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T.P. HOVORUN,[1] O.A. BILOUS,[1] O.P. GAPONOVA,[1] L.V. DEKHTYARUK,[2]
A.M. CHORNOUS [1]

[1] Sumy State University
(2, Rymskyi-Korsakov Str., Sumy 40007, Ukraine; e-mail: yushchenko@phe.sumdu.edu.ua)
[2] Kharkiv National University of Construction and Architecture
(40, Sums'ka Str., Kharkiv 61002, Ukraine)

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RESEARCH OF CONDUCTIVITY IN POLYCRYSTALLINE FILMS WITH A THIN COATING USING THE MODIFIED MAYADAS–SHATZKES MODEL

The experimental testing of the theoretical concepts of a modified Mayadas–Shatzkes model for the conductivity in thin polycrystalline films with a thin coating has been carried out. It is shown that the deposition of Ni coatings onto Co films increases the specific conductance in bilayer samples with respect to the conductivity in the basic (uncovered) specimen. At the same time, the coating of the Ni films with Cu and of the Cu films with Ni leads to a decrease of the film conductivity. This phenomenon takes place due to the enhancement or worsening of the grain boundary transparency when the coating is deposited.

Keywords: Mayadas–Shatzkes model, polycrystalline film, coating, diffusion, conductivity.

1. Introduction

Modern requirements to conducting elements in microelectronic and spintronic devices lead to the necessity to create materials with predicted electrophysical properties. The features in the nanocondensate structure are known to dictate, as a rule, conditions for the additional relaxation of charge carriers and give rise to differences between the charge transfer parameters in the film and bulk materials. The electrophysical properties of nanocrystalline metal films are defined, first of all, with the scattering of conduction electrons by the film surface and grain boundaries, which is testified by the results of researches carried out by Ukrainian and foreign scientists [1–5]. Size effects in the conductivity of thin films and condensates were studied in works [3, 4]. In particular, it was shown that the size restriction in films and nanostructures gives rise to the appearance of some physical effects, which are weakly pronounced or not observed at all in massive specimens. As a rule, the surface scattering of charge carriers is described in the framework of the semiclassical approach [6, 7]. In so doing, some electrons are supposed to be reflected specularly and the others diffusively by the film surface, with the coefficient of specular reflection from the surface varying from 0 to 1. The value of this scattering parameter is affected by the state of the film surface.

As a rule, the scattering of charge carriers in multilayer systems or films with a thin coating is associated with the presence of external boundaries and film interfaces. As the layer thickness grows, the specific resistance of the specimen increases and reaches a maximum, because charge carriers penetrate into neighbor film layers. If the interlayer thickness grows further, the maximum degenerates, and the specific resistance changes monotonically, as the specimen thickness increases. The authors of work [8] studied the character of electron scattering by interfaces in epitaxial Cu/(111)Si and Co/(111)Si films with a thin Co or

Cu, respectively, coating layer. As the thickness of the Co coating on Cu increases, the resistance was found firstly to increase. Afterward, when the thickness attains a value corresponding to 1–1.5 monolayer, the resistance decreases.

The authors of work [9] researched the electric resistance of two-layer structures Cu/Ta depending on the thickness of a Cu monolayer, which was varied from 10 to 500 nm. The resistance of the Ta multilayer was found to increase with a reduction of the monolayer thickness. When the monolayer became narrower that 100 nm, the resistance drastically increases and, at a thickness of 50 nm, reached a maximum. The authors of the cited work explained this effect by the influence of the interface scattering of charge carriers. At the same time, they marked that the influence of the grain-boundary scattering on the specific resistance became significant for the monolayer thickness varying from one nanometer to submicron sizes.

In work [10], conditions of electron transport in two-layer film systems consisting of a thin palladium film deposited on a germanium film were studied. It was shown that the germanium sublayer with a subatomic thickness deposited beforehand on a dielectric substrate accelerates the process of palladium film metallization. At the same time, according to the cited authors, the atoms in as-deposited quench-condensed films can be regarded almost motionless at the substrate surface, so that the diffusion of germanium atoms into the metal film bulk can be excluded from consideration. The germanium suiblayer was shown to reduce the percolation thickness threshold and to promote the 2D growth regime of ultrathin palladium films. The size dependences of the specific resistance in palladium films were described with the help of the Wissman and Namba theories for the charge transfer in spatially confined systems. It was found that those dependences for ultrathin palladium films with the thicknesses $d < 3.5$ nm cannot be described in the framework of examined models.

