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A Charge Transport in Ultrathin Electrically Continuous Metal Films

R. I. Bigun, Yu. A. Kunitsky*, and Z. V. Stasyuk

*Ivan Franko Lviv National University,
1, Universytetska Str.,
79000 Lviv, Ukraine
*Technical Centre, N.A.S. of Ukraine,
13, Pokrovs'ka Str.,
04070 Kyiv, Ukraine*

The problems of ultrathin stable electrically continuous metal films fabrication and their electron-transport properties are discussed. To prevent the coagulation process of metal grains during metal film condensation, the surfactant underlayers utilizing is discussed. Analysis of current theoretical concepts concerning electron-transport properties of metal film is performed. The experimental data are explained within the scope of the modern theoretical models.

Обговорено проблему створення надтонких (товщина шару від 2 нм до 50 нм) електрично суцільних стабільних провідних шарів металів і вивчення їхніх електричних властивостей. Розглянуто можливість застосування сурфактантних підшарів для запобігання коагуляції зародків кристалізації в процесі росту плівок. Здійснено аналізу сучасного стану модельних уявлень про перенесення заряду в металевих зразках обмежених розмірів, і на його основі проведено трактування результатів експериментального дослідження надтонких металевих плівок.

Обсуждается проблема создания сверхтонких (толщина слоя от 2 нм до 50 нм) электрически сплошных проводящих стабильных слоев металлов и исследования их электрических свойств. Рассмотрена возможность применения сурфактантных подслоев для предотвращения коагуляции зародышей кристаллизации в процессе роста пленок. Сделан анализ современного состояния модельных представлений о переносе заряда в металлических образцах ограниченных размеров, и на его основе проведена трактовка результатов экспериментального исследования сверхтонких металлических пленок.

Key words: thin films, classical and quantum size effects, charge surface scattering, surfactant sublayer, ballistic charge transport.

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1. INTRODUCTION

Thin layers of substance are basic elements of many devices of modern electronic techniques. The further development of electronics is impossible without microminiaturisation of electronic systems by nanotechnology, in particular, by techniques of stable ultrathin covering formation.

Properties of ultrathin slabs can essentially differ from properties concerning thick layers, which are used in up-to-date engineering. This difference is caused, above all, by prevailing influence of the surface phenomena on ultrathin layer structure and electric parameters. In our work, the current state of theoretical and experimental researches on ballistic charge transport in ultrathin (layer thicknesses are 2–12 nm) electrically continuous metal films (temperature coefficient of resistance $\beta > 0$) under the condition of inequality realisation $d < l$ is analysed. Here, d is the film thickness; l is the charge mean free path. Crystal lattice parameters and the crystalline average linear sizes are considered as peculiarities of film structure.

The ultrathin electrically continuous metal film deposition on dielectric substrate surface is a problem of considerable difficulty due to the action of surface tension forces. These phenomena lead to coagulation of metal particles. As a result, there is some metal layer critical thickness d_c , at which current starts to flow into a metal film (*percolation threshold* is observed here). The mean d_c is determined by technological features of film formation (the rate of material condensation, the substrate temperature at layer deposition, the modes of further heat treatment) as well as by the properties of condensing material, in particular by its fusion temperature. Essential decrease of d_c may be reached at epitaxial growth of a metal film on the oriented substrate. The use of surfactant underlayers of subatomic thickness preliminary deposited on a dielectric substrate inhibits coagulation of metal condensates in other effective way of d_c decrease.

The mentioned technique makes possible formation of ultrathin conductive coatings of several atom layers of metal in thickness. In particular, the Hall voltage investigation on 1–3 nm thickness chrome films deposited on germanium surfactant underlayer was performed in [1]. Electrically continuous ultrathin films of some metals also have been obtained due to application of surfactant underlayer (see, for example, [2–4]). We shall analyse some peculiarities of modern view on the mechanisms of charge carrier's relaxation in ultrathin layers and the application of these theoretical models for the experimental results treatment.

