and on the external surfaces ($q_i = 1$), and provided that the layers are identical both by purity ($\lambda_i = \lambda_j$) and structure ($\alpha_i = \alpha_j$), a numerical value of the 2LF conductivity is equal to the bulk conductivity value, and the 2LF can be formally considered as a bulk specimen, whose conductivity is described by (15). And vice versa, if the LI is absolutely transparent for electrons ($Q_{ij} = Q_{ji} = 1$) and equations $\lambda_i = \lambda_j$ and $\alpha_i = \alpha_j$ are in action, the 2LF conductivity is determined by the MS generalized formula [10] describing the conductivity of a uniform polycrystalline film with a thickness $d = d_1 + d_2$,

i.e. the 2L specimen may be regarded once more as a single-layer film of a thickness d [16].

Thus, a general analytical expression (9) for the 2LF conductivity was obtained. Integrals, entering into (9), cannot be expressed through elementary functions, so the further analysis of the 2LF conductivity is possible only on the basis of numerical calculations. Nevertheless, for large and small values of parameters k_i and α_i , rather simple asymptotic formulas for the specific conductivity σ can be derived, which make much easier a comparison of theoretical and experimental results.

If $k_i \gg 1$, i.e. if a mean free path of electrons λ_i is much less than the layer thicknesses d_i , the exponents in (9) are small and may be neglected. Integrating over φ and x , the following formula for the thickness-depending function Φ_i is obtained, being valid for arbitrary values of the parameters q_i , P_{ij} , Q_{ji} , and α_i :

$$\Phi_i = f(\alpha_i) - \frac{3}{16k_i} \{ (2 - q_i - P_{ij}) \Gamma_{1,i} - Q_{ji} \tau_{0j,i} \Gamma_{2,i} \}, \tag{16}$$

$$\Gamma_{1,i} = 1 - \frac{32}{3\pi} \alpha_i + 12\alpha_i^2 + \frac{16}{\pi} \alpha_i^3 \{ 5 - (4 - 5\alpha_i^2) I_i \} - 40\alpha_i^4, \tag{17}$$

$$\Gamma_{2,i} = 1 - \frac{16}{3\pi} \left\{ \alpha_i + \alpha_j - \frac{3\pi}{4} (\alpha_i^2 + \alpha_i \alpha_j + \alpha_j^2) - 3(\alpha_i^3 + \alpha_i^2 \alpha_j + \alpha_i \alpha_j^2 + \alpha_j^3) + \frac{3\pi}{2} (\alpha_i^4 + \alpha_i^3 \alpha_j + \alpha_i^2 \alpha_j^2 + \alpha_i \alpha_j^3 + \alpha_j^4) + \frac{3}{\alpha_i - \alpha_j} [\alpha_i^4 (1 - \alpha_i^2) I_i - \alpha_j^4 (1 - \alpha_j^2) I_j] \right\}, \tag{18}$$

$$I_i = \begin{cases} \frac{1}{\sqrt{1-\alpha_i^2}} \ln \frac{1+\sqrt{1-\alpha_i^2}}{\alpha_i}, & \alpha_i \leq 1; \\ \frac{\arccos(\frac{1}{\alpha_i})}{\sqrt{\alpha_i^2-1}}, & \alpha_i > 1. \end{cases} \tag{19}$$

For a 2LF, the polycrystalline layers of which have the same structure ($\alpha_i = \alpha_j$), $\Gamma_{1,i} = \Gamma_{2,i}$ and Eq. (16) becomes much simpler:

$$\Phi_i = f(\alpha_i) - \frac{3}{16k_i} \{ 2 - q_i - P_{ij} - Q_{ji} \tau_{0j,i} \} \Gamma_{1,i}. \tag{20}$$

If polycrystalline metal layers are composed of crystallites, whose dimensions L_i are much greater than the electron mean free path λ_i ($L_i \gg \lambda_i$), or the GBs are almost transparent for current carriers ($R_i \ll 1$), then $\alpha_i \ll 1$. In the inverse case, $\alpha_i \gg 1$, either the 2L specimen has a fine-grained structure ($L_i \ll \lambda_i$) or the

GBs are almost opaque for current carriers ($1 - R_i \ll 1$). For those limiting cases of the parameter α_i , Eq. (16) reads:

$$\Phi_i = 1 - \frac{3}{2}\alpha_i - \frac{3}{16k_i} \left\{ (2 - q_i - P_{ij}) \left(1 - \frac{32}{3\pi}\alpha_i \right) - Q_{ji}\tau_{0j,i} \left(1 - \frac{16}{3\pi}(\alpha_i + \alpha_j) \right) \right\} \text{ для } \alpha_i \ll 1, \quad (21)$$

$$\Phi_i = \frac{3}{4\alpha_i} \left\{ 1 - \frac{1}{8k_i\alpha_i} \left[(2 - q_i - P_{ij}) \left(1 - \frac{512}{105\pi\alpha_i} \right) - Q_{ji}\tau_{0j,i} \frac{\alpha_i}{\alpha_j} \left(1 - \frac{256}{105\pi} \frac{\alpha_i + \alpha_j}{\alpha_i\alpha_j} \right) \right] \right\} \text{ для } \alpha_i \gg 1. \quad (22)$$