The other works of researchers belonging to the Lviv scientific school [11–13] concern the influence of surfactant germanium sublayers on the conductivity in metal films of copper, gold, silver, and lead. The effect takes place, because the deposition of a thin layer onto the lower film surface results in the appearance of a film structure, which governs the electrophysical properties of the film and affects the parameters of the film surface reflectivity near the substrate.

Nevertheless, despite that there are publications on this topic, the issue concerning the satisfactory agreement between the theoretical relations obtained for a model film and the results of experimental researches on how a thin coating on the film surface affects the electrophysical properties of the film and the parameters of charge transfer in it remains challenging.

In work [14], the conductivity in thin films with single- and polycrystalline structures and with one of their surfaces being covered with an ultrathin diffusing layer of a different metal was analyzed theoretically. Exact and asymptotic expressions for the film specific conductance were derived, and a detailed numerical analysis of its dependence on the diffusion annealing time was carried out for various values of the parameters characterizing the specimen structure. The authors of work [14] drew conclusion that the presence of an ultrathin diffusing layer of impurities on one of the surfaces of a thin single- or polycrystalline film substantially affects its specific conductance. The change in the conducting properties of the single- or polycrystalline metal layer after its diffusion annealing allows the very processes of bulk and grain-boundary diffusion to be studied, the effective penetration depth of diffusant atoms into the specimen bulk and along the grain boundaries to be determined, and the coefficients of bulk and grain-boundary diffusion to be evaluated. In the framework of this model, the grain-boundary and surface scatterings in metal films were studied; in particular, in the films that are used for the fabrication of spin-valve structures.

Proceeding from the model for the film conductivity, which was proposed theoretically and analyzed by the authors of work [14], we have studied experimentally the influence of a thin metal coating deposited onto the film surface on the conductivity in film specimens. We also verified the asymptotic relations for the film conductivity, which were derived in the framework of this model [15], by using Cu, Ni, and Co films as examples.

2. Asymptotic Relations for Coated Films in the Framework of the Modified Mayadas–Shatzkes Model

The Mayadas–Shatzkes model describes the conductivity in polycrystalline metal films making allowance for surface and grain-boundary scatterings [15]. In

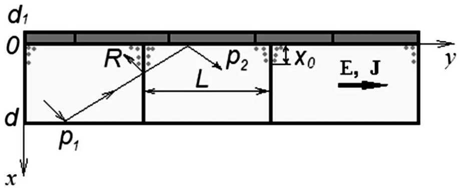


Fig. 1. Schematic diagram of a thin polycrystalline film with grain-boundary diffusion of atoms. The broken line schematically illustrates one of the electron trajectories [14]: x_0 is the penetration depth of coating atoms in the course of their diffusion, $\mathbf{E}$ the external electric field strength, and J the electric current density

the framework of this model, the authors of work [14] derived exact and asymptotic relations for the conductivity in a polycrystalline film, one of its surfaces being covered with a thin layer of a different diffusant material. The cited authors took into account that the coating is much narrower than the basic film, so that the contribution made by this coating layer to the total conductivity of the film system is insignificant and can be neglected. Let us briefly consider the main theoretical grounds of work [14].

Figure 1 illustrates the model of a thin film with thickness d. One of its surfaces is covered with a thin coating of a different metal with thickness d_1. If the diffusion annealing temperature T_0 of the polycrystalline film is relatively low, namely, if $T_0 < 0.3T_m$, there is almost no bulk diffusion in the polycrystalline specimen. Then, impurity atoms mainly migrate only along crystallite interfaces, which results in a change of the character of charge carrier scattering by grain boundaries and, accordingly, to a modification of the conductivity in the coated film. In other words, the resistance of the basic film has to change owing to the appearance of foreign atoms at the grain boundaries [16].

