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I. Protsenko, L. Odnodvoretz, A. Chornous

Sumy State University, Physical-Engineering Department
2 Rimsky-Korsakov St., UA-244007 Sumy, Ukraine

ELECTROCONDUCTIVITY AND TENSOSENSIBILITY OF MULTILAYER FILMS

The experimental-research results for a size effect on the thermal coefficient of resistance (TCR) and coefficient of longitudinal-strain sensitivity (CS) of two-layer and multilayer structures based on Cr, Co, Ni, and Mo are discussed. The results obtained are in satisfactory agreement with microscopic and macroscopic theoretical models. The diagrams in the co-ordinates 'TCR—thickness' and 'CS—thickness' are plotted.

Наведено результати експериментальних досліджень розмірного ефекту термічного коефіцієнта опору (ТКО) та коефіцієнта тензочутливості (КТ) двох- та багатошарових плівкових структур на основі Cr, Co, Ni та Mo, обробку яких проведено в рамках мікроскопічної та макроскопічної моделей. Побудовано діаграми в координатах «ТКО—товщина» та «КТ—товщина».

Приведены результаты экспериментальных исследований размерного эффекта термического коэффициента сопротивления и коэффициента тензочувствительности двух- и многослойных пленочных структур на основе Cr, Co, Ni и Mo, обработка которых была проведена в рамках микро- и макроскопической моделей. Построены диаграммы в координатах «ТКС—толщина» и «КТ—толщина».

Key words: multilayer film's structure, electrophysical properties, thermal coefficient of resistance, coefficients of strain sensitivity.

1. Introduction

Recently a considerable attention has been focused on the research of electrophysical properties of general-type multilayer-film structures (*i.e.* $d_n/.../d_2/d_1/S$, where S — is a substrate, $d_1 \neq d_2 \neq \neq ... \neq d_n$) or periodic ones, in which the films' structure of general type acts as a fragment (that is $d_n/.../d_2/d_1/d_n/.../d_2/d_1/S$).

Such multilayer structures have their special features in reference to size phenomena in view of the fact that along with the traditional mechanism of electric-current carriers scattering on phonons, defects of a crystal lattice, grain boundaries, and external surfaces, there occurs an additional one which is connected with interfaces of separate layers.

With reference to technical applications of the multilayer film systems, they have a number of potential advantages in comparison with one-layer systems.

The results of investigation of the size effect on electrophysical properties (the thermal coefficient of resistance (TCR) and the coefficient of strain sensitivity (CS)) in multilayer film structures on the basis of Cr, Co, Ni, and Mo films are presented in this paper. A succession of the layers was such that two ferromagnetic films (Co, Ni) with or without a separating paramagnetic Cr film represented a fragment of a periodic structure.

The submitted results permit to the understanding of size phenomena as well as to a generalized understanding for film systems prepared on the basis of other metals.

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2. Theoretical Models of Size Effects in Electroconductivity and Strain Sensitivity of Multilayer Metal Films

2.1. Double-layer films

A question about the size effect on the electroconductivity (σ) and the thermal coefficient of resistance (β) of two-layer metal films has been repeatedly discussed in literature [1—5]. Thus, in Refs. [3, 5] a subject of research was single crystal films; Refs. [3, 6] considered basically polycrystalline samples, and Ref. [5] was dedicated to the conditions of an atomic interdiffusion under arbitrary ratio between the thickness of layers, d_1 and d_2, and the mean free path, λ_{01} and λ_{02}.

In these works, a model of a double-layer film of thickness $d = d_1 + d_2$ (an axis Oz is parallel to the normal of its surface) is considered, in which the interdiffusion of atoms takes place or does not.

