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# The Size Effect on the Temperature Dependence of the Resistivity of Metal Films

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The analysis of the temperature dependences of the electrical conductivity of thin metal films is performed. It is shown that the size dependence of the effective parameter of the electron-phonon interaction is connected with amplification of high-temperature electron-phonon interaction at the decrease in the film thickness which appears as a result of the shift of the phonon spectrum to higher frequencies. We have found that the slope of the experimental temperature dependences of the resistance for Pd, Pt, and Sc films, as in the case of Mo, Cr, Ag, Au, and PdAu films, according to the data of other authors, increases with the decrease in the thickness. As the film thickness decreases the average phonon energy increases that leads to the increase in the efficiency of the electron-phonon scattering and, finally, to the increase in the resistivity.

**Keywords:** Thin films, Temperature dependence of resistivity, Electron-phonon interaction, Debye temperature, Phonon spectrum.

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## 1. INTRODUCTION

Modern development of microelectronics and sensor technology allows to obtain film materials of nanometer thickness with the properties which are not typical for the given materials in a bulk state. It is experimentally established that transition from the bulk to the film material leads to the changes of its physical properties, the root causes of which are connected with

– different structural characteristics;

– the influence of the thickness or crystallite sizes, which is manifested in the size effects appearing as a result of the restriction of the mean free path of electric current carriers by the external film surfaces or crystallite geometry;

– the change in the frequency and energy characteristics of the crystal lattice atoms under the action of thermal and mechanical factors.

A question about the features of the temperature dependences of the resistivity $\rho(T)$ of film materials, connected with the change in the scattering mechanisms, remains current, since it is very important for the prediction of the working characteristics of sensitive elements of microelectronic devises. A number of works is devoted to the theoretical and experimental aspects of this question.

The author of the work [1] has performed the theoretical investigations of the role of the electron-phonon scattering in the resistivity of metal nanowires which were modeled as two-dimensional plane lattices of the length $N{\cdot}a$ and width $M{\cdot}a$ (Fig. 1). If consider $N$ to be an infinitely large value ($N \to \infty$), then the value of $M$ (a number of monolayers) will be limited by the value which is not more than some hundreds.

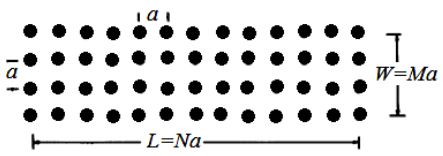


**Fig. 1** – Two-dimensional lattice of the size of $Na \times Ma$ which shows the nanowire structure. $a$ is the lattice parameter; $L$ and $W$ are the nanowire length and width, respectively. From [1]

Correlation for the phonon spectrum of monatomic lattice which takes into account the deformation potential using the Kubo formula was written in [1] as:

$$E_{k_1,k_2} = 2t_1 \cos(k_1 a) + 2t_2 \cos(k_2 a) - E_F ,$$

where $t_1$ and $t_2$ are the matrix parameters of the electron-phonon interaction along the length and width of the sample, respectively; $k_1$ and $k_2$ are the electron momenta in the directions along and across the nanowire.

In spite of the fact that parameters $k_1$ and $k_2$ in the equation are formally much alike, there is a qualitative difference between them [1]. Indeed, parameter $k_1$ is constantly changed from $-\pi/a$ to $+\pi/a$ with the step $\Delta k_1 = 2\pi/Na$, which tends to 0 at $N \to \infty$. On the other hand, parameter $k_2$ is discretely changed with the step $\Delta k_2 = 2\pi/Ma$ and is equal to $k_2 = 0.15$ nm$^{-1}$, for example, for $M = 100$ and $a = 0.4$ nm.

Dependence of the resistance on the nanowire width, temperature and Fermi energy was investigated by numerical calculations using typical values of different parameters. It is established [1] that nanowires resistance increases with decrease in their width (film thickness analog) and increasing temperature (Fig. 2), at that temperature dependence of the resistivity has the most linear behavior for nanowires of the thickness of $M = 20$ that is explained by the increase in the contribution of electron-phonon interaction with decreasing nanowire width.