Impurity atoms diffuse along the grain boundaries. The normal to the film surface is parallel to the axis Ox (see Fig. 1), which is directed downward, and the coordinate x is reckoned, as a rule, from the upper boundary of the basic layer. The axis Oy is directed along the surface. The external electric field strength $\mathbf{E} = (0, E, 0)$ is also directed along the film surface.

The influence of the low-temperature grain-boundary diffusion on the specific conductance in a thin polycrystalline film (Fig. 1) is evaluated, by taking advantage of theoretical relations of the Mayadas–Shatzkes model. In the framework of this model, the grain-boundary scattering is described with the help of the coefficient of electron scattering by crystallite interfaces, R, and the parameter of grain-boundary scattering α, which is determined by the formula

$$\alpha = \frac{\lambda}{L}\,\frac{R}{1-R}, \tag{1}$$

where L is the average grain size, R the coefficient of electron scattering by grain boundaries in the absence of impurity atoms, and λ the mean free path of charge carriers.

The presence of a thin coating on the film surface gives rise to a modification of the grain-boundary scattering characteristics. Taking the diffusion into account, the latter acquire the values R' and α', respectively. At a low concentration of atoms diffusing along the grain boundaries, the magnitude of R' is determined by the formula [16]

$$R' = R + \gamma_g C_g, \tag{2}$$

where R is the coefficient of charge carrier scattering by the grain boundary in the absence of diffusing atoms; γ_g a coefficient determining the character of a grain boundary (namely, the impurity atoms make the boundary more transparent, if $\gamma_g < 0$, and more scattering, if $\gamma_g > 0$), $C_g = C_0 \exp\{-\mu x_0\}$ is the diffusant distribution function for the grains, x_0 the penetration depth of atoms, C_0 the initial surface concentration of diffusing elements,

$$\mu = \left\{ \frac{2}{\delta D_g} \left(\frac{D_V}{\pi\, \tau_D} \right)^{1/2} \right\}^{1/2}$$

is a characteristic penetration depth of impurity atoms into the grain boundaries, D_V the coefficient of bulk diffusion, D_g the coefficient of grain-boundary diffusion, δ the diffusion width of the grain boundary, and τ_D the diffusion annealing time. At the same time, the expression for the parameter of grain-boundary scattering takes the form

$$\alpha' = \alpha \frac{1 + (\gamma_g/R)\, C_g}{1 - (\gamma_g/(1-R))\, C_g}. \tag{3}$$

According to the results of work [15], the expression for the specific conductance in the film specimens that satisfy the condition $d \gg \lambda$ can be presented in the following form:

for films with the large-grain structure ($\alpha \ll 1$),

$$\frac{\sigma}{\sigma_\infty} \cong 1 - \frac{3}{2}\alpha \times$$
$$\times \left[1 + \frac{1}{\mu d R} \ln\left(1 + \gamma_g C_g \frac{1 - \exp(-\mu d)}{1 - (R + \gamma_g C_g)}\right)\right]; \qquad (4a)$$

and for films with the fine-grain structure ($\alpha \gg 1$),

$$\frac{\sigma}{\sigma_\infty} \cong \frac{3}{4\alpha} \times$$
$$\times \left[1 + \frac{1}{\mu d (1 - R)} \ln\left(1 - \gamma_g C_g \frac{1 - \exp(-\mu d)}{R + \gamma_g C_g}\right)\right]; (4b)$$

where σ_∞ is the conductivity of a polycrystalline film specimen with the infinite thickness. In the case $\alpha \ll 1$, formula (4a) can be simplified to the following asymptotic limit for the specific conductance of a polycrystalline film with a thin coating:

$$\frac{\sigma}{\sigma_\infty} = 1 - \frac{3}{2}\alpha' - \frac{3\lambda (2 - p_1 - p_2)}{16d} \left(1 - \frac{32}{3\pi}\alpha'\right), \qquad (5)$$

where p_1 and p_2 are the coefficients of specular reflection from two opposite external film surfaces: the substrate/film and film/coating, respectively, interfaces; and d is the film thickness.