To determine the specific conductivity of such a film, it is necessary to solve the Boltzmann kinetic equation for the nonequilibrium additive to the Fermi—Dirac distribution function $f_0(\varepsilon_i)$. Authors of [3—6] have obtained the expressions for σ_f, having an identical mathematical structure, but have written them in different ways. In a general form it can be presented as:

$$\sigma = \sum_{i=1}^{2}\{\sigma_{gi} - (3\sigma_{oi}\lambda_{0i}/d_i)\int d\varphi \int dt(t^{-3} - t^{-5}) \times \\ \times \frac{\cos^2\varphi}{H_i^2(t, \varphi)}(F_i + G_i)\}, \quad (1)$$

where σ_g and σ_o are the specific conductivity of a one-layer film, limited by grain boundaries (*i.e.* $d \to \infty$) and that of a massive crystal; $(F + G)$ is a complex function of single-crystal parameters $P^* = (1 - W - P)$ and $Q^* = (W - Q)$, which characterize the probability of reflection and tunneling of an electron with a tangential component being kept with respect to the interface of the layers of the quasi-impulse component (W, P and Q are the probabilities of a tunneling electron through the interface, the diffusion scattering without transition and with transition into the next layer).

For short times of the diffusive annealing t_D, when the characteristic length of the decreased concentration of atoms on other sort is shorter than the characteristic scale of the change of the electron distribution function, the size effect in double-layer systems determines the width of a pure region. If the characteristic depth of the penetration of atoms of another sort has a value of the order of the thickness d_i, but $\lambda_{0i} > d_i$, the size effect is takes place.

In this respect, the expression of the double-layer film, obtained in Ref. [7], opens wide possibilities for comparison with the experimental results:

$$\beta = \sum_{i=1}^{2} A_i\beta_{oi}\left(1 - \frac{d\ln F_i}{d\ln k_i} - \frac{d\ln F_i}{d\ln L_i}\right), \quad (2)$$

where $A_i = d\rho_{oi}F_i/\Sigma d\rho_{oi}F_i$, $F_i = \sigma_i/\sigma_{0i}$ — the Fusch function; $k_i = d_i/\lambda_{0i}$ and $L_i = D_i/\lambda_{0i}$ are the reduced thickness and reduced average size of crystallites, β_{0i} is TCR for massive polycrystalline samples.

In limiting case of large thicknesses

$$\beta = \sum_{i=1}^{2} A_i\beta_{oi} \quad . \quad (3)$$

A comparison with experimental results indicated, that it was more correct to take the TCR value β_{gi} at $d_i \to \infty$ instead of β_{0i}.

Recent decade a considerable attention was directed to investigations of the strain sensitivity effect in double-layer films. General physical considerations indicate that the strain sensitivity of the multilayer seems to be of importance in comparison with the one-layer film of the same thickness, which is connected with an additional electron scattering at the interface of the layers. In Ref. [8] the theory of the strain-sensitivity effect was offered for the double-layer single-crystal films of metals. The basis of the theory is the representation of the parallel connection of two films.

After differentiation of the logarithm of the film resistance R on the longitudinal ($d\varepsilon_l = dl/l$) and transverse deformation flow ($d\varepsilon_t = da/a$), Khater and El-Hiti [8] have obtained an expression for coefficients of longitudinal ($\gamma_l = R^{-1}dR/d\varepsilon_l$) and transverse ($\gamma_t = R^{-1}dR/d\varepsilon_t$) strain sensitivities. In view of their identical mathematical structure, we shall present the expression only for γ_l:

$$\frac{dR/R}{dl/l} = -A_1\left(\frac{dd_1/d_1}{dl/l} + \frac{d\sigma_{01}/\sigma_{01}}{dl/l} + \frac{dF_1/F_1}{dl/l}\right) -$$

$$- A_2\left(\frac{dd_2/d_2}{dl/l} + \frac{d\sigma_{o2}/\sigma_{o2}}{dl/l} + \frac{dF_2/F_2}{dl/l}\right) - \frac{da/a}{dl/l} + 1, \quad (4)$$

which, under assumption that $\gamma_{01l} = 1 + \eta_{01l} = \gamma_{1l}$, has been transformed by rearranging as in Ref. [9]:

$$\gamma_l = A_1[\gamma_{1l} + (\beta_f/\beta_{01} - 1)\left(\eta_{2l} - \mu_2\frac{1-\mu_S}{1-\mu_2}\right)\beta_{01}/\beta_{02}] +$$

$$+ A_2[\gamma_{2l} + (\beta_f/\beta_{02} - 1) \times$$

$$\times \left(\eta_{1l} - \mu_1\frac{1-\mu_S}{1-\mu_1}\right)\beta_{02}/\beta_{01}], \quad (4a)$$

where $\eta_l = -\frac{1 d\lambda_0}{\lambda_0 d\varepsilon_l}$ is the deformation coefficient of the mean free part, μ_i and μ_S are the Poisson coefficients of the one-layer film and the substrate.

In Ref. [10] an attempt to modify the Eq. (4a) has been made so that it was suitable for polycrystalline films. The allowance for the grain-interface scattering was realized only by means of the multipliers F_1 and F_2, an explicit view of which was written within the framework of the model of the effective length of the mean free part.

Ultimately the expression, which (see section 4.1) corresponds to the experimental results in the best way, has been written as follows

$$\gamma_l = A_1[(\gamma_{g1l} + \mu_{f1}) + (1 - \beta/\beta_{g1})(2(1 - \gamma_{g1l}) + \mu_{f1} +$$

$$+ \mu_{f2}(\beta_{g1}/\beta_{g2}))] + A_2[(\gamma_{g2l} + \mu_{f2}) + (1 - \beta/\beta_{g2}) \times$$

$$\times (2(1 - \gamma_{g2l}) + \mu_{f2} + \mu_{f1}(\beta_{g2}/\beta_{g1}))] + 1 + 2\mu_S. \quad (5)$$

2.2. Multilayer films

It is known that electrophysical properties of double-layer films are sufficiently investigated both in theoretical and in the experimental aspects, which can hardly be said to periodical multilayer structures or the general forms because the achievements of this field are not so significant. So, Ref. [2], which was devoted to the theoretical consideration of electrical properties of the periodical multilayer structures with the fragment (period) $d_f = d_1 + d_2$, was reduced totally to the analysis of properties of a separate fragment, which represents the same double-layer film. The simplified expressions (2) and (3) of the Dimmich theory can be generalized for the case of the arbitrary number of layers [11].

However, there are great difficulties of the mathematical character, making impossible to obtain rigorous relations of type (2), (3), (4) within the framework of the microscopic models. It has not been realized now, on the other hand the analysis of physical processes, as well as a forecast of electrophysical properties can be realized also in the framework of the macroscopic approach.

According to Ref. [1] in terms of the representation of the parallel connection of the sample layers the expressions for TCR and CS for the film with arbitrary (n) number of layers have been obtained. The derivation of the above mentioned expressions is based on taking the logarithm and the subsequent temperature differentiation or the deformation of the following expression for the resistivity (ρ):

$$\rho = \frac{\rho_1\rho_2...\rho_n(d_1 + d_2 + ... + d_n)}{d_1\rho_1\rho_2...\rho_n + d_2\rho_1\rho_3...\rho_n + ... + d_n\rho_1\rho_2...\rho_{n-1}}. \quad (6)$$

In the case of TCR we obtain

$$\beta = \frac{1}{\rho}\frac{\partial\rho}{\partial T} = \sum\beta_i + \frac{\sum d_i\rho_i}{\sum d_i} - \frac{\sum d_i\rho_i\rho_k\rho_m...}{\sum d_i\rho_k\rho_m...} -$$

$$- \frac{\sum d_i(\beta_k + \beta_m + ... + \beta_n)\rho_k\rho_m...}{\sum d_i\rho_k\rho_m...} \quad (i \neq k \neq m ...). \quad (7)$$