But if the layer thicknesses d_i are much less than the electron mean free path λ_i in the layer, i.e. $k_i \ll 1$, the following approximations can be obtained for Φ_i :

$$\Phi_i \approx \frac{3}{4} \frac{(1 + q_i) [(1 + P_{ij})(1 - q_j P_{ji}) + q_j Q_{ij} Q_{ji} + (1 + q_j) Q_{ji} d_{j,i}]}{(1 - q_i P_{ij})(1 - q_j P_{ji}) - q_i q_j Q_{ij} Q_{ji}} k_i \begin{cases} \ln \frac{1}{k_i}, & \alpha_i \leq k_i, \\ \left(\ln \frac{1}{k_i} - \frac{4}{\pi} \alpha_i \right), & k_i < \alpha_i \ll 1, \\ \ln \frac{1}{\alpha_i k_i}, & 1 < \alpha_i \ll \frac{1}{k_i}. \end{cases} \quad (23a)$$

$$(23a)$$

$$(23b)$$

$$(23c)$$

From the viewpoint of forming the ultimate value of the 2LF conductivity, as well as that of a thin single-layer film [14], there is a group of "effective" (responsible for the result, see the Pippard's concept of "nonefficiency" [17]) current carriers which move in parallel to the external and internal specimen interfaces and have no scattering on them within the mean free path. The fraction of such electrons is of the order of d_i/λ_i . The factor $\ln \frac{1}{k_i}$ takes into account a contribution to the 2LF conductivity of the electrons, which move almost in parallel with the LI and external surfaces, and collide with them.

In the case $\alpha_i \leq k_i$, the electron scattering on the interface and external surfaces is the main mechanism of electron relaxation, and the polycrystalline specimen can be considered as a single-crystal one, since the contribution of electrons, which are scattered on crystal interfaces, to the 2LF conductivity is negligible [18]. Along with the increasing of the GB parameter α_i , the contribution of the current carriers to σ (their fraction is equal to $\frac{4}{\pi}\alpha_i$, see Eq. (23b)), which are scattered on the GBs, also increases. If the parameter α_i rises further and attends values $\alpha_i \gg 1/k_i$, the scattering of electrons on crystal interfaces (see equation (23c)) becomes the main mechanism of their relaxation.

For numerical calculations, it is convenient to write down the 2LF conductivity (9) in the form:

$$\frac{\sigma}{\sigma_{01}} = \frac{\Phi_1}{1 + d_{2,1}} \{1 + B_{2,1}\}, \quad (24)$$

the asymptotics of which for $d_{j,i} \ll 1$ is as follows:

$$\sigma = \sigma_{0i}\Phi_i \{1 - d_{j,i} + B_{j,i}\}, \quad (25)$$

where

$$B_{j,i} = \frac{d_j \sigma_{0j} \Phi_j}{d_i \sigma_{0i} \Phi_i}. \quad (26)$$

The curves depicted in Fig. 1, which were obtained by numerical calculations according to the exact formula (24), illustrate the dependence of the 2LF conductivity, normalized by σ_{01} , on the ratio $d_{2,1}$ of the metal layer thicknesses for various values of 2LF parameters ($k_2 = k_1 d_{2,1} \lambda_{2,1}$). The dependences $\sigma(d_{2,1})$ show that in the range of small $d_{2,1} \ll 1$, the 2LF conductivity is determined by a character of the interaction of current carriers with the LI, while for $d_{2,1} \gg 1$, the value of σ/σ_{01} is determined by the ratio $\lambda_{2,1}$ of the electron mean free paths in metals and by the bulk value of the upper layer conductivity $f(\alpha_2)$, i.e. $\frac{\sigma}{\sigma_{01}} \cong \frac{\lambda_2}{\lambda_1} f(\alpha_2)$. For $d_2 \sim d_1$, a minimum appears in the dependence $\frac{\sigma}{\sigma_{01}}$ vs $d_{2,1}$, which is stipulated by a competition of bulk, surface, and GB scattering contributions to the conductivity.

To simplify an interpretation of numerical results, let us suppose that the crystal interfaces are almost transparent for current carriers ($R_i \ll 1$), while the layer interface and external surfaces scatter electrons diffusively ($P_{ij}, Q_{ij} \ll 1$). In this instance, we may admit that $F_i \sim k_i$, $B_i \sim d_{2,1}^2$, so Eq. (24) acquires the form [19]:

$$\frac{\sigma}{\sigma_{01}} \cong \frac{k_1}{1 + d_{2,1}} \{1 + d_{2,1}^2\}. \quad (27)$$

On investigating Eq. (27) for an extremum, one can find that the conductivity of a 2L polycrystalline