2. CHARGE TRANSPORT MODELS IN SIZE-LIMITED METAL SAMPLES

Thin film is a classical example of the size-limited sample in which surface phenomena play an essential role owing to the restriction in one of the film sizes. The relative contribution of these phenomena can vary from the negligibly small to the dominating due to the thickness d changes. In Figure 1, the areas of films thickness are specified for various mechanisms of carrier relaxation in metal films. In a mode of diffusive charge-transport scattering, which is observed in films of micron thickness, the charge transport phenomena are well described within the framework of free electron model. The electrophysical properties of films are basically determined by the processes occurring in the film volume. When the mean free path of electron becomes commensurable to the thickness of a metal film d , the electron-transport phenomena are essentially influenced by electron scattering on film surface. Thus, the contribution of surface scattering to the total electron relaxation time is close to the contribution of volume scattering. The kinetic parameters thickness dependence of electrically continuous metals films is described within the framework of the classical size effect theory (the theory of Fuchs–Sondheimer [5, 6] and its modifications) and internal size effects theory (Mayadas–Shatzkes [7], Tellier–Tosser–Pichard [8], Varkusz [9] models). Those are the models, in

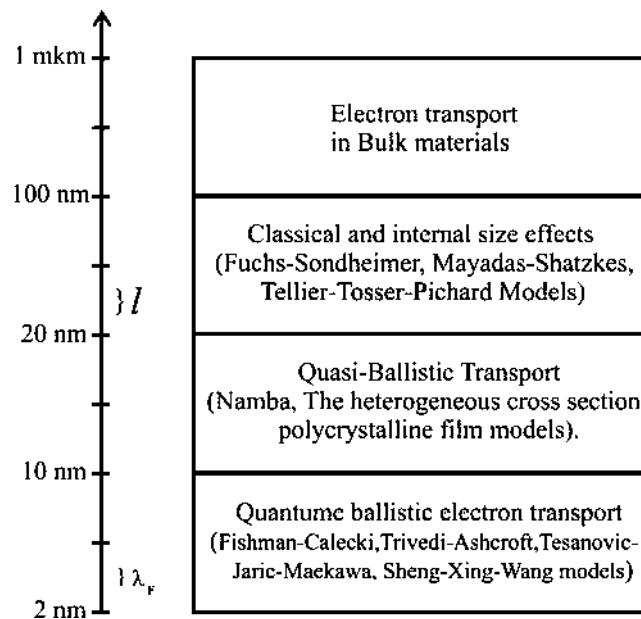


Fig. 1. Charge transport models in finite-size metal samples.

which the contribution to total relaxation time of electron surface scattering on flat external film surfaces and on grain boundaries in the film volume are considered. The account of the electron scattering contribution on grain boundaries in the most cases is necessary, as in the massive polycrystalline metal samples, the mean linear grain sizes essentially exceed the charge carriers mean free path l . The mean grain linear sizes in metal films are usually less or commensurable to l .

With further reduction of metal layer thickness when the electron mean free path satisfies the condition $d < l$, the quasi-ballistic electron transport in a film (without changes of the power spectrum of electron in metal film) is realised. Thus, charge carriers surface scattering in metal film becomes dominating. The contribution of surface scattering has essentially influenced the macroscopic surface inhomogeneity because the mean linear grain sizes are commensurable to film thickness. The quasi-ballistic electron transport in metal films can be described by size dependencies of kinetic coefficients proposed in Namba theory [10] and within the framework of polycrystalline layer heterogeneous cross section [11]. The treatment of experimental data by the mentioned theories makes possible the reliable calculation of the average amplitude of one-dimensional surface asperity h . The calculated values h correlate well with the results of direct microscopy tunnel scanning structure investigation. It should be noticed that in terms of quasi-ballistic electron transport the film state and the surface morphology play a dominant role in charge carriers' relaxation. The detailed analysis considered above of the geometrical size effect theories and the discussion of the possibilities of their application for experimental results explanation was carried out in [12].

When the film thickness does not exceed 5–8 nm, the quantum effects, which have influence on electron transport in film, are possible. We will consider the general regularities of these phenomena on the example of the influence of size restriction along Z -axis in thin film thickness direction in Sommerfeld electron gas, possessing a spherical form of Fermi surface. In an initial stage of size restriction, the k_z quantization (k_z is quasi-impulse component) is observed. As a result, there is a set of the discrete resolved k_z states on spherical Fermi surface. The evident display of quasi-impulse component quantization is oscillation dependences of the metal film kinetic on its thickness coefficient by the oscillation period, which depends on electron de Broglie wavelength.