For the coefficient of the longitudinal strain sensitivity it is possible to write down a similar expression

$$\gamma_l = \frac{d\ln R}{d\ln l} = \frac{d\ln\rho}{d\ln l} + 1 + 2\mu_f =$$

$$= \sum\gamma_{li} - \frac{\sum d_i(\gamma_{lk} + \gamma_{lm} + ... - \mu_{fi})\rho_k\rho_m}{(1/2)\sum d_i\rho_k\rho_m...} -$$

$$- \frac{\sum\mu_{fi}d_i}{\sum d_i} + 1 + 2\mu_f \quad (i \neq k \neq m \neq ...). \quad (8)$$

In the limiting cases $d_i \to \infty$ and $d_i \to 0$, it follows from (8) that

$$\lim_{d_i\to\infty}\gamma_l = \lim_{d_i\to\infty}\gamma_{l1} + ... - \lim_{d_i\to\infty}\gamma_{l1} - ... = \gamma_{gi},$$

$$\lim_{d_i\to 0}\gamma_l = \gamma_l^{(i-1)}. \quad (9)$$

Here $\gamma_l^{(i-1)}$ is the coefficient of strain sensitivity of

Table 1. **Experimental and calculated data on the TCR (300 K)**

Films (thickness, nm)	$\beta_{f\,exper}\cdot 10^3$, K^{-1}	$\beta_{f\,calc}\cdot 10^3$, K^{-1}
Cr(40)/Co(40)/S	0.90	1.70
Cr(80)/Co(65)/S	2.02	2.20
Co(25)/Cr(45)/S	0.80	1.72
Co(75)/Cr(55)/S	1.56	2.60
Co(25)/Ni(20)/S	2.00	2.95
a^* — Mo(90)/Cr(180)/S	-2.00	-2.50

*a is an amorphous phase.

multilayer films with the number of layers (i-1). The expressions (7)—(9) will be used in section 4.2 for plotting the size diagrams β—d and γ_l—d and for forecasting the electrophysical properties of periodic and general type multilayer structures.

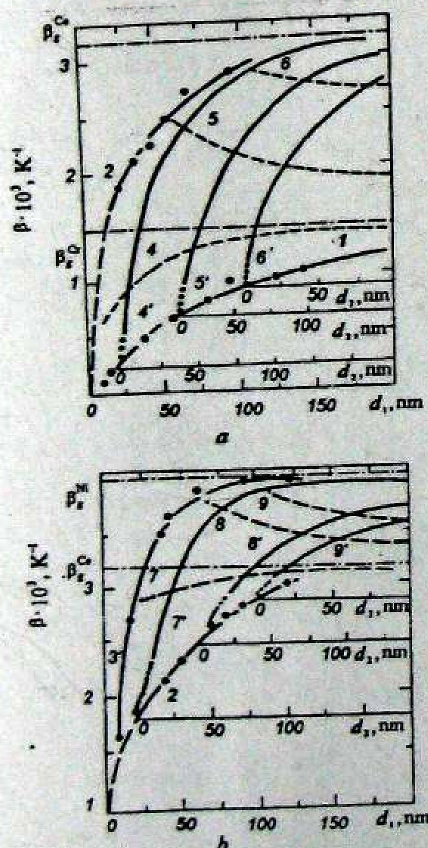

Fig. 1. The dependence of β on the thickness for double-layer films of Cr/Co and Co/Cr (a) and Ni/Co and Co/Ni and Co/Ni (b): 1—3 are the experimental of dependences β (d_1) for films of Cr, Co and Ni; 4—6, 4'—6', 7—9, 7'—9' are the calculation dependences β (d_2) for Cr/Co, Co/Cr, Co/Ni and Ni/Co, respectively

3. Experiment

Condensation of multilayer film structures (up to 10 layers) on to carbon (for the electron-microscopic researches), glass (for measurements of a specific resistance and TCR) and textolite substrates (for CS measurement) was realized using a standard vacuum apparatus ВУП-5М (~ 10^{-3} Pa) at the substrate temperature T_S = 300 K. The annealing of samples was performed in vacuum within the temperature interval from 300 to 650 K (when researching the crystal structure, resistance, and TCR) or 300 to 500 K (when researching CS) according to a scheme 'heating—cooling'.