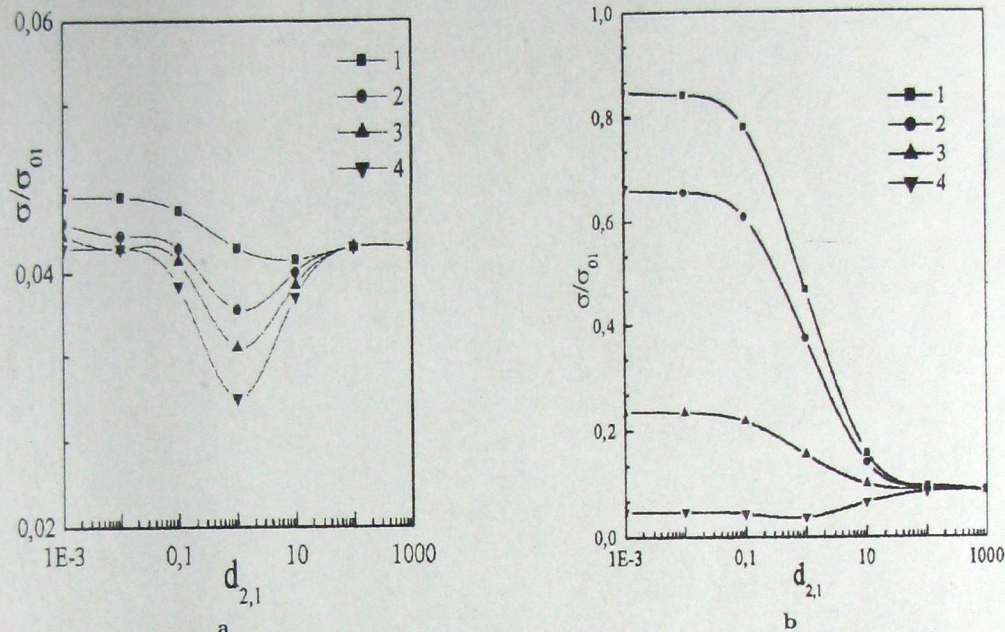


Fig. 1. Dependences of the normalized specific conductivity σ/σ_{01} of the two-layer plate on the ratio of thicknesses of the adjacent layers $d_{2,1}$ in the case $Q_{12} = Q_{21} = Q$ and $P_{12} = P_{21} = P$, and for various parameters: a - $q_1 = q_2 = 0.2$, $P = 0.2$, $\lambda_{1,2} = 10$, $k_1 = 0.01$, $\alpha_1 = \alpha_2 = 1$, $Q = 0.8$ (1), 0.5 (2), 0.3 (3), and 0 (4); b - $q_1 = q_2 = 0.1$, $P = 0.2$, $Q = 0.1$, $\lambda_{1,2} = 10$, $\alpha_1 = \alpha_2 = 0.1$, $k_1 = 10$ (1), 1 (2), 0.1 (3), and 0.01 (4)

specimen is minimal at $d_{2,1 \min} \cong 0.414$, which was confirmed by numerical calculations. A shift of $d_{2,1 \min}$ towards greater values (this is also observed in numerical calculations) is due to the contribution to the 2LF conductivity of electrons moving almost in parallel to the LI and external surfaces which is made allowance for, and because the specimen has a polycrystalline structure.

With the increasing of either the tunneling probability of current carriers into the adjacent layer without scattering (Fig. 1,a) or the base layer thickness (Fig. 1,b), this minimum degenerates and the conductivity of the 2L polycrystalline specimen monotonously decreases with increase in $d_{2,1}$.

2. Temperature Coefficient of Resistance

The temperature change of a resistance R_f of the polycrystalline 2LF, when the conditions for external and internal size effect manifestations are favorable, is determined by a temperature dependence of the mean free path of current carriers λ_i , on the one hand, and by

a temperature dependences of the specimen geometric dimensions and the average crystallite diameter L_i in the 2LF plane (hereafter, the parameters q_i , P_{ij} , P_{ji} , Q_{ij} , Q_{ji} , and R_i are assumed temperature-independent). The TCR equals [20]

$$\beta = \frac{d \ln R_f}{dT}, \quad (28)$$

where

$$R_f = \frac{1}{\sigma} \frac{b}{ad}. \quad (29)$$

Here, a and b are the specimen width and length, respectively, and σ is the 2LF specific conductivity determined by (9).

Substituting (9) into (29), and the result of substitution into (28), a general analytical formula is obtained for the TCR of a two-layer film, taking into account the temperature variations of the specimen geometric dimensions and the average crystallite size:

$$\beta = \sum_{i \neq j} \frac{\beta_{0i}}{1 + B_{j,i}} \left\{ 1 - \left(1 + \frac{2\chi_{di}}{\beta_{0i}} \right) \frac{\partial \ln \Phi_i}{\partial \ln k_i} \right\}$$

$$-\beta_{0j,i} \left(1 + \frac{2\chi_{dj}}{\beta_{0j}} \right) \frac{\partial \ln \Phi_i}{\partial \ln k_j} + \left(1 + \frac{\chi_{di}}{\beta_{0i}} + \frac{\chi_{Li}}{\beta_{0i}} \right) \frac{\partial \ln \Phi_i}{\partial \ln \alpha_i} - \Theta_i - \frac{x}{k_i E_i H_i} (\Lambda_i + 2G_i) \Bigg\}, \quad (37)$$

$$+\beta_{0j,i} \left(1 + \frac{\chi_{dj}}{\beta_{0j}} + \frac{\chi_{Lj}}{\beta_{0j}} \right) \frac{\partial \ln \Phi_i}{\partial \ln \alpha_j} - \left(\left(1 + \frac{\chi_{di}}{\beta_{0i}} \right) - \right. \quad J_{\alpha_i}^* = \left\langle \frac{k_j E_j}{x} (H_j - 1) \left\{ \Theta_i^* + \frac{x}{k_j E_j H_j} \Lambda_i \right\} \right\rangle, \quad (38)$$

$$\left. -\beta_{0j,i} \left(1 + \frac{\chi_{dj}}{\beta_{0j}} \right) \right) \frac{\partial \ln \Phi_i}{\partial \ln \tau_{0j,i}} \Bigg\}, \quad (30) \quad J_{\tau_i} = \langle \Lambda_i \rangle; \quad (39)$$

where function $B_{j,i}$ is determined by (26), $\beta_{0j,i} = \beta_{0j}/\beta_{0i}$ are the ratio of the TCR bulk values of the layers, $\chi_{di} = \frac{d \ln d_i}{dT}$ and $\chi_{Li} = \frac{d \ln L_i}{dT}$ are phenomenological parameters determining the temperature variation of the layer width d_i and the average grain size L_i , respectively. The temperature coefficient β_{0i} of the total resistance of a specimen without LI equals [20,21]:

$$\beta_{0i} = -\frac{d \ln \sigma_{0i}}{dT} - \frac{d \ln d_i}{dT}. \quad (31)$$

