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Experimental Test of a Three-Dimensional Model for Electrophysical Properties of Metal Films

Anatoly M. CHORNOUS, Nadia M. OPANASYUK, Alexander D. POGREBNJAK[1,2,*] and Ivan Yu. PROTSENKO

[1]*Sumy State University, R.-Korsakov str. 2, 40007 Sumy, Ukraine*
[2]*Sumy Institute for Modification, P.O. Box 163, 40030 Sumy, Ukraine*
[3]*Department of Electrical Engineering, Laboratory of Beam Technology, Nagaoka University of Technology, Nagaoka, Niigata 940-2188, Japan*

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A three-dimensional model of strain sensitivity proposed by [Tellier, Tosser: Thin Solid Films **59** (1979) 163; Tosser, Tellier and Pichard: J. Mater. Sci. **16** (1981) 944] has been tested for thin Cr, Cu and Co films. The films were obtained by electron-beam evaporation in a vacuum of 10^{-4}–10^{-5} Pa. Film structure stabilization was carried out by heating and cooling at the rate of 3 K/min in the range of 300 to 520 K. The identity of properties of the films obtained on the glass (during the termal coefficient of resistance (TCR) measuring) and the textolite glass (during the strain-sensitivity coefficient (SSC) measuring) substrates was examined according to Vand method on lattice distortion energy spectra for films of different thickness, where the spectra were calculated from the resistance-temperature data. It has been shown that the experimental results of the strain sensitivity agree with the calculated ones only under the assumption of size dependence of the electron mean-free path.

KEYWORDS: thin film, size effects, thermal coefficient of resistance, strain sensitivity, grain boundary

1. Introduction

The problem of the determination of electron transfer parameters of metal films is being considered by both experimenters and theorists. Recently, several theoretical models, which take into account both external and internal size effects (SEs), have been proposed. In particular, the models involving the effective free path, the linearized relations of the Mayadas—Shatzkes model, the isotropic scattering of electron current carriers, the thermal coefficient of resistance (TCR)[1–3] and the strain-sensitivity coefficient (SSC)[2,4] have been extensively applied. A general disadvantage of these models is that they can be applied either when $L > d$ (where L is average crystallite size and d is thickness)[1,2] or when the ratio between L and d is arbitrary, assuming a crystal isotropy. Further development of the SE theory for the TCR and SSC is confirmed by the three-dimensional model, which assumes that crystals are of arbitrary form, and that their sizes, L_x, L_y and L_z, do not coincide in general cases.

The purpose of the present work was to test the three-dimensional model of SEs for SSC. As investigation objects, copper, cobalt and chromium films were determined for the above purpose, since the L in Cr films and Co films is smaller, and in Cu films it is greater than the thickness of the samples, which permits testing of the model for these two cases. It should be noted that testing of the three-dimensional model for TCR with Sn and Pb films was performed for the first time by Pichard *et al.*[5] And with Sc and Re it was performed by Protsenko.[6] We carried out the testing of the three-dimensional model for SSC first.

2. Basic Relations

To take into account both external and internal SEs Tosser *et al.*[3] introduced two parameters of electron scattering: one is for a grain boundary (ν), and another is for a film surface (μ):

$$\nu = L\lambda_0^{-1}\left(\ln\frac{1}{r}\right)^{-1}, \quad \mu = d\lambda_0^{-1}\left(\ln\frac{1}{p}\right)^{-1} \tag{1}$$

where λ_0 is the mean-free path (MFP) of electrons in the bulk; r is the transmission coefficient at the grain boundary (GB), which is related to R (the reflectivity coefficient of the GB) as $R(1-R)^{-1} \cong \ln(r^{-1})$;[3] p is the reflectivity coefficient of the external surfaces.

2.1 *TCR of metallic films*

The most common relation, which relates the TCR to the parameters ν, μ and $a = (\nu + c^2)(1-c)^{-1}$, where $c = 4/\pi$, has been obtained from ref. 7 as

$$\frac{\beta_g}{\beta_0} = \frac{\nu}{1-c}\frac{a^{-1} - 2 + 2a\ln(1+a^{-1})}{a - 1/2 + (1-a^2)\ln(1+a^{-1})} = \frac{\nu}{1-c}\cdot\frac{V(a)}{U(a)}. \tag{2}$$

where β_g and β_0 are the TCR of an infinitely thick polycrystalline film ($d \to \infty$) and a bulk sample, respectively.