The technique of the measurement of the longitudinal and transversal coefficient of the strain sensitivity consists in determination of the relative resistance change $\Delta R/R_н$ under bending deformation of a film on a substrate ($\Delta\varepsilon$ = 0—0.06%) or under stretching one ($\Delta\varepsilon$ = 0—4%) with the aid of a microscrew [12, 13].

Processes of the interdiffusion were investigated using the method of ion-secondary mass-spectrometry (MC 7201 M) with argon for producing primary ions.

4. Results

We have accumulated a significant experimental material on double-layer metal films on chromium, cobalt, nickel and molybdenum basis.

Table 2. **The comparison of the experimental and calculated data for the coefficient of the longitudinal strain sensitivity**

Films (thickness, nm)	$\gamma_{l\ exper.}$	$\gamma_{l\ calc.}$ by (5)	Films (thickness, nm)	$\gamma_{l\ exper.}$	$\gamma_{l\ calc.}$ by (5)
Bending deformation ($\Delta\varepsilon_l$ = 0—0.06%)					
Cr(60)/Co(90)/S	26.7	16.1	Co(65)/Cr(60)/S	21.0	16.3
Ni(60)/Co(30)/S	14.4	3.6	Co(80)/Cr(50)/S	9.0	11.1
Ni(50)/Cr(70)/S	7.7	4.7	Cr(60)/Ni(30)/S	8.7	5.0
Stretching deformation ($\Delta\varepsilon_l$ = 0—4%)					
Ni(115)/Co(90)/S	3.9	4.0	Cr(60)/Co(90)/S	26.7	15.1
Ni(70)/Cr(80)/S	12.9	3.2	Ni(60)/Co(30)/S	14.4	15.6
Co(90)/Cr(120)/S	11.8	5.0	Ni(90)/Cr(70)/S	9.0	11.6
Co(100)/Ni(70)/S	5.0	2.0	Co(60)/Cr(60)/S	22.0	18.6
Cr(110)/Ni(90)/S	1.5	1.1	Co(90)/Ni(50)/S	8.0	17.2

It is important here to emphasize, that a level of understanding of physical processes, occurring in a double-layer sample, largely predetermine understanding of these processes in multilayer films.

4.1. Double-layer films

The most characteristic data on TCR for double-layer films are generalized in Table 1. The calculated values have been obtained both using the expression (2) and the expression (7), in which the summation to the index i was taken from 1 up to 2.

Let us note, that in view of the affinity of the calculated results according to the expressions (2) and (7), Table lists only the calculation and the auxiliary data for a microscopic model. The significant difference in the experimental and calculated values can be explained by the influence of the residual atmosphere $(\Delta\beta_f)_{tech}$, macrostresses, arising on the interface, $(\Delta\beta_f)_S S$ and the interdiffusion processes $(\Delta\beta_f)_{dif}$. The contribution of the first two mechanism to our experimental conditions is 16—20% and 2—5%, respectively.

Hence, the main mechanism, which influences the difference of the experimental and calculated values of β most essentially, is the interdiffusion of elements. The results of the researches of these processes have been repeatedly published by us earlier [14].

The simplicity of the formula (7) permits to calculate the size dependence b_f, on the thickness d_2 with the fixed thickness of the lower layer d_1.

In Ref. [10] it has been shown, that the laws of size effects on TCR of double-layer films essentially differ from those of the one-layer films; when the thickness of the upper layer (d_1 = const) increases, the value β increases (if $\beta_1 < \beta_{g2}$ or $\beta_{g1} < \beta_{g2}$) or decreases (if $\beta_{f1} > \beta_{g2}$ or $\beta_{g1} > \beta_{g2}$), taking the asymptotic value β_{g2}, while in one-layer films β is only increasing monotonously with growing thickness d_1 taking the value β_{g1}. The diagrams, which show the size effects on TCR for Cr/Co, Co/Cr, Ni/Co and Co/Ni are represented in Fig. 1. For other film systems the similar diagrams take place. Experimentally the plotted diagrams are completely similar to the calculated ones, but a correspondence between them is the same as in Table 1.