Further restriction of sample sizes in Z -direction leads to the change of chemical potential level position and to important changes of electronic structure in metal sample. As a result, the oscillation dependences of Fermi energy on a film thickness occur. In this case, the treatment of experimental results of kinetic phenomena in films is rather inconvenient. The calculation of simple metal electron structure of free films was performed in a number of works [13–16], *etc.* It was

shown that the given phenomenon occurs in the films which thickness does not exceed 7–10 atomic layers that is $d < 2\text{--}3$ nm.

Existence of changes in the transport phenomena caused by dimensional quantization was predicted by Lifshitz and Kosevich [17]. Experimentally quantum size effect was found in [18] in the research of semimetal bismuth films properties. The theory of this phenomenon was developed by Sandomirski [19]. Further investigations of size quantization effect on semimetal and semiconductor films properties were implemented in a number of scientific institutions, in particular, under the supervision of Prof. Komnik [20]. Quantum size effects are the most brightly displayed in semimetal films. The length of the electron de Broglie wavelength in these materials is 10 times larger than interatomic distances and, consequently, the interference of electronic waves is influenced poorly by imperfections of film surface. In metal films, the situation is essentially different as the de Broglie electron wavelength is commensurable to interatomic distances. Therefore, to observe oscillations of the kinetic coefficients in thin metals layers, it is necessary to provide high perfection of samples surface structure.

Modern theoretical approaches to quantum size effects in kinetic phenomena of metal films have been developed. In the majority of works, Kubo formalism is used for calculation of surface scattering effect on charge transport under the conditions of size quantization [21–24], being considered the contributions of separate scattering mechanisms, which are non-additive (that is the Mathiessen's rule is violated). The effect of a surface on an electronic system is considered by introduction of a surface potential to Hamiltonian function. Under constant chemical system potential, the density of states and, accordingly, the conductivity in a film plane oscillate with the period, which is equal to a half-length of de Broglie electron wave.

A peculiarity of the mentioned works is ignoring of foreign dispersion of charge carriers contribution in the current carriers relaxation. In the given approach, it is impossible to carry out any coupling of quantum theories results with the known classical theories. In Ref. [25], an attempt was made to coordinate conclusions of quantum and classical theories by introduction of the dissipating potentials caused by the surface impurities on both film surfaces into model Hamiltonian. Owing to it, the film conductivity size dependences reminding similar Fuchs–Sondheimer theory formalism were received. As a result, quantization influence on σ occurs from quasi-classical approach by the consideration of partial conditions nature and taking into account new treatment of angular dependence of smooth surface reflection parameters. The approach [25] to the solving of this problem was developed in [26], where the relationships describing the effect of surface inhomogeneity of various configurations on films conductivity were obtained. The results of the theory were investigated when treat-

ing the data of the experimental researches in CoSi_2 films carried out in numerous works [27] and for gold films [28]. According to the estimation [26], on the average a metal film surface can be considered as atomically smooth, and a quantization condition is the existence of parallel to each other sites of the $L \times L$ size on the surfaces, where $L/a > 2(d/a)^{1/2}$. Here, d is the layer thickness, a is crystal lattice constant. Features of quantum transport should manifest themselves in films of the metals, the thickness of which does not exceed 10 nm.

The problems of transition from classical to quantum charge transport at reduction of film thickness were considered in numerous theoretical works of Moroz and Makarova (see, for example, [29–31]) and Mejerovich with collaborators (see, for example, [32–34]). In these works, questions of the electronic waves interference with surface and the possibilities of kinetic coefficient oscillations were discussed in details. In Ref. [35], new basic approaches to experimental formation of metal size-quantum systems were proposed.

In summary, we noticed that, in works stated above, only qualitative physical picture of the influence of classical and quantum size effects on the peculiarities of charge transport in metals films is presented due to insufficient volume of the publications. An extended review of theoretical works with corresponding mathematical conclusions is prepared for printing and will be published in the nearest future.

2. EXPERIMENTAL RESEARCH OF SIZE QUANTIZATION EFFECT ON CHARGE TRANSPORT IN METAL FILMS

Influence of kinetic phenomena on geometrical size effect in metal films was being investigated for years. The results of these researches were discussed in detail in a number of works. Therefore, we will consider only the experimental works devoted to studying of size quantization influence on the phenomena of charge transport in metal films and works in which issues of transition from prevailing quantum to quasi-classical charge transport are discussed.