Equation (2) was detailed in ref.8 for the case of the polycrystalline film (p-index), or one which satisfies the conditions of monocrystallinity (m-index):

$$\frac{\beta_p}{\beta_0} = \frac{a_p}{1 + \dfrac{c^2}{\nu}}\cdot\frac{V(a_p)}{U(a_p)}, \quad \frac{\beta_m}{\beta_0} = \frac{a_m}{1 + \dfrac{c^2}{\nu}}\cdot\frac{V(a_m)}{U(a_m)},$$

where

$$a_p = \left(1 + \frac{c^2}{\nu}\right)b_p^{-1}, \quad a_m = \left(1 + \frac{c^2}{\nu}\right)b_m^{-1},$$

$$b_p = \frac{1}{\mu} + \frac{1-c}{\mu}, \quad b_m = \frac{1}{\mu} - \frac{c}{\nu}.$$

In ref. 8, the relation for the TCR of polycrystal films was obtained. It was linearized over parameters ν and μ under conditions $0,1 < \nu < 4$ and $\mu > 0,1$. A similar simplification was made in ref. 9 under the condition of sample monocrystallinity, i.e., when $\mu \ll 1 \ll \nu$ and $p < 1$, $r \cong 1$:

*E-mail address: apogrebnjak@simp.sumy.ua

$$\left(\beta_m \ln\frac{\lambda_0}{d}\right)^{-1} \approx \beta_0^{-1}\cdot\left(1+\frac{c^2}{\nu}\right)\left(1+\left(\ln\frac{\lambda_0}{d}\right)^{-1}\cdot\ln\frac{\ln\left(\frac{1}{p}\right)}{1+\frac{c^2}{\nu}}\right). \quad (3)$$

When films are polycrystalline and the condition $\mu < 1$ and $\nu > 1$ is satisfied, one can use the similar linearized relation obtained from ref. 8:

$$\left(\beta_p \ln\frac{\lambda_0}{d}\right)^{-1} \approx 1.43\beta_0^{-1}\cdot\left(1+\frac{c^2}{\nu}\right)\left(1+\left(\ln\frac{\lambda_0}{d}\right)^{-1}\cdot\ln\frac{\ln\left(\frac{1}{p}\right)}{1+\frac{c^2}{\nu}}\right). \quad (4)$$

2.2 *SSC of metallic films*

The three-dimensional model of strain sensitivity may be applied when $\nu > 0,4$ for the coefficient of longitudinal strain sensitivity (γ_l); its relation has the following form:

$$\gamma_l = (\eta_l + 1) - \eta_l\frac{F^*(\nu_x)+G^*(\nu_y)+G^*(\alpha^*)}{M(\nu_x,\nu_y,\alpha^*)} + \frac{\mu_s G^*(\nu_y) - F^*(\nu_x) - \mu' G^*(\alpha^*)}{M(\nu_x,\nu_y,\alpha^*)}, \quad (5)$$

where $M(\nu_x,\nu_y,\alpha^*) = F(\nu_x)^{-1}+G(\nu_y)^{-1}+G(\alpha^*)^{-1}-2 = \frac{\rho}{\rho_0}$, ($\rho$, ρ_0 are the specific resistance of the film and the bulk sample, respectively); $(\alpha^*)^{-1} = \mu^{-1}+\nu_z^{-1}$; the functions $F(\nu_x)$, $G(\nu_y)$, $G^*(\alpha^*)$, their derivatives $f(\nu_x) = \frac{dF}{d\nu_x}$, $g(\nu_y) = \frac{dG}{d\nu_y}$, $g(\alpha^*) = \frac{dG}{d\alpha^*}$—are summarized in tables in ref. 3, and $F^*(\nu_x) = \nu_x f(\nu_x) F(\nu_x)^{-2}$;

$$G^*(\nu_y) = \nu_y g(\nu_y) G(\nu_y)^{-2};\ G^*(\alpha^*) = \alpha^* g(\alpha^*) G^*(\alpha^*)^{-2};$$

μ_s, μ' are Poisson coefficients of a substrate material and a reduced Poisson coefficient, respectively; $\eta_l = -\lambda_0^{-1}\frac{d\lambda_0}{d\varepsilon_l}$ is a deformation coefficient of the MFP (ε_l is a longitudinal deformation).

3. Experimental Technique

Cr, Cu and Co films were obtained by electron-beam evaporation and where thermally treated using the apparatus with a magnetically discharged pump (10^{-4}–10^{-5} Pa). Glass plates were used for TCR measurements. We used textolite substrates for SSC measurements.

The element composition of samples was studied by secondary-ion mass spectrometry. To measure the mean size of crystallites L_x and L_y in electron-microscopy investigations, the sample was oriented so that the x-axis direction coincided with the current flow direction in resistance measurements. Size L_z was assumed to be equal to the film thickness.