Additionally, let us note that in calculations according to the expression (2) the earlier obtained data for one-layer films on such values as λ_0, p and the transmittance coefficient r, which had been calculated in the framework of the linearization expression [15] or in the framework of an isotropic model [16] were used.

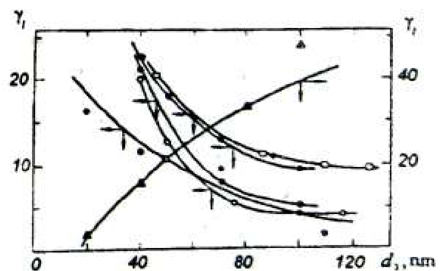

Fig. 2. The dependence γ_l on d_2 for double-layer films: ◒ — Ni/Cr (d_1 = 80 nm); □ — Co/Cr (d_1 = 120 nm); • — Co/Ni (d_1 = 70 nm); ○ — Ni/Co (d_1 = 90 nm); ◐ — Cr/Ni (d_1 = 90 nm); ▲ — Cr/Co (d_1 = 95 nm)

Table 3. **Comparison of experimental and calculated data of the TCR**

Films (thickness, nm)	$\beta \cdot 10^3$, K^{-1} experimental data	$\beta \cdot 10^3$, K^{-1} data calculated by (2)
Cr(60)/Co(30)/Cr(40)/S	0.88	1.22
Co(50)/Cr(50)/Co(35)/S	1.29	1.46
Ni(50)/Co(20)/Ni(20)/S	1.86	2.38
Co(20)/Ni(40)/Co(20)/S	2.05	2.43
Cr(20)/Co(20)/Ni(20)/S	1.80	2.32
Ni(75)/Co(75)/Cr(65)/S	1.25	2.09
Cr(150)/Co(10)/Cr(80)/Co(10)/S	0.42	1.04
Co(10)/Cr(150)/Co(10)/Cr(80)/Co(10)/S	0.9	0.87
Cr(80)/Co(10)/Cr(150)/ Co(10)/Cr(80)/Co(10)/S	0.32	1.05
Co(10)/Cr(80)/Co(10)/Cr(150)/ Co(10)/Cr(80)/Co(10)/S	1.15	0.93
Cr(150)/Co(10)/Cr(80)/Co(10)/ Cr(150)/Co(10)/Cr(80)/Co(10)/S	0.91	1.12
Co(12)/Ni(20)/Co(12)/Ni(20)//Co(12)/Ni(20)/Co(12)/Ni(20) /Co(12)/Ni(20)/S	1.9	2.84

Turning to the phenomenon of the strain sensitivity, let us note, that the comparison with the experimental data of expressions (5) and (8) was performed using different series of experiments, *i.e.* for the various values of d_1 and d_2 (Table 2).