Substituting the thickness-dependent functions Φ_i in the form (10) into Eq. (30), the exact value for the TCR of the 2LF is obtained for any relation between the mean free path λ_i of electrons and the layer thicknesses d_i , and for any character of current carrier interaction with the internal and external interfaces of the specimen:

$$f^*(\alpha_i) = \frac{3}{2} \alpha_i - \frac{3\alpha_i^2(2+3\alpha_i)}{1+\alpha_i} + 9\alpha_i^3 \ln \left(1 + \frac{1}{\alpha_i} \right) \cong \begin{cases} \frac{3}{2} \alpha_i - 6\alpha_i^2 & \alpha_i \ll 1, \\ \frac{3}{4\alpha_i} - \frac{6}{5\alpha_i^2} & \alpha_i \gg 1. \end{cases} \quad (40)$$

$$\Theta_i = \{ (q_i + P_{ij} - 2q_i P_{ij})(1 - q_j P_{ij} E_j^2) + q_j(1 - 2q_i) Q_{ij} Q_{ji} E_j^2 - q_i Q_{ji} \tau_{j,i} (1 - E_j)(1 + q_j E_j) \} \Delta^{-1} + G_i U_i, \quad (41)$$

$$\Theta_i^* = \{ 2q_j P_{ij} E_j (2 - q_i - P_{ij} + E_i(q_i + P_{ij} - 2q_i P_{ij})) + 2q_j Q_{ij} Q_{ji} E_j (1 - E_i + 2q_i E_i) - Q_{ji} \tau_{j,i} (1 + q_i E_i)(1 - q_j + 2q_j E_j) \} \Delta^{-1} - G_i U_j, \quad (42)$$

$$U_i = 2q_i E_i \{ P_{ij} + q_j (Q_{ij} Q_{ji} - P_{ij} P_{ji}) E_j^2 \} \Delta^{-1}, \quad U_i = 2q_i E_i \{ P_{ij} + q_j (Q_{ij} Q_{ji} - P_{ij} P_{ji}) E_j^2 \} \Delta^{-1}, \quad (43)$$

$$\Lambda_i = Q_{ji} \tau_{j,i} (1 - E_j)(1 + q_i E_i)(1 + q_j E_j) \Delta^{-1}. \quad (43)$$

Functions G_i , E_i , H_i , $f(\alpha_i)$, and angle brackets were defined by Eqs. (11)–(15). The exact expression (32) for the TCR of a two-layer film can be simplified for the limiting cases of thick ($k_i \gg 1$) and thin ($k_i \ll 1$) layers of 2LFs.

For $k_i \gg 1$, the TCR of a 2L specimen for arbitrary values of q_i , P_{ij} , Q_{ij} , and α_i is determined again by (32), where functions J_{di} , J_{di}^* , $J_{\alpha i}$, $J_{\alpha i}^*$, and $J_{\tau i}$ can be written as

$$J_{di} = f(\alpha_i); \quad J_{di}^* = 0; \quad (44)$$

$$J_{\alpha i} = f^*(\alpha_i) - \frac{\alpha_i}{\pi k_i} \{ 2(2 - q_i - P_{ij}) \Gamma_{3,i} - Q_{ji} \tau_{0j,i} \Gamma_{4,i} \}, \quad (45)$$

$$J_{\alpha i}^* = \frac{\alpha_j}{\pi k_i} Q_{ji} \tau_{0j,i} \Gamma_{4,j}; \quad J_{\tau i} = \frac{3}{16k_i} Q_{ji} \tau_{0j,i} \Gamma_{2,i}. \quad (46)$$

Here,

$$\Gamma_{3,i} = 1 - \frac{9\pi}{4} \alpha_i - 30\alpha_i^2 + 15\pi\alpha_i^3 +$$

$$\begin{aligned}
 & 6\alpha_i^2 (3 - 5\alpha_i^2) I_i + \frac{3\alpha_i^2 (1 - \alpha_i^2 I_i)}{2(1 - \alpha_i^2)}, \\
 & \Gamma_{4,i} = 1 - \frac{3\pi}{4}(2\alpha_i + \alpha_j) + \\
 & + \frac{3\pi}{2} \{4\alpha_i^3 + 3\alpha_i^2 \alpha_j + 2\alpha_i \alpha_j^2 + \alpha_j^3\} \times \\
 & \times \left\{ 1 - \frac{2}{\pi} \frac{1 - (1 - \alpha_i^2) I_i}{\alpha_i} \right\} - \frac{3\alpha_j^4}{\alpha_i(\alpha_i - \alpha_j)} \times \\
 & \times \left\{ 1 + \frac{\alpha_j(1 - \alpha_i^2) I_i - \alpha_i(1 - \alpha_j^2) I_j}{\alpha_i - \alpha_j} \right\} - \frac{3\alpha_i^3 I_i}{\alpha_i - \alpha_j}. \quad (48)
 \end{aligned}$$

It should be noted that if the equality $\alpha_i = \alpha_j$ is valid, $\Gamma_{3,i} = \Gamma_{4,i}$.