The SSC was calculated using an angular coefficient of the deformation dependence $\Delta R/R_0$ on (ε_l (R_0 is an initial resistance, ΔR is its change in deformation, $\varepsilon_l = \frac{\Delta l}{l_0}$, where l_0 is an initial film length), which was obtained as a result of a longitudinal deformation of the film on the substrate after tension by a microscrew ($\varepsilon_{max} = 2\times10^{-2}$).

Annealing samples for the purpose of their recrystallization and stabilization of their electrical properties was realized in a vacuum chamber, according to a "heating-cooling" scheme (2-3 circuits) from 300 to 520 K with 3 K/min. The heat treatment was carried out in such a way that the maximum height of the F_{om} spectrum and the corresponding activation energy E_m (see ref. 11 for details) coincided with those of film samples of the metal obtained on glass or textolite substrates. Thus, the maximum for the films investigated was fixed on the spectrum with the following values of E_m: 0.55; 0.60–0.65 and 0.80 eV at $F_{om} \sim 10^{-8}$ Ωm/eV (Cr); 0.70 and 0.80 eV at $F_{om} \sim 10^{-8}$ Ωm/eV (Cu) and 0.62–0.65; 0.70–0.75 and 0.81 eV at $F_{om} \sim 10^{-7}$ Ωm/eV (Co). The initial state of the film as well as the substrate material is of no consequence with this control technique.

The technique of measurement of the longitudinal coefficient of strain sensitivity consists of the determination of the relative resistance change, $\Delta R/R_0$ under stretching deformation of a film on a substrate ($\Delta\varepsilon_l = 0$–2%) with the aid of a microscrew.

The element composition was investigated by secondary-ion mass-spectrometry (measuring apparatus—MS7201 M) using argon to produce primary ions.

4. Results and Discussion

The experimental dependencies of the TCR on thicknesses are presented in Fig. 1 in different coordinates. A rough estimation of the μ and ν parameters and values of p and r coefficients reveals that the results for Cu films may be described within the framework of eq. (3), while for Cr and Co films this would be eq. (4). Using MFP λ_0 values obtained from ref. 9 in the model of isotropic scattering[3] the precise values of p and r parameters were calculated on the basis of relations presented in Figs. 1(b), and 1(c) (along the intercept on the abscissa axis and the angle coefficient, p and r results are presented in Table I). They were used for detecting μ, ν and α^* parameters under relation testing in eq. (5).

In addition to the experimental data for SSC γ_l, Figure 2 presents the calculated data. As is evident, the poorest correspondence is found when the value $M(\nu_x,\nu_y,\alpha^*) = \rho/\rho_0$ is theoretically calculated. At the same time the application of experimental values ρ/ρ_0 or ρ/ρ_g (where ρ_g is a specific resistance of an infinitely thick film ($d \to \infty$), i.e., a bulk sample of the same type and concentration of defects as in a film) results in an approximately equal value γ_l, which differs essentially from the experimental one. This allows the conclusion that the deformation coefficient of the MFP is a size-dependent parameter of an electroconductivity (Fig. 2). It can be accounted for by the MFP depending on the