The authors of the works [2-4] have carried out the experimental investigations of the temperature dependence of the resistivity of Ag [2, 4] and Au [3] films in the low-temperature range (to 300 K) and Au films with different concentration of Co clusters [5] and concluded that decrease in the Debye temperature in small samples is caused by softening of the surface phonons spectrum compared with their volume spectrum. In Fig. 3 we show the temperature dependences of the resistivity for epitaxial Au films [3] of different thickness which imply the dependence of the slope of $\rho(T)$ on the thickness of film material. A series of publications [6-8] was devoted to the question of the influence of internal and external size effects on the temperature dependence of electrical conductivity of transition-metal thin films.

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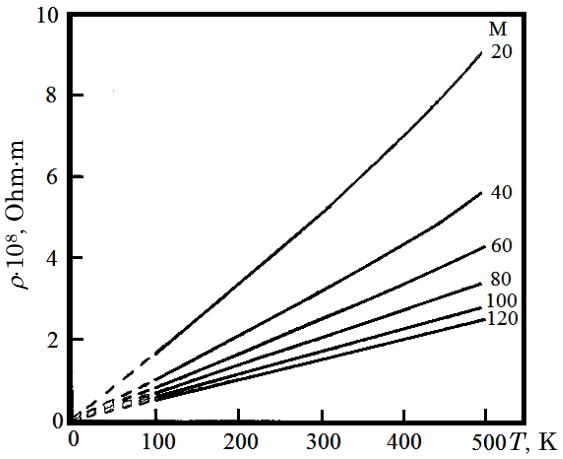


**Fig. 2** – Temperature dependences of the resistivity at $M$ = 20-100 calculated by two-dimensional model. From [1]

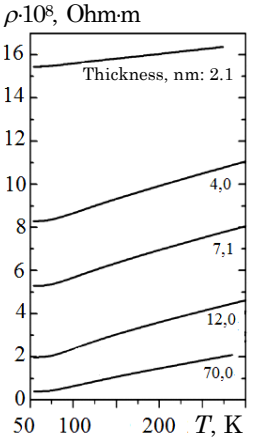


**Fig. 3** – Temperature dependences of the resistivity for Au films [3] of different thickness on sapphire ($d$ = 2-12 nm) and mica ($d$ = 70 nm) substrates

The main result of the above mentioned publications consists in the fact that angular coefficient tg$\alpha$ of the linear section of the dependence $\rho(T)$ depends on the film thickness. According to the cited works, the slope of the temperature dependence of the resistivity increases with decreasing nanowire width [1] or film thickness [2-8]. In accordance with the authors of [7], a weak dependence of tg$\alpha$ on the thickness for Cr and Mo films is explained by the fact that during $s$-$d$ scattering there is a partial localization of $s$-electrons which leads to the increase in their effective mass and gives a certain contribution to the hole conductivity of these metals.

We note that the same decrease in tg$\alpha$ with increasing thickness was also observed in PdAu (this intermetallic is formed on the basis of multilayer Au/Pd films) [9] and Pt [10] films.

All aforesaid has caused the relevance of the given work, whose aim consisted in the analysis of the temperature dependences of the resistivity of monolayer metal films depending on their thickness and influence of the phonon spectrum deformation and change of the Debye temperature ($\Theta_D$) on the efficiency of phonon scattering of $s$- and $d$-electrons that leads to the significant difference of the electrophysical properties between film and bulk materials.

## 2. DEPENDENCE OF THE TEMPERATURE DEPENDENCE OF THE FILM RESISTIVITY ON ITS THICKNESS

Condensation of Pd, Pt and Sc monolayer metal films was carried out in the HV chamber VUP-5M (vacuum ~ $10^{-3}$-$10^{-4}$ Pa) by the method of thermal evaporation. Thermal treatment of the samples was performed during three "heating ↔ cooling" cycles in the temperature range of 300-700 K. Calculation of the thermal coefficient $\beta$ was realized based on the third cooling cycle.

