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

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A three-dimensional model of strain sensitivity proposed by [1] for thin Cr, Cu and Co films. The films were obtained by electron-beam evaporation in a vacuum of 10-1 - 10-4 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 thermal coefficient of resistance (TCR) measurement) and the metallic glass (during the strain-sensitivity coefficient (SSC) measurement) substrates was examined according to Frenkel method on lattice distortion energy spectra for films of different thickness, where the spectra were calculated from the resistivity-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 films, strain 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 (SEEs), have been proposed. In particular, the models involving the effective free path, the linearized relations of the Mayak—Schatkes model, the isotropic scattering of electron current carriers, the thermal coefficient of resistance (TCR)1,2 and the strain-sensitivity coefficient (SSC)2,3 have been extensively applied. A general disadvantage of these models is that they can be applied either when \( L > d \) (where \( L \) is the average crystallite size and \( d \) is thickness)1,2 or when the ratio between \( L \) and \( d \) is arbitrary, assuming a crystalline structure. 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, \( L_1, L_2 \), and \( L_3 \), 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.1,2 And with Sc and Re it was performed by Protsenko.1,2 We carried out the testing of the three-dimensional model for SSC first.

2. Basic Relations

To take into account both external and internal SEEs, Tosser et al.1,2 introduced two parameters of electron scattering: one is for a grain boundary (\( \nu \)), and another is for a film surface (\( \mu \))

\[
\nu = \frac{L_{in}^{-1} \ln \left( \frac{1}{r} \right)^{-1}}{r}, \quad \mu = \frac{d_{in}^{-1} \ln \left( \frac{1}{p} \right)^{-1}}{p},
\]

where \( L_{in} \) 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 \( R(1 - R) \approx \ln(r^{-1})^2 \); \( p \) 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 \( \nu, \mu \) and \( \sigma = (\nu + \sigma) \xi (1 - 1) \), where \( \xi = 4/\pi \), has been obtained from ref. 7 as

\[
\beta_2 = \frac{v}{1 - \sigma} - 2 \sigma \ln(1 + a^{-1}), \quad \beta_0 = \frac{v}{1 - \sigma} \ln(1 + a^{-1})
\]

where \( a \) is the TCR of an infinitely thick polycrystalline film \( d \rightarrow \infty \) and \( \beta_0 \) is the TCR of 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):

\[
\beta_p = \frac{a_p}{1 + \frac{a_p U(a_p)^{-1}}{U(a_p)^{-1}}}, \quad \beta_m = \frac{a_m}{1 + \frac{a_m U(a_m)^{-1}}{U(a_m)^{-1}}},
\]

where

\[
a_p = \left( 1 + \frac{\nu}{\mu} \right)^{-1}, \quad a_m = \left( 1 + \frac{\nu}{\mu} \right)^{-1}
\]

In ref. 8, the relation for the TCR of polycrystalline films was obtained. It was linearized over parameters \( \nu \) and \( \mu \) under conditions \( 0 < \nu < 4 \) 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 \( p \ll 1, r \gg 1 \):
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:

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

![Image](image0.png)

### 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 ($\gamma_1$); its relation has the following form:

$$
\gamma_1 = (\eta_1 + 1) - \frac{F(v_1) + G(v_1) + G(\alpha^*)}{M(v_1, v_2, \alpha^*)} + \frac{\mu_1 G(v_2) - F(v_1) - \mu G(\alpha^*)}{M(v_1, v_2, \alpha^*)} \tag{5}
$$

where $M(v_1, v_2, \alpha^*) = F(v_1)^{-1} + G(v_1)^{-1} + G(\alpha^*)^{-1} - 2 = \frac{1}{\rho_0}$, $(\rho_0, \rho_0)$ are the specific resistance of the film and the bulk sample, respectively; $(\alpha^*)^{-1} = \mu_1 + \nu_1^{-1}$; the functions $F(v_1)$, $G(v_1)$, $G(\alpha^*)$, their derivatives $f(v_1), g(v_1) = \frac{df}{dv_1}, g(\alpha^*) = \frac{dg}{d\alpha^*}$—are summarized in tables in ref. 3, and $F(v_1) = v_1 f(v_1) F(v_1)^{-2}$.

$$
G(v_1) = v_1 g(v_1) G(v_1)^{-2}, G(\alpha^*) = \alpha^* g(\alpha^*) G(\alpha^*)^{-2};
$$

where $\mu_1, \mu_2$ are Poisson coefficients of a substrate material and a reduced Poisson coefficient, respectively; $\eta_1 = -\frac{\lambda_0}{\alpha^*} \frac{d\alpha^*}{d\alpha^*}$ is a deformation coefficient of the MFP ($\alpha$ 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 crystals Lx and Ly, in electron-microscopy investigations, the sample was oriented so that the x-axis direction coincided with the current flow direction in resistance measurements. Size $L_x$ 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 $(\theta_1 R_0$ is an initial resistance, $\Delta R$ is its change in deformation, $\theta_1 = \frac{A}{L}$, where $A$ 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 microscrope ($\varepsilon_{max} = 2 \times 10^{-2}$).

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 $F_{max}$ spectrum and the corresponding activation energy $E_a$ coincide 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 $E_a$: 0.55–0.60–0.65 and 0.80 eV at $F_{max} \sim 10^{-4}$ $\Omega m/eV$ (Cr); 0.70 and 0.80 eV at $F_{max} \sim 10^{-4}$ $\Omega m/eV$ (Cu) and 0.55–0.65, 0.70–0.75 and 0.81 eV at $F_{max} \sim 10^{-7}$ $\Omega m/eV$ (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 ($\Delta \varepsilon_0 = 0–25\%$) with the aid of a microscrope.

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 $\mu$ and $\nu$ 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 $\lambda_0$ values obtained from ref. 9 in the model of isotropic scattering 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 1). They were used for detecting $\mu$, $\nu$ and $\alpha$ parameters under relation testing in eq. (5).

In addition to the experimental data for SSC $\gamma$, Figure 2 presents the calculated data. As is evident, the poorest correspondence is found when the value $M(v_1, v_2, \alpha^*) = \rho_0$ is theoretically calculated. At the same time the application of experimental values $\rho_0 \rho_0$ or $\rho_0 \rho_0$ (where $\rho_0$ is a specific resistance of an infinitely thick film ($d \rightarrow \infty$), i.e., a bulk sample of the same type and concentration of defects as in a film) results in an approximately equal value $\gamma$, 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
film thickness. The poor agreement of the three-dimensional model with the experimental results under relatively small thickness is due to the theory when \( \eta \) = 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 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, \( \tau, \tau_x, \omega \). We could also observe that even an insignificant change in the size and form of a grain resulted in an essential change of \( \rho \) and \( \gamma \). 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 \( \eta \) produces deformation coefficient \( \eta \) in thin films as well as in the bulk samples since surface scattering of electrons does not influence on the value of \( \gamma \) 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 \( \eta \) 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 $\rho/\rho_0$ value; □ using the experimental $\rho/\rho_0$ value; ● using the experimental $\rho/\rho_0$ value; ▲ experimental plot. The numbers presented in the vicinity of point ▲ are the calculated value of the deformation coefficient $\eta_f$ obtained using the experimental $\rho/\rho_0$ value.