The derived asymptotic formula for the temperature coefficient of resistance of 2LF, valid for arbitrary α_i , can be greatly simplified for the limiting values of the parameter. If $\alpha_i \ll 1$ or $\alpha_i \gg 1$, then the TCR of a 2L specimen is determined again by Eq. (32) for any value of the layer thickness ratio $d_{j,i}$, where functions M_i and M_i^* are as follows:

$$\begin{aligned}
 M_i &= 1 - \frac{3}{2}\alpha_i - \frac{3}{16k_i} \left\{ (2 - q_i - P_{ij}) \left(1 - \frac{12}{\pi}\alpha_i \right) + \right. \\
 & + \frac{3}{5\pi} Q_{ji} \tau_{0j,i} \alpha_i \left. \right\} - \left\{ \frac{3}{2}\alpha_i + \frac{3}{8k_i} \left[(2 - q_i - P_{ij}) \times \right. \right. \\
 & \times \left. \left. \left(1 - \frac{9}{\pi}\alpha_i \right) - \frac{1}{2} Q_{ji} \tau_{0j,i} \left(1 - \frac{16}{3\pi} \left(\alpha_j + \frac{1}{4}\alpha_i \right) \right) \right] \right\} \times \\
 & \times \frac{\chi_{di}}{\beta_{0i}} - \frac{3}{2}\alpha_i \left\{ 1 - \frac{3}{4\pi k_i} \left[(2 - q_i - P_{ij}) \left(1 - \frac{13}{2}\alpha_i \right) - \right. \right. \\
 & \left. \left. \frac{1}{10} Q_{ji} \tau_{0j,i} \left(1 - \frac{7\pi}{3}(\alpha_i + \alpha_j) \right) \right] \right\} \frac{\chi_{Li}}{\beta_{0i}}, \quad \alpha_i \ll 1, \quad (49)
 \end{aligned}$$

$$\begin{aligned}
 M_i^* &= \frac{3}{16k_i} Q_{ji} \tau_{0j,i} \left\{ \left[1 - \frac{32}{3\pi} \left(\alpha_j + \frac{1}{16}\alpha_i \right) \right] \left(1 + \frac{\chi_{dj}}{\beta_{0j}} \right) - \right. \\
 & - \frac{16}{3\pi} \alpha_j \left[1 - \frac{3\pi}{4} \left(2\alpha_j + \frac{9}{25}\alpha_i \right) \right] \frac{\chi_{Lj}}{\beta_{0j}} \left. \right\}, \quad \alpha_i \ll 1, \quad (50)
 \end{aligned}$$

$$M_i = \frac{4}{5\alpha_i} - \frac{3}{16k_i \alpha_i^2} \left\{ (2 - q_i - P_{ij}) \left(1 - \frac{8}{5\alpha_i} \right) - \right.$$

$$\begin{aligned}
 & \left. - \frac{\pi}{100} Q_{ji} \tau_{0j,i} \frac{\alpha_i}{\alpha_j} \right\} - \left\{ 1 - \frac{4}{5\alpha_i} + \frac{1}{4k_i \alpha_i} \left[(2 - q_i - P_{ij}) \times \right. \right. \\
 & \times \left. \left. \left(1 - \frac{3}{5\alpha_i^2} \right) - Q_{ji} \tau_{0j,i} \frac{\alpha_i}{\alpha_j} \left(1 - \frac{\pi}{4\alpha_j} + \frac{\pi}{5\alpha_i^2} \right) \right] \right\} \frac{\chi_{di}}{\beta_{0i}} - \\
 & - \left\{ 1 - \frac{4}{5\alpha_i} - \frac{1}{4k_i \alpha_i} \left[(2 - q_i - P_{ij}) \left(1 - \frac{3}{2\alpha_i} \right) - \right. \right. \\
 & \left. \left. - \frac{\pi}{135\alpha_j} Q_{j0j,i} \right] \right\} \frac{\chi_{Li}}{\beta_{0i}}, \quad \alpha_i \gg 1, \quad (51)
 \end{aligned}$$

$$\begin{aligned}
 M_i^* &= \frac{Q_{ji} \tau_{0j,i}}{4k_i \alpha_j} \left\{ \frac{\pi}{4\alpha_j} \left(1 + \frac{4}{5\alpha_i} - \frac{5(2\alpha_i + \alpha_j)}{2\pi \alpha_i \alpha_j} \right) \times \right. \\
 & \times \left. \left(1 + \frac{\chi_{dj}}{\beta_{0j}} \right) - \left(1 + \frac{4}{5\alpha_i} - \frac{\pi(2\alpha_i + \alpha_j)}{4\alpha_i \alpha_j} \right) \frac{\chi_{Lj}}{\beta_{0j}} \right\}, \\
 & \alpha_i \gg 1. \quad (52)
 \end{aligned}$$

In the case of a 2LF composed of thin metal layers ($k_i \ll 1$), the following approximate expressions for the TCR can be obtained:

$$\begin{aligned}
 \beta &\cong \sum_{i \neq j} \frac{\beta_{0i}}{1 + B_{j,i}} \left\{ \left(1 + \frac{2\chi_{di}}{\beta_{0i}} \right) \frac{1}{\ln(1/k_i)} - \frac{2\chi_{di}}{\beta_{0i}} \right\}, \\
 \alpha_i &\leq k_i; \quad (53)
 \end{aligned}$$