As known (see, for example, [13]), resistance of conductors appears as a result of scattering of conduction electrons and holes by electrons, phonons and defects of the crystal structure. Temperature dependence of the resistivity for bulk samples of transition $d$-metals (the Debye-Gruneizen theory) is expressed by the following correlation:

$$\rho_0(T) = \rho_{0r} + A\cdot T^2 + B\left(\frac{T}{\Theta_{0D}}\right)^3 \times \times j_3\left(\frac{\Theta_{0D}}{T}\right) + C\left(\frac{T}{\Theta_{0D}}\right)^5 \cdot j_5\left(\frac{\Theta_{0D}}{T}\right), \quad (1)$$

where $\rho_{0r}$ is the residual resistance connected with scattering of current carriers by lattice defects which does not depend on the temperature; $A$, $B$, $C$ are the coefficients of proportionality; $\Theta_{0D}$ is the Debye temperature for a bulk material;

$j_n(\Theta_{0D}/T) = \int_0^{\Theta_{0D}/T} \frac{x^n dx}{(e^x-1)\cdot(1-e^{-x})}$ is the Debye integral.

Let us characterize the physical meaning of the terms entering correlation (1). The term which is proportional to $T^2$ is connected with electron-phonon scattering and is manifested only at $T < 10$ K. The terms proportional to $T^3$ and $T^5$, respectively, are conditioned by scattering of $s$-electron by phonon to $d$- or $s$-zone.

In the case of metal films correlation (1) is transformed to the form of [6]

$$\rho(T) = \rho_з + a\cdot T^2 + c^*\cdot\left(\frac{T}{\Theta_D}\right)^3 \cdot j_3\left(\frac{\Theta_D}{T}\right), \quad (1')$$

where $a$, $b$, $c$ are the coefficients similar to $A$, $B$, $C$; $\Theta_D$ is the Debye temperature for the film; $c^* = b + c$ is the effective parameter of the electron-phonon interaction.

Parameter $c^*$ is determined by the formula

$$c^* = tg\alpha \frac{\Theta_D^3}{T^2}\cdot\frac{1}{J_3\left(\frac{\Theta_D}{T}\right)},$$

where tg$\alpha$ is the slope of the linear section of $\rho(T)$.

It is taken into account in correlation (1) that in thin samples (foil, wires and films) the law $\rho_0 \sim T^5$ is transferred to the law $\rho \sim T^3$ at low temperatures, and this gives the possibility to combine the terms which are responsible for the Mott $s$-$d$- and $s$-$s$-scattering. Moreover, we should emphasize that Debye temperature in film materials differs from $\Theta_{0D}$.

As seen from Fig. 4, for Pd, Pt and Sc films the value of tg$\alpha$ increases with decreasing film thickness $d$.

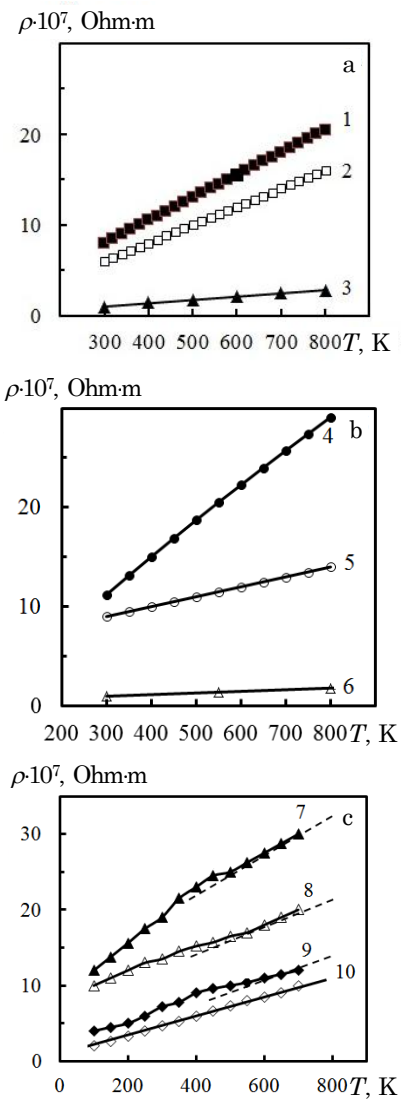


**Fig. 4** – Dependences $\rho(T)$ for Pd (a), Pt (b) and Sc (c) films. Thickness, nm: 20 (1); 35 (2); 10 (4); 50 (5); 54 (6); 50 (7); 60 (8); 190 (9) and for bulk samples [11, 12] (3, 6 and 10)

In whole, in accordance with our and obtained by other authors [2-10] data, the value of tg$\alpha$ for Pd, Pt, Sc, Mo, Cr, Ag, Au and PdAu films is larger than for bulk samples, at that the values of tg$\alpha$ and $c^*$ increase with decreasing film thickness.