By equating the parameters of specular reflection from the opposite external surfaces: $p_1 = p_2 = p$, where p is the coefficient of specular reflection for the coated film, expression (5) can be presented in the form

$$\frac{\sigma}{\sigma_\infty} = 1 - \frac{3}{2}\alpha' - \frac{3\lambda (1 - p)}{8d} \left(1 - \frac{32}{3\pi}\alpha'\right). \qquad (6)$$

3. Experimental Method and Procedure

In order to verify theoretical principles of the asymptotic model proposed in work [14], experimental specimens were fabricated, by using the following technique. Metal films were condensed on polished polycore substrates containing preliminarily deposited contact platforms with corresponding geometrical configurations Cu(150–200 nm)/Cr(30–50 nm)/substrate. Metals were deposited by the method of thermal (Cu, Ni) or electron-beam (Co) evaporation.

The thermostabilization of electric properties of the basic film was carried out, by applying two to three "heating-cooling" cycles in a temperature interval of 300–550 K (Cu), 300–630 K (Ni), or 300–650 K (Co). At the next experimental stages, thin metal coatings with the thickness $d \leq 1 \div 2$ nm were deposited onto the basic films, and the specimen was subjected to the thermostabilizing annealing.

The structural characteristics and the phase composition of the films were studied by the methods of transmission electron microscopy and electronography (Fig. 2). The conditions of thermal treatment were identical for the specimens intended either for the study of their electrophysical properties or for electron microscopy researches.

The results of our researches (Fig. 2) show that the phase composition of film specimens correspond to that of massive specimens. For Cu films, the average grain size L increases with the film thickness d and amounted to $L \approx 40 \div 110$ nm in the interval $d = 20 \div 70$ nm. Ni films had the average grain size $L \approx 30 \div 60$ in the film thickness interval $d = 30 \div 60$ nm. For Co films with the thickness $d = 20 \div 70$ nm, the average grain size amounted to $L \approx 17 \div 20$ nm.

4. Results and Their Discussion

Relations (5) and (6) were used to calculate the specific conductance in the metal film specimens. Those relations include the parameters of charge transfer, such as the parameter of specular reflection p, the mean free path of electrons λ, and the parameter of grain-boundary scattering α, which depends on the coefficient of electron scattering by grain boundaries, R. When calculating the specific conductance on the basis of formulas (5) and (6), we used the parameters obtained earlier in our works [17–19], when analyzing the experimental dependences of the temperature coefficient of film resistance on the film thickness measured for Cu, Ni, and Co films. Besides that, the indicated parameters were calculated for the films of those metals with a coating making use of a linearized model and the Tellier–Tosser–Pichard (TTP) model of isotropic scattering [20]. The application of the linearized TTP model allowed the magnitude of quantity $\lambda(1 - p)$ to be determined.

In works [21, 22] and some others, their authors used the diffusion approximation and took the mean free path determined from the linearized model as a parameter. In this work, we followed this approach. The mean free path values were obtained in the framework of the linearized model and in the diffusion approximation for the scattering at external