$$\begin{aligned}
 \beta &\cong \sum_{i \neq j} \frac{\beta_{0i}}{1 + B_{j,i}} \left\{ \frac{1}{\ln(1/k_i) - (4/\pi)\alpha_i} \left[1 + \frac{2\chi_{di}}{\beta_{0i}} - \right. \right. \\
 & - \left. \left. \left(1 + \frac{\chi_{di}}{\beta_{0i}} + \frac{\chi_{Li}}{\beta_{0i}} \right) \frac{4}{\pi} \alpha_i \right] - \frac{2\chi_{di}}{\beta_{0i}} \right\}, \quad k_i < \alpha_i \ll 1; \quad (54)
 \end{aligned}$$

$$\begin{aligned}
 \beta &\cong - \sum_{i \neq j} \frac{\beta_{0i}}{1 + B_{j,i}} \left\{ \frac{1}{\ln(1/\alpha_i k_i)} \left[\left(1 + \frac{\chi_{di}}{\beta_{0i}} + \frac{\chi_{Li}}{\beta_{0i}} \right) \times \right. \right. \\
 & \times \left. \left. \frac{3}{4\alpha_i^2} + \frac{\chi_{Li}}{\beta_{0i}} - \frac{\chi_{di}}{\beta_{0i}} \right] + \frac{2\chi_{di}}{\beta_{0i}} \right\}, \quad 1 < \alpha_i \ll \frac{1}{k_i}. \quad (55)
 \end{aligned}$$

The dependences of the TCR for a 2L polycrystalline specimen on the layer-thickness ratio $d_{2,1}$ for various values of the parameters characterizing the 2LF, are depicted in Fig. 2. For small $d_{2,1} \ll 1$, the TCR of the 2LF is almost constant, but its numerical value is determined by the interaction of current carriers with the external and internal specimen interfaces. For $d_{2,1}$

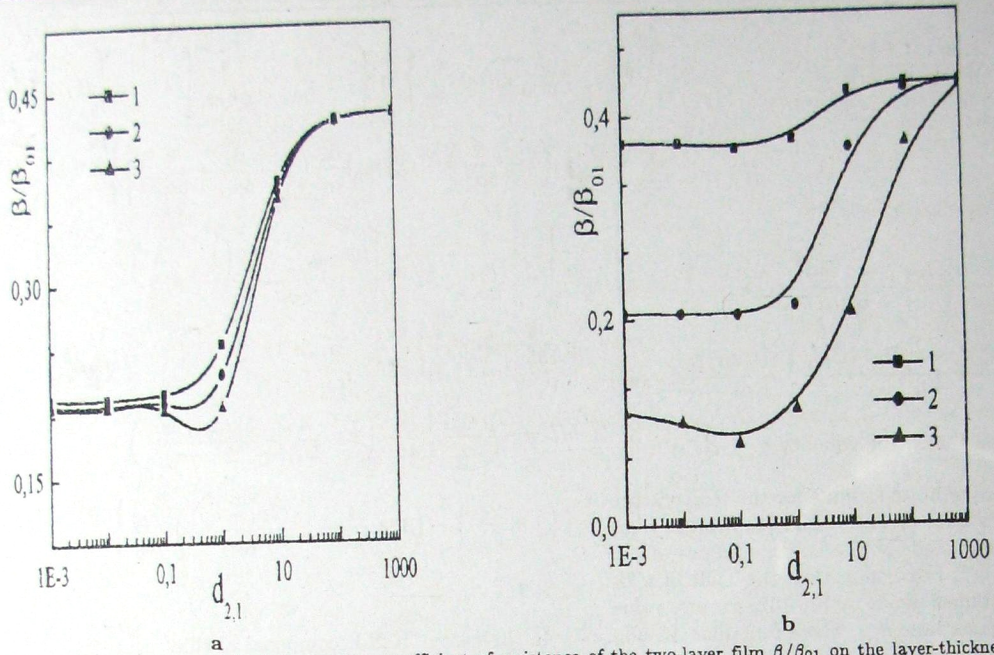


Fig. 2. Dependences of the normalized temperature coefficient of resistance of the two-layer film β/β_{01} on the layer-thickness ratio $d_{2,1}$ in the case $Q_{12} = Q_{21} = Q$ and $P_{12} = P_{21} = P$, and for various parameters: a - $q_1 = q_2 = 0.1$, $P = 0.1$, $\lambda_{1,2} = 1$, $k_1 = 0.1$, $\alpha_1 = \alpha_2 = 1$, $\chi_{d1} = \chi_{d2} = \chi_{L1} = \chi_{L2} = 10^{-4}$, $Q = 0.9$ (1), 0.4 (2), and 0.1 (3); b - $q_1 = q_2 = 0.1$, $P = 0.1$, $Q = 0.1$, $\lambda_{1,2} = 1$, $\alpha_1 = \alpha_2 = 1$, $\chi_{d1} = \chi_{d2} = \chi_{L1} = \chi_{L2} = 10^{-4}$, $k_1 = 1$ (1), 0.1 (2), and 0.01 (3)

increased, so that $d_2 \sim d_1$, the function $\beta(d_{2,1})$ has a minimum of the same origin as the minimum in the 2LF-conductivity dependence (22). While either the probability of electron tunneling into the adjacent layer (Fig. 2,a) or the base layer thickness (Fig. 2,b) increases, this minimum degenerates and the temperature coefficient of resistance becomes monotonous in $d_{2,1}$.

3. Experimental Method

Two-layer films were produced by condensing the metal in a vacuum of 10^{-3} – 10^{-4} Pa on the glass substrate with the temperature $T_{\text{subs}} = 300$ K. After the base layer to condense, it was held at the substrate temperature for 30 min, then the deposition of the upper layer was done. The thermal treatment of the specimens was carried out by triple "heating-cooling" cycles in the temperature interval 300–670 K.