Experimental researches of size quantization effect on charge transport in metals films were first carried out by Fisher and Hoffmann [36–38]. Size dependences of platinum films resistivity in thickness range 3–300 nm were investigated. Films were deposited on polished glass by thermal evaporation under high vacuum condition (pressure of residual gases 10^{-5} Pa). It was shown that conductivity size dependence for thick films ($d > 10$ nm) is in good agreement with similar dependence predicted in Fuchs–Sondheimer theory. Within the range of thickness $8 < d < 10$ nm, the size dependence of σ is described by the approximated formula of Namba theory [11] considering presence of macroscopic irregularity on polycrystalline film surface. In the range of small Pt film thickness ($d < 8$ nm), electrical-current size os-

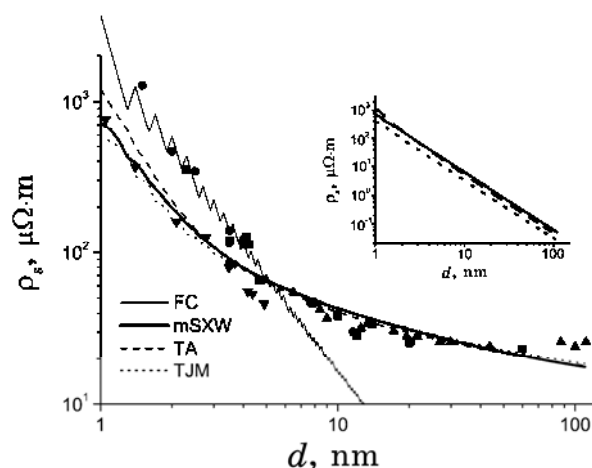


Fig. 2. Size dependences of CoSi_2 resistivity. Points—experimental data. Theoretical curves: FC—[24]; mSXW—[25]; TA—[23]; TJM—[21].

cillations under constant voltage applied to the film showed the oscillation period size dependences with $d_0 = \lambda_F/2$, where λ_F is electron Fermi wave length.

There are a few experimental works devoted to the research of size quantization effect on the electron transport phenomena in such metal films. Probably, the scarcity of these works is caused by the complexity of experiments with this type of metal films. In the majority of the theoretical works, the data of electrical properties of epitaxial CoSi_2 films were used. Metallic behaviour of electron transport in CoSi_2 films is retained until the thickness $d \sim 1$ nm and, in the range of small thickness, the behaviour of conductivity size dependences essentially differs from those foreseen in classical size effect theories. As an illustration, in Fig. 2, the results of CoSi_2 -films resistivity size dependences calculated by four theoretical models of quantum size effect (continuous curves) and some experimental data (the points) are presented [27].

The influence of size quantization on charge transport in lead and gold films was studied in [39, 40]. The conductivity and the Hall constant of lead films under the conditions of size quantization were studied in [41, 42]. The oscillations of resistivity with thickness change of Ag, In, and Ga films deposited on annealed gold and silver films were observed in [43]. The influence of quantum size effect for sliding electrons on electronic conductivity of films of refractory metals was studied in [44].

Direct comparison of experimental results with the corresponding theoretical modelling representations in some cases is inconvenient. Therefore, we will consider the possibility of such a comparison analysing data experimentally obtained in our works. Relative contribution of surface scattering to total time of the relaxation of current carriers

increases with the reduction of film thickness. Thus, if the possible change in the film structure is neglected with the reduction of its thickness, the residual conductivity, which can be written in the form

$$\sigma_{\text{res}} = 1/[\rho(d) - \rho_{\infty}], \quad (1)$$

is the feature of surface scattering contribution. Here, $\rho(d)$ is resistivity of metal film d in thickness, ρ_{∞} is metal film with infinite thickness ($d \rightarrow \infty$) resistivity; a structure being similar to the structure of the investigated film. The analysis of classical size effect theoretical expressions [4, 5] showed that, in all cases, the residual conductivity σ_{res} is directly proportional to the film thickness d . In particular, for the theory of Fuchs–Sondheimer,