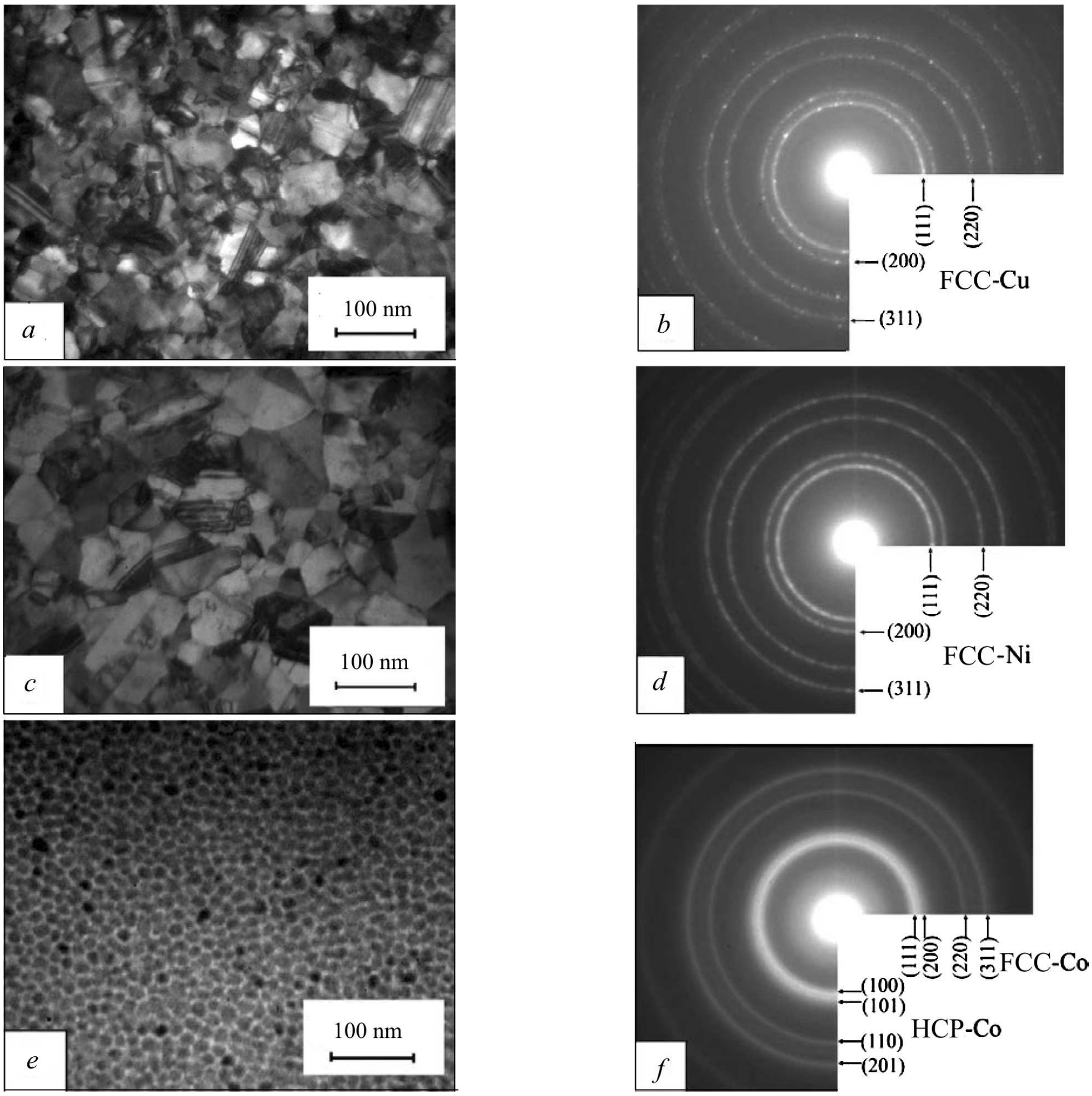


Fig. 2. Crystal structures and corresponding electron diffraction patterns for Cu ($d = 42$ nm) (a and b), Ni ($d = 52$ nm) (c and d), and Co ($d = 50$ nm) (e and f) films after their thermal treatment

surfaces. Then they were used, when determining the coefficients p, R, and r in the framework of the TTP isotropic scattering model. Note that the calculations were carried out, by using the data obtained for temperatures of 360 (Cu) and 300 K (Ni and Co). Those data and the data on the average grain size that were used in calculations are quoted in Table 1.

Note also that, while calculating the parameters of charge transfer in coated films, we supposed that the deposition of a thin coating layer does not change the mean free path, but only the scattering conditions at the external and internal film boundaries. This approach was applied, for example, in work [23]. We would like to point out that, in principle, a variation of the scattering coefficients should affect the total free path length of charge carriers, but the corresponding change is smaller than the error of the mean free path determination.