Figure 2 permits to understand the size de-

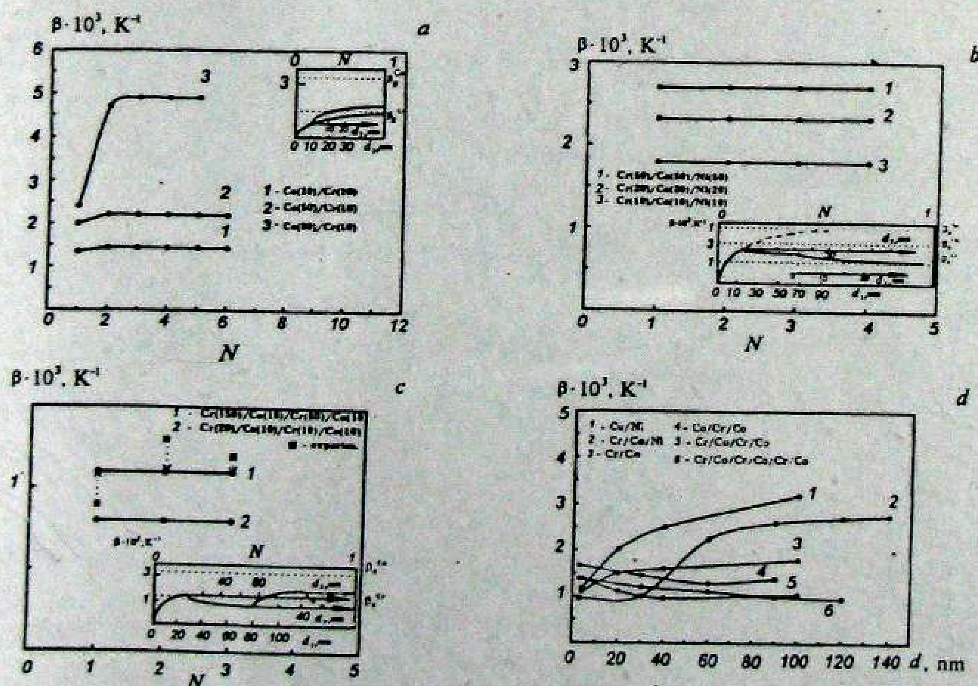

Fig. 3. The dependence β on the number (*a—c*) and the thickness (*d*) of the fragment. In the insert there is a change β within the separate fragment

Table 4. **Comparison of experimental and calculated data on the longitudinal strain sensitivity coefficient**

Films (thickness, nm)	$\gamma_{l\,exper.}$	$\gamma_{l\,calc.}$ by (8)
Cr(20)/Co(20)/Ni(20)/S	22.7	24.3
Ni(75)/Co(75)/Cr(65)/S	24.4	13.8
Co(50)/Cr(50)/Co(55)/S	19.8	21.2
Cr(60)/Co(30)/Cr(40)/S	21.6	22.1
Ni(50)/Cr(10)/Ni(50)/Cr(10)/S	20.7	22.2
Ni(30)/Co(30)/Cr(30)/Ni(30)/Co(30)/Cr(30)/S	50.0	52.5
Ni(50)/Cr(10)/Ni(50)/Cr(10)/Ni(50)/Cr(10)/Ni(50)/Cr(10)/Ni(50)/Cr(10)/S	21.7	22.2

pendence γ_l on the thickness of the upper layer with the fixed thickness of the base one under stretching deformation. Let us note, that in comparison with the other film pairs the inversion of dependence for Cr/Co films can be explained both from the point of view of a stronger surface scattering if to compare with that of the grain interfaces and by the influence of atoms of the residual atmosphere, which seems to be more probable. The same anomaly takes place also in Ni/Co/S films under bending deformation.

If to generalise all the obtained data on the tensosensibility, one can make the following conclusions.

First, the value γ_l may differ up to two times when measured in the region of small deformations (bending) and large (stretching).

Second, in double-layer films the value γ_l is higher in comparison with one-layer ones of the same thickness, the ratio being as follows: γ_l/γ_{l1} = = 1.3—3.1 and γ_l/γ_{l2} = 1.4—2.4 (Cr/Ni, Ni/Cr, Cr/Co, Co/Cr, Ni/Co films) or γ_l/γ_{l1} = 1.0—1.1 and γ_l/γ_{l2} = 1.0—1.1 (Co/Ni films). This seems to be explained by an additional electron scattering at the layer interface in the process of film deformation.

4.2. Multilayer films

To investigate the size effect on TCR and CS, a series of samples of a multilayer structure of general type and a multilayer periodic structure with various numbers of layers in a fragment and various thicknesses has been prepared. Table 3 represents a degree of conformity between the experimental and calculated data on TCR (according to the expression (2), where $i = 1,2$, for i from 1 to n).

Figure 3 shows the calculated (on the basis of the experimental data for one-layer films) dependence β_f on the number of fragments (N) and the thickness of the fragment (d_f). The inserts in this figure show the experimental dependences $\beta_f(d)$ within the limits of the fragment. The data of Table 3 and Fig. 3 permit to make following conclusions.