The electrical resistance of the films was measured with an accuracy of $\pm 0.06\%$ by an APPA-109 digital voltmeter (with a rate of two measurements per second). The temperature was monitored making use of a

chromel-alumel thermocouple and a UT-70B voltmeter, which provided an accuracy of ± 1 K. Resistance and temperature data were entered into a computer in a routine of measuring, which made possible to obtain the resistance vs temperature plots. The example of such a dependence for a triple cycle of thermostabilization of the Sc/Cu/subs film system is shown in Fig. 3. The film thicknesses were measured by a MII-4 interferometer, with a Panasonic KXL-600A digital camera and a computer being used for the image registration, which ensured a measurement accuracy of ± 2 nm for layer thicknesses of 50–200 nm.

The electronographic investigations of the phase content of single- and two-layer films evidence for that neither impurity phases based on metals or residual gases, nor intermediate intermetallic phases are formed. The studies of the element content and diffusion processes by mass-spectrometry of secondary ions (an MS-7201 device) showed that, in the case of 2LF systems, mainly the GB diffusion takes place (it is reduced for the systems based on Cu, Cr, Sc, and Co), although the individuality of each layer preserves. All

studied film systems, but Ni/Co/subs and Co/Ni/subs ones, can be therefore regarded as double plates with the eutectic phase content at the LI.

Note that correct selections of the temperature interval and the number of thermostabilizing cycles compose an important methodological issue for the TCR value calculations, since peculiarities in the $R_f(T)$ dependence reveal themselves at the Debye, Néel, and Curie points. Furthermore, those characteristic temperatures change with increase in the thermostabilizing cycle number, which is stipulated by a bulk diffusion of components. Moreover, thermal annealing results in macrostresses of the thermal nature (S_T) leading to the TCR variation. It is also known that the TCR value is connected to the temperature interval ΔT of its measurement.

In this connection, the following conditions for the procedure of the TCR measuring were used:

- the number of thermostabilizing cycles was selected to induce the minimal (for the specimens of the bilayer type) or the maximal (for the film alloy case) mixing of elements;
- the maximal annealing temperature should not result in oxidizing the films;
- the temperature interval of the $R_f(T)$ dependence was selected to exclude the characteristic temperatures;
- the interval ΔT for differentiation (28) was selected according to the conditions (i) that S_T would cause the TCR variation by at most $2 \times 10^{-4} \text{ K}^{-1}$, and (ii) that for the 2-fold increasing of ΔT , the difference of the relevant TCR values, i.e. for ΔT and $2\Delta T$, should not be more than $2 \times 10^{-4} \text{ K}^{-1}$.

In accordance with those criteria, we calculated the TCR for some 2LF systems (see Table).

4. Verification of Relations for TCR

To handle the experimental data for 2LFs on the basis of relation (32) for M_i and M_i^* functions, the simplified expressions (49) and (50) were used, because the investigated films corresponded to the condition $\alpha_i < 1$. Further simplifications of (49) and (50) were made taking into account that χ_d and χ_L are of the order of 10^{-6} K^{-1} , and $\beta \sim 10^{-3} \text{ K}^{-1}$. The values of the TCR $\beta_{\infty i}$ and the specific conductivity $\sigma_{\infty i}$ of the infinitely thick film were used instead of β_{0i} and σ_{0i} , respectively, because the asymptotic values, contrary to the bulk ones, make allowance for the actual structure of film specimens.

Bearing in mind the all above-mentioned, relation (32) can be written in a rather convenient form to handle

experimental results ($\chi_{di} = \chi_{Li} = 0$):

$$\beta = A_1 \beta_{\infty 1} \left\{ 1 - \frac{3}{2} \alpha_1 - \frac{3}{16} \frac{\lambda_1}{d_1} \left[(2 - q_1 - P_{12}) \times \left(1 - \frac{12}{\pi} \alpha_1 \right) + \frac{3}{5\pi} Q_{21} \frac{\lambda_2}{\lambda_1} \alpha_1 \right] + \frac{3}{16} Q_{21} \frac{\lambda_2}{d_1} \frac{\beta_{\infty 2}}{\beta_{\infty 1}} \left[1 - \frac{32}{3\pi} \left(2\alpha_2 + \frac{1}{16} \alpha_1 \right) \right] \right\} + A_2 \beta_{\infty 2} \left\{ 1 - \frac{3}{2} \alpha_2 - \frac{3}{16} \frac{\lambda_2}{d_2} \left[(2 - q_2 - P_{21}) \times \left(1 - \frac{12}{\pi} \alpha_2 \right) + \frac{3}{5\pi} Q_{12} \frac{\lambda_1}{\lambda_2} \alpha_2 \right] + \frac{3}{16} Q_{12} \frac{\lambda_1}{d_2} \frac{\beta_{\infty 1}}{\beta_{\infty 2}} \left[1 - \frac{32}{3\pi} \left(2\alpha_1 + \frac{1}{16} \alpha_2 \right) \right] \right\}, \quad (56)$$

where $A_i = 1/(1 + B_{ji}) = d_i \sigma_{\infty i} \Phi_i / (d_i \sigma_{\infty i} \Phi_i + d_j \sigma_{\infty j} \Phi_j)$.