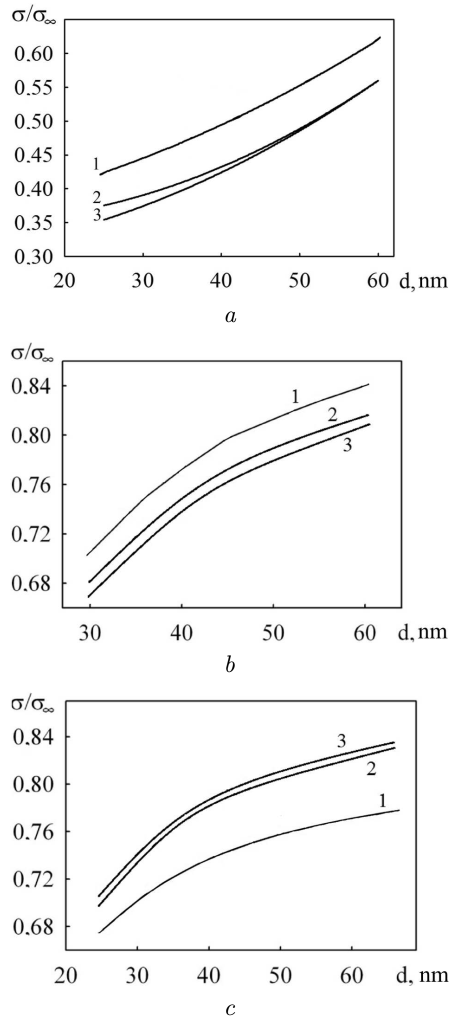
The results of calculations are illustrated in Fig. 3. Here, the specific conductances for basic Cu, Ni, and Co films with various thicknesses, as well as for the films with coatings, calculated by formulas

(5) and (6) are shown. While comparing the results obtained for the Cu/substrate and Ni/Cu/substrate films (Fig. 3, *a*), as well as the Ni/substrate and Cu/Ni/substrate ones (Fig.3, *b*), the conclusion can be drawn that the deposition of a thin coating leads to a reduction in the specific conductance of the basic film. This reduction is associated with the diffusion of coating atoms along the grain boundaries, which changes the coefficients of electron scattering by and electron transmission through the grain boundaries in the Ni and Cu films with the Cu and Ni coatings, respectively, so that the transparency of grain boundaries worsens.

By comparing the calculated dependences depicted in Fig. 3, *a* with the experimental data for the Ni/Cu/substrate films, we may assert about their agreement. For example, the calculated specific resistances ρ (this is a value reciprocal to the specific conductance) for the specimens 36 and 60 nm in thickness amount to 3.2×10^{-8} and 2.6×10^{-8} Ω m, respectively, whereas the corresponding experimental data are equal to 3.3×10^{-8} and 2.6×10^{-8} Ω m, which corresponds to the literature data, e.g., of works [22, 24, 25]. For the Cu/Ni/substrate film system, the experimental values of ρ obtained, e.g., for thicknesses of 30 and 60 nm amount to 19.4×10^{-8} and 17.6×10^{-8} Ω m, respectively, and the calculated values equal 19.6×10^{-8} and 16.3×10^{-8} Ω m, respectively.

In the case of nanocrystalline Co basic films, the size dependences of the specific conductance for the Co films with and without the Ni coating calculated by relations (5) and (6) demonstrate that the deposition of a thin Ni coating results in an increase of their specific conductance (Fig. 3, *c*). In other words, the diffusion of nickel coating atoms along the grain boundaries in the cobalt film results in the enhancement of the grain boundary transparency. By comparing the theoretical and experimental values of the specific resistance for the cobalt films with the nickel coating, we may say about their quantitative agreement. For instance, the specific resistance amounts to 36.3×10^{-8} and 23.9×10^{-8} Ω m for film thicknesses of 32 and 50 nm, respectively. Our earlier experimental results obtained for the same thicknesses equaled 37.7×10^{-8} and 24.8×10^{-8} Ω m, respectively.