$$\sigma_{\text{res}} = 8d/[3\rho_{\infty}l(1-p)]. \quad (2)$$

Here, l is the mean free path of current charge carriers, p —coefficient of surface reflexion. Linearity of the given dependency is broken in the area of thickness, at which quasi-ballistic charge transport takes place $l > d$ [10, 11]. In this case, the film thickness is irregular in charge transport direction due to the macroscopic surface asperities existing in a polycrystalline film. It should be noted that size dependence of polycrystalline film resistivity in the presence of surface asperities with amplitude h was obtained in [11]:

$$\rho(\bar{d}) = \rho_{\infty} \left[1 - \left(\frac{h}{\bar{d}} \right)^2 \right]^{-1/2} \left\{ 1 + \frac{3l_0(1-p)}{8\bar{d}} \left[1 - \left(\frac{h}{\bar{d}} \right)^2 \right]^{-1} \right\}. \quad (3)$$

This dependence may be easily transformed into corresponding formulae of theories [7] and [9], provided that $h \ll \bar{d}$. Here, \bar{d} is an average thickness of a non-uniform thickness film, ρ_0 and l_0 —resistivity and the mean free path of current charge carriers (the characteristic of a single crystal sample), $f(\alpha)$ —grain-boundary function of Mayadas–Shatzkes [7].

In the case, when $h \leq \bar{d}$, considering that $\rho_0 = \rho_{\infty}f(\alpha)$, and $l = l_0f(\alpha)$ [7], the expression (3) is transformed into the known approximated expression $\rho = \rho(\bar{d})$ of Namba theory [10]:

$$\rho(\bar{d}) = \rho_{\infty} \left[1 - \left(\frac{h}{\bar{d}} \right)^2 \right]^{-1/2} \left\{ 1 + \frac{3l_0(1-p)}{8\bar{d}} \left[1 - \left(\frac{h}{\bar{d}} \right)^2 \right]^{-1} \right\}. \quad (4)$$

The formula (4) describes well films resistivity size dependence in an initial area of film thickness, at which the deviation from dependency (3) predicted by the theory [4, 5] is observed. In Figure 3, the depend-

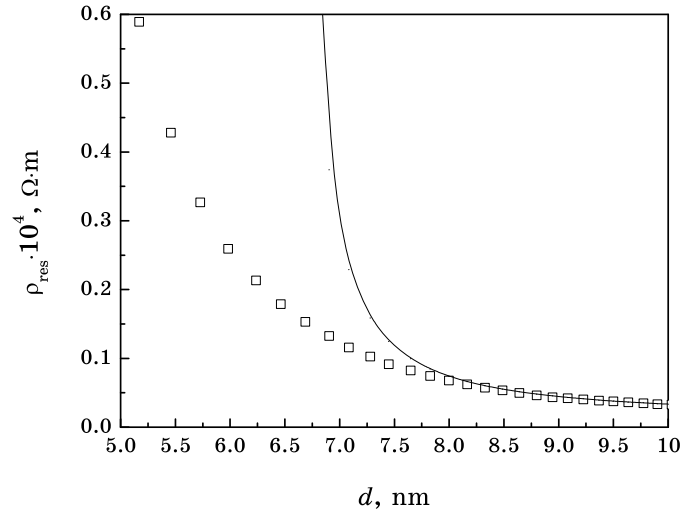


Fig. 3. Dependences of $\rho_{\text{res}} = \rho_{\text{res}}(d)$ as-deposited copper film ($T = 78$ K). Points—experimental data; continuous curve—approximated data of Namba theory expression at $h = 6.5$ nm.

ence $\rho(\bar{d})$ of copper films during metal condensation on polished glass substrate cooled to $T = 78$ K under the ultrahigh vacuum conditions ($p \leq 10^{-7}$ Pa) is shown. The comparison between the experimental points fixed by computer during film deposition and the theoretical curve calculated from (4) under the condition $h = 5.4$ nm shows that expression (4) well describes the dependence $\rho = \rho(\bar{d})$ for the films thickness of which exceeds 8 nm. It should be noted that d is film mass thickness here (and below). The experimental data deviation from theoretical curve is caused by transition to quantum charge transport. All the researches were carried out on electrically continuous films ($\beta > 0$). In particular, for as deposited films, β was measured for the temperature range from 78 K (liquid nitrogen) to 90 K (liquid oxygen).