Practically, starting from the second fragment, the value β is hardly changed. The thickness d_f defines it size dependence. In spite of the fact that β does not almost change at $N > 2$—3, within the limits of each fragment is observed size oscillation dependence, on character similar to that, that is observed in the case of two-layer films (Fig. 1). In accordance with an increase N this oscillation dependence is completely smoothed.

Table 4 gives an idea about the level of accordance between the experimental and calculated data of strain sensitivity coefficient of multilayer films.

In Fig. 4 the dependences CS on number of fragments and thickness of i-layer are submitted. As well as in the case TCR, the value of CS is weak depends from N, though strongly depends from d_f and d_i within the limits of a fragment.

The value γ_l and it size dependence is completely defined by quantity of layers and value γ_{li}. For any (i + 1)-th layer, γ_l has larger value in comparison with γ_l for i layers.

At increasing thickness $d_{(i+1)}$, γ_l at first grows and then decreases, coming nearer to asymptotic significance $\gamma_{gl(i+1)}$. In Fig. 4, d the settlement de-

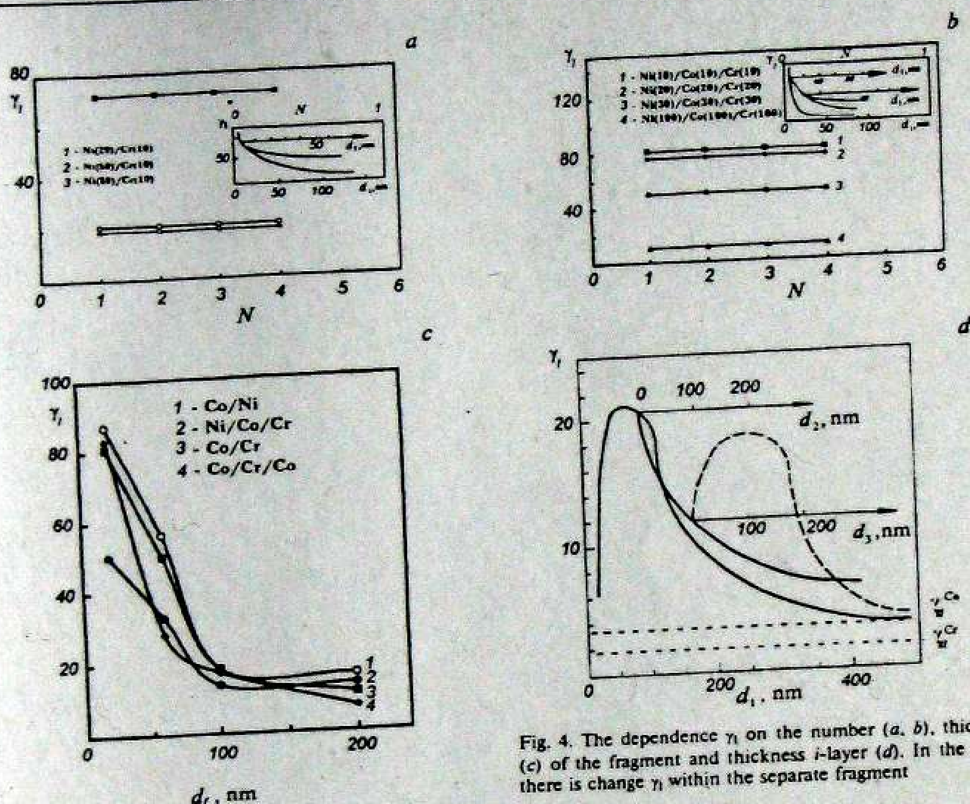

Fig. 4. The dependence γ_l on the number (*a*, *b*), thickness (*c*) of the fragment and thickness *i*-layer (*d*). In the insert there is change γ_l within the separate fragment

pendences, about a degree of conformity of which to experimental results are submitted, it is possible to judge under Table 4.

So, receiving result allowed prognosis of electrophysical properties of multilayer films by macroscopic theory.

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