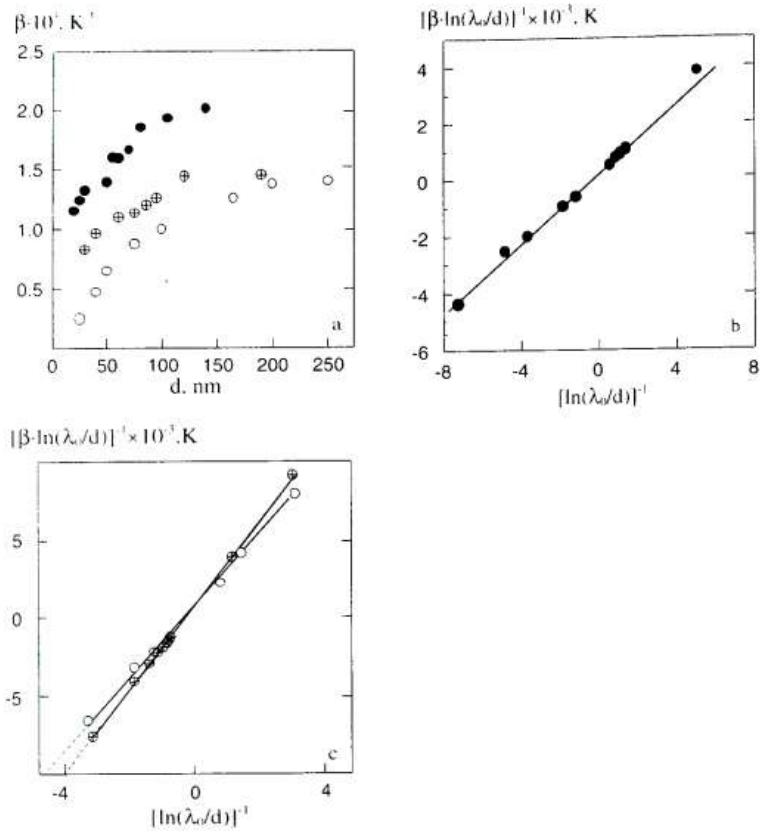

Fig. 1. The size dependence of the TCR for films: ○: Cr, ⊕: Cu, ●: Co at different coordinates ($T = 300$ K).

Table I. The parameters of electron transfer.

Films	λ_∞, nm[9]	r	p
Cr	129	0.99–0.93	0.01
Cu	39	0.80–0.72	0.001
Co	61	0.99–0.87	0.001

film thickness. The poor agreement of the three-dimensional model with the experimental results under relatively small thickness is due to the theory when η_l = const. The idea of size dependence of the given parameter (film thickness and grain size) cannot bring about objections even for the consideration that many different physical quantities also depend on the film thickness. It is essential to stress, however, that in the formulae the description on of the three-dimensional model of strain sensitivity there are some values that can hardly be measured experimentally as: L_x, L_y, L_z and, respectively, r_x, r_y, α'. We could also observe that even an insignificant change in the size and form of a grain resulted in an essential change of ρ and γ_l. In this case the considerable part play the grain boundaries and their types (the small-angle type and the high-angle type) as well as the concentration of impurity atoms, which localize on the boundaries, etc.

5. Conclusions

The obtained results permit to come to the conclusion that the basic influence on the value γ_l produces deformation coefficient η_l in thin films as well as in the bulk samples since surface scattering of electrons does not influence on the value of γ_l but influences only on its dimensional correlation and the scatter of electrons on the (GB) under the defined conditions may come either to the decreasing or to the increasing of SSC. The degree of difference of SSC in the films and in the bulk samples depends on the difference in the value of η_l completely.

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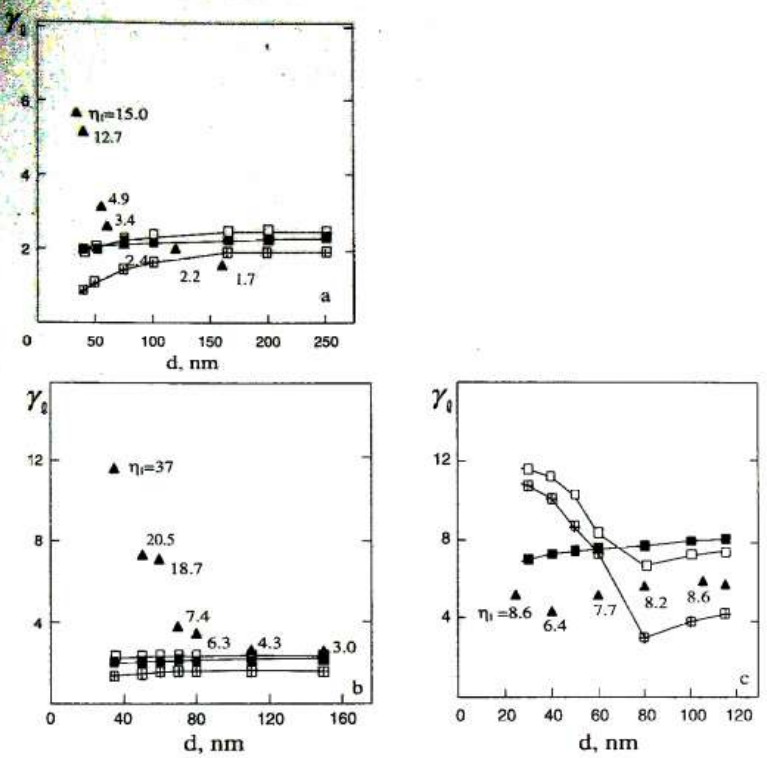

Fig. 2. The size dependence of the SSC for films Cr (a), Cu (b) and Co (c): ■: obtained using the calculated ρ/ρ_0 value; □: using the experimental ρ/ρ_0 value; ⊞: using the experimental ρ/ρ_g value; ▲:experimental plot. The numbers presented in the vicinity of point ▲ are the calculated value of the deformation coefficient η_t obtained using the experimental ρ/ρ_0 value.

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