In the quantum electron transport, the conductivity size dependencies σ_{res} differ slightly. The theoretical expressions obtained by Fishman and Calecki [23, 24] are the most convenient for direct comparison with experimental data:

$$\sigma_{\text{res}} \sim d^2 \left\{ 1 - \frac{6}{(3n\pi^5)^{2/3}} \frac{1}{d} \right\}, \quad (5)$$

where n is the current carriers concentration, d —film thickness. This expression may be transformed to $\sigma_{\text{res}} \sim d^\alpha$, where α changes from 2.1 (pure metals) to 6 (semiconductors). The power dependence of metal film residual conductivity on the film thickness was obtained also by Trivedi and Ashcroft [23]: $\sigma_{\text{res}} \sim d^2$. As noted above, the expressions of

theories [23, 24] were successfully applied for experimental size dependences description in many works.

The techniques for preparation of ultrathin electrically continuous copper, gold, silver, and palladium films were developed in [45–50]. The experiment was carried out under ultrahigh vacuum conditions (the pressure of residual gases $p \leq 10^{-7}$ Pa, the pressure of active components was less than 10^{-9} Pa) in evacuated glass devices. To overcome the influence of metal-condensates coagulation on the glass substrate thermally degasified for a long time (about 40 hours at $t = 400^\circ\text{C}$ in vacuum not worse than $p = 10^{-5}$ Pa), the surfactant (germanium, silicon, and antimony) underlayers of the thickness of some atomic layers were predeposited on surface directly before metal films deposition. The deposition techniques for metal layers the crystal grain sizes D of which did not depend on the film thickness and thickness did not exceed 50–60 nm were developed. Metal films and surfactants were deposited on a cooled substrate ($T = 78$ K) with condensation speed not exceeding 0.01 nm/s. The film thermostabilization was carried out with low-temperature annealing at $T \leq 373$ K. Application of this technique with change of thickness surfactant underlayers predeposited on the substrate made possible to prepare the metal films with presubscribed linear crystalline sizes on the parallel plane substrate. These facts were confirmed by experimental results of electron-microscopy and electron-diffraction studies of metal films and scanning tunnelling microscopy of surface topology investigation of palladium film.

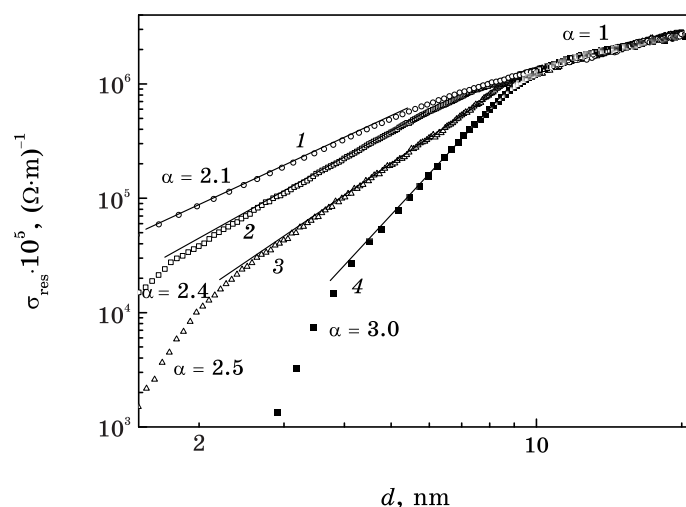


Fig. 4. Size dependences of gold films residual conductivity $\rho_{\text{res}} = \rho_{\text{res}}(d)$ as-deposited on germanium surfactant underlayers with thickness of 3 nm (1), 2 nm (2), 1 nm (3) and deposited on clean glass surface. Points—experimental data; curve segments—linear approximation.