The maximum discrepancy between the calculated results and the experimental data obtained for the



a

b

c

Fig. 3. Calculated dependences of the specific conductance on the film thickness for Ni/Cu/substrate and Cu/substrate films (*a*); Cu/Ni/substrate and Ni/substrate films (*b*); and Ni/Co/substrate and Co/substrate films (*c*): (*1*) films without the coating; (*2*) films with the coating, relation (5); and (*3*) films with the coating, relation (6)

Parameters of charge transfer in Cu, Ni, and Co films without and with the coating

Substrate	*d*, nm	*L*, nm	$\lambda(1-p)$, nm	*p*	*R*	α
Cu/substrate	25–60	50	22.4	0.25	0.46	0.424
Ni/Cu/substrate				0.10	0.51	0.489
Ni/substrate	30–60	30–60	28.7	0.19	0.01–0.03	0.03
Cu/Ni/substrate				0.10	0.03–0.06	0.11
Co/substrate	25–60	20	42.2	0.11	0.12	0.107
Ni/Co/substrate				0.06	0.14	0.06

Ni/Cu/substrate, Cu/Ni/substrate, and Ni/Co/substrate films amounted to 8%.

The analysis of the plotted dependences testifies to an insignificant discrepancy of the results obtained by relations (5) and (6), when calculating the specific conductance in specimens with a thin metal coating. This fact is most likely associated with the substitution of the factor $(2 - p_1 - p_2)$ in relation (5) by $(1 - p)$, when changing to expression (6). In the case of Eq. (5), the value of p_1 was selected to be the coefficient of specular reflection from the film surface without the coating, which was obtained in the framework of the isotropic TTP model [17], and p_2 was the parameter of specular reflection from the film surface with the coating. In relation (6), the parameter p is the coefficient of specular reflection from the film surface with the coating. Evidently, the former approach is more accurate, although it is practically impossible to resolve the contribution of scattering at each film surface into the magnitude of the parameter p, when treating the experimental results of works [17–19] in the framework of the isotropic TTP model.

5. Conclusions

In this work, the experimental verification of the asymptotic ($\alpha \ll 1$ and $k \gg 1$) relation derived in the framework of the theoretical model for the specific conductance of a polycrystalline metal film with a thin coating developed on the basis of the Mayadas–Shatzkes theory was carried out for the first time. The obtained calculation results concerning the influence of impurity (coating) atoms on the specific conductance in the basic Cu, Ni, and Co film specimens made it possible to draw the following conclusions. In the Ni/Cu/substrate and Cu/Ni/substrate film systems, the diffusion of coating atoms gives rise to a reduction of the specific conductance, i.e. the grain boundaries of the basic Cu and Ni films become less transparent to charge carriers. At the same time, in the Ni/Co/substrate films, the nickel coating enhances the transparency of grain boundaries, and the specific conductance of the cobalt films becomes higher.

With the growth of their thickness, all researched film specimens demonstrated the increase of their specific conductance. At the same time, the presence of a coating on Co films increases the conductivity of two-layer specimens with respect to the conductivity of the basic specimen, whereas the coating on the Ni- and Cu-based specimens results in a reduction of their specific conductance.

The maximum difference between the theoretical and experimental results obtained for the Ni/Cu/substrate, Cu/Ni/substrate, and Ni/Co/substrate films amounts to 8%.

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Т.П. Говорун, О.А. Білоус,
О.П. Гапонова, Л.В. Дехтярук, А.М. Чорноус

ДОСЛІДЖЕННЯ ПРОВІДНОСТІ ПОЛІКРИСТАЛІЧНИХ ПЛІВОК З ТОНКИМ ПОКРИТТЯМ ІЗ ВИКОРИСТАННЯМ МОДИФІКОВАНОЇ МОДЕЛІ МАЯДАСА–ШАТЦКЕСА

Резюме

В роботі проведено експериментальну апробацію теоретичних положень асимптотичного співвідношення, які запропоновані із використанням модифікованої моделі Маядаса–Шатцкеса для провідності полікристалічних плівок з тонким покриттям. Показано, що нанесення покриття із Ni на плівки Co збільшує провідність двошарових зразків по відношенню до провідності базового зразка, в той час, як покриття із Cu і Ni на плівках Ni та Cu відповідно призводить до зменшення питомої провідності плівок. Це пов'язано із покращенням чи погіршенням прозорості меж зерен при нанесенні покриття.