The electrical transport parameters for individual layers, which enter into (56) and were used in calculations, were determined from the analysis of experimental data for the TCR dependences of single-layer films on the layer thickness. A difficulty consists in determinating the scattering, P_{ij} , and transmission,

Comparison of experimental and theoretical values for TCR

Specimen	Thickness, nm		TCR, 10^{-3} K^{-1}	
	d_1	d_2	expr.	theory
Cu/Cr/subs	15	48	2.22	1.80
Cu/Cr/subs	55	48	2.58	2.37
Cr/Cu/subs	90	180	1.90	1.17
Sc/Cu/subs	48	18	2.10	1.65
Sc/Cu/subs	30	60	1.40	2.80
Sc/Cu/subs	43	65	1.90	1.86
Sc/Cu/subs	38	93	1.69	2.14
Sc/Cr/subs	45	70	0.15	0.89
Co/Cr/subs	55	75	1.56	2.44
Co/Cr/subs	75	75	1.65	2.45
Co/Cr/subs	120	85	1.42	2.29
Cr/Co/subs	85	35	1.62	2.27
Cr/Co/subs	65	80	2.02	2.66
Cr/Co/subs	90	70	2.20	2.22
Ni/Co/subs	20	55	1.20	2.18
Ni/Co/subs	55	80	2.05	1.98
Ni/Co/subs	70	80	2.50	1.79
Co/Ni/subs	30	90	1.19	1.34
Co/Ni/subs	80	75	2.42	1.73
Co/Ni/subs	100	65	1.47	1.93

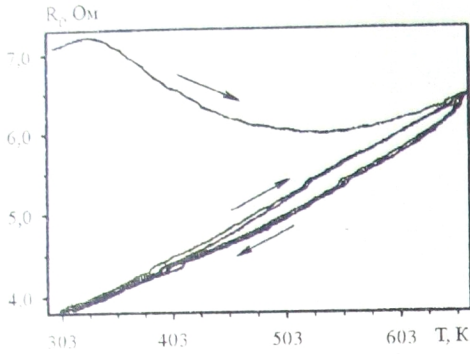


Fig. 3. Dependence of the Sc/Cu/subs film resistance on the annealing temperature. Layer thicknesses: $d_{Sc} = 60$ nm, $d_{Cu} = 30$ nm

Q_{ij} , coefficients of the layer interface, which cannot be evaluated from experimental data. Taking into account that the grain boundaries and the layer interface represent identical scattering centers for electrons, one may use, with a certain error, the GB coefficients of scattering R_i and transmission r_i instead of P_{ij} and Q_{ij} .

On the basis of (56), the TCR vs layer thickness dependences were plotted for 2L films. In Fig. 4, a three-dimensional example of the TCR dependence on the layer thickness for the Sc/Cr film system is shown. The vertical axis corresponds to the TCR, and two horizontal ones to the thicknesses of the base and upper layers. The results obtained imply that, provided the thickness of the base layer is fixed, the TCR value for a 2LF changes monotonously, while d_2 increases. Moreover, if $\beta_{\infty 2} > \beta_{\infty 1}$, the TCR value grows, or diminishes, otherwise. Such a tendency agrees with theoretical dependences $\beta(d_2)$ obtained for $d_1 = \text{const}$ when using the simplified relation of the Dimmich's theory [11] for 2L films based on Co, Cr, and Ni [4].

The degree of consistency between the theoretical and experimental values for the TCR can be judged from the consideration of the table. One of the factors affecting the coincidence of the relevant values is a coefficient of the grain-boundary scattering α_i . Relation (56) was derived for the case $\alpha_i < 1$. In our instance, $\alpha_i = \text{const}$ is supposed, being 0.034, 0.76, 0.59, 0.66, and 0.28 for Cr, Cu, Sc, Co, and Ni single-layer films, respectively. The variable α_i was determined by solving Eq. (15) under the condition $f(\alpha_i) = \beta_{\infty i} / \beta_{0i}$. In real film specimens, the dispersion degree of crystallites increases with decrease in the film thickness, and, as a

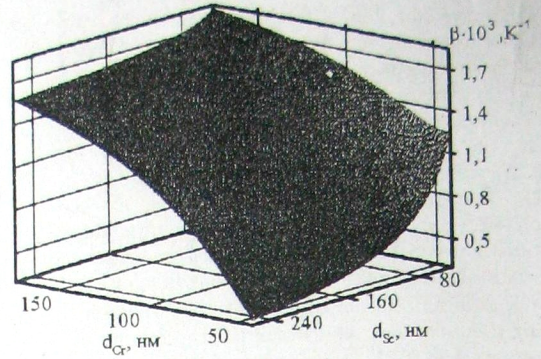


Fig. 4. Three-dimensional diagram for the TCR of the Cr- and Sc-based two-layer films

result, α_i grows. Therefore, the larger the α_i , the worse should be the correspondence between theoretical and experimental TCR data. The fact that the capability of the grain boundaries to scatter charges will change due to diffusion processes occurring along them, has also to be paid attention to. This must affect the calculated TCR value as well, because the above-mentioned model does not make allowance for mutual diffusion processes.

Conclusions

In this work, exact and asymptotic relations for the specific conductivity and TCR in two-layer polycrystalline films, considered in the framework of the Mayadas-Shatzkes theory, were derived. Making use of the electric transport parameters for single-layer films, the calculations of the TCR for two-layer film specimens and its comparison with experimental data were performed. The TCR value was shown to increase or decrease along with the increasing of the upper layer thickness, provided the fixed value for the base layer one, which depends on the ratio between $\beta_{\infty i}$ and $\beta_{\infty j}$ values. The verification of the relations for the TCR in the limiting case $\alpha_i \ll 1$, showed that the proposed model can be used for tentative evaluations of the temperature coefficient of resistance in two-layer films.

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