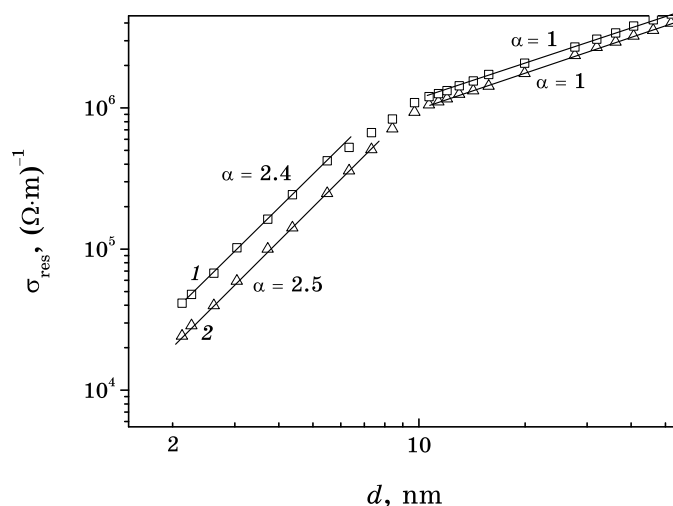


Fig. 5. Size dependences of palladium films residual conductivity $\rho_{\text{res}} = \rho_{\text{res}}(d)$ as-deposited on Al surfactant underlayers with mass thickness 0.3 nm (1) and clean glass surface (2). Points—experimental data; curve segments—linear approximation.

The investigation of film conductivity during film deposition in a continuous mode and fixed thickness films annealed at $T_{\text{an}} = 293$ K or $T_{\text{an}} = 373$ K were performed. The application of surfactant underlayers made possible to decrease considerably the critical metal film thickness corresponding to the transition to electrically continuous metal layers ($\beta > 0$). The metal films with stable (at temperatures not exceeding 300 K) and reproduced electrical properties with thickness $d \geq 2$ –3 nm were obtained. The size dependences of residual conductivity in metal films in the thickness range 3–8 nm are well described using expression (5) of the theory [24]. Those results are confirmed by the data in Figs. 4 and 5 obtained for as-deposited palladium films on Ge underlayers deposited on glass substrates and on Al underlayers of subatomic thickness ~ 0.3 nm.

The analysis of experimental results in Figs. 4 and 5 shows that in the range of large thickness $d > 12$ –15 nm the size dependences of residual conductivity σ_{res} may be explained within the framework of classical and internal size effects. In transitive area of thickness $8 < d < 12$ nm, behaviour of dependence of $\sigma_{\text{res}} = \sigma_{\text{res}}(d)$ can be explained by theories [11, 12]. If $d < 8$ nm, the film peculiarities of quantum transport display the behaviour $\sigma_{\text{res}} \sim d^\alpha$. In the films deposited on surfactants underlayers, the σ_{res} dependence can be observed up to the thickness of 3–4 nm. For the metal films as-grown on pure glass substrate, the quantum electron-transport behaviour is narrow and its lower limit reaches only 5–7 nm. The behaviour deviation of $\sigma_{\text{res}} \sim d^\alpha$ in the smaller thickness region is caused by both gradual transition to island structural state of metal lay-

ers, and chemical potential change with the film thickness d that, in turn, leads to the deviation from the approach taken in theories [23, 24].

From the data considered above, it is also clear that on σ_{res} size dependences any oscillations are absent. Measured oscillation is the consequence of the interference of electron wave reflected by film surface. For fine-grained layers, the coherent electron wave reflexion is hardly probable. This fact was widely discussed in theoretical and experimental works.

The issue concerning the behaviour of σ_{res} average thickness dependence is of significance in polycrystalline film research. It should be also noted that investigation of surfactant underlayer effect on the formation of ultrathin conductive films remains a problem of importance. As known from literature, this problem is currently central for purposeful techniques development for formation of conductive layers with prespecified structure and electric properties.

4. CONCLUSIONS

The problem of ultrathin metal film formation and study of their electrical properties have been analysed. These films may be used in modern micro- and nanoelectronics. The use of surfactant underlayers of subatomic thickness allows the control of the processes of formation and growth of metal film on the surface of dielectric substrates. Electrically stable metal films under low temperature condition ($T \leq 370$ K) with different mean linear crystal sizes on surface of dielectric substrates can be formed by the way of supervised change of surfactant underlayers parameters.

The possibility of harnessing of classical and quantum size effects modern theoretical models for analysis of experimental data results of charge transport phenomena in continuous metal film (2–3 nm–100 nm) is discussed. The modern theories of quantum size effect are used to explain the film conductivity size dependence when the film electrochemical potential is close to a similar parameter of bulk material. The possibility of using different theoretical models for explanation of experimental results of transport phenomena research in metal films in a wide range of film thickness is confirmed by analysis of modern modelling construction concepts on electron-charge transport in thin metal layers.

The possibility of control of structure and electric properties of metal films is supported by the example of experimental results discussion for some metal films deposited on surfactant underlayer.

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