It should also be borne in mind that correlation (1) describes the temperature dependence of the resistivity of non-ferromagnetic samples in a general case; therefore it does not take into account the features in the Curie and Néel points and at the Debye temperature.

Since electron-phonon scattering plays the key role in the resistivity of thin films, we will consider below the influence of the phonon spectrum and change of the Debye temperature on the efficiency of phonon scattering of *s*- and *d*-electrons which induces the size effect in thermoresistive properties of thin films.

## 3. DEFORMATION OF THE PHONON SPECTRUM

A detailed study of the oscillations of microscopic objects (atoms, ions) in the crystal lattice requires the application of concepts and methods of quantum theory, according to which any oscillations or waves are simultaneously particles and any particles – waves. Phonons as quasi-particles with zero magnetic moment and spin which correspond to elastic oscillations of the crystal lattice are characterized by the quasi-momentum (defines the propagation direction and wavelength) and oscillation frequency. In the case of acoustic phonons, unit cell is shifted as a whole during oscillations. Frequency distribution of normal modes, i.e. the phonon spectrum, is the key question of the crystal lattice oscillation theory, and the phonon state density $g(\omega)$, with which the Debye temperature $\theta_D = \hbar\omega_{\max}/k_B$ is connected, is an important characteristic of the crystal lattice state. Since change of the phonon spectral density in thin metal films is connected with the deformation of the phonon spectrum induced by a finite size of the film thickness, we will consider its features.

Distribution function has the following form (see, for example, [14]):

$$g(\omega) = \frac{dg(q)}{dq}\frac{dq}{d\omega}, \quad q \equiv \frac{2\pi}{\lambda} = \frac{2\pi\nu}{V_{ph}} = \frac{\varpi}{V_{ph}},$$

where $V_{ph}$ is the phase velocity; $q$ is the wavenumber.

For small samples one can write [14]

$$v_2^{-2} = \frac{2v_l^4 - 3v_t^2v_l^2 + 3v_l^4}{v_t^2v_l^2\left(v_l^2 - v_t^2\right)},$$

$$g(\omega) \cong \frac{V\omega^2}{2\pi v_3^3} + \frac{S\omega}{8\pi v_2^2} \cong \frac{V\omega^2}{2\pi v_3^3},$$

where $S$ and $V$ are the surface area and volume of the sample, respectively.

Frequency dependence of the phonon spectrum density is illustrated in Fig. 5. The presented correlations indicate that acoustic spectrum in small samples is softened from the side of $\omega_{\max}$ that is connected with the contribution to the phonon spectrum of Rayleigh waves (renormalization of the phonon spectrum in small samples). Along with this, phonon spectrum becomes more rigid in the low-frequency region, i.e. near $\omega_{\min}$, since $\lambda^z_{\max} = 2d$, and $\omega^z_{\min} = 2\pi V^z_{ph} / \lambda^z_{\max} = \frac{\pi V_{ph}}{d}$. Thus, deformation of the phonon spectrum leads to the decrease in $\Theta_D$ with decreasing film thickness, but as a result of the competition of these two mechanisms the resulting amplification of the electron-phonon interaction takes place that leads to the increase in tg$\alpha$ with decreasing film thickness.

At that, one or another feature of the phonon spectrum can be exhibited in a specific phenomenon. But, if degree of softening is sharply attenuated with increasing number of atoms, then effect in the low-frequency region remains.

Thus, size dependence of the effective parameter of the electron-phonon interaction is connected with the fact that branch of the phonon spectrum in the direction perpendicular to the film plane is limited below by the boundary values of the wavenumber that leads to the size effect of $c^*$, which is determined not by the film structure, but by the rigidity of one of the branches of

the phonon spectrum. Increase in the parameter $c^*$ with decreasing thickness is conditioned by strengthening of the high-temperature electron-phonon interaction which is exhibited due to the shift of the phonon spectrum from the low-frequency region to the region of higher frequencies. Grain boundary scattering, which, in contrast to the surface one, is mainly manifested at relatively large film thicknesses, can also influence the value of $c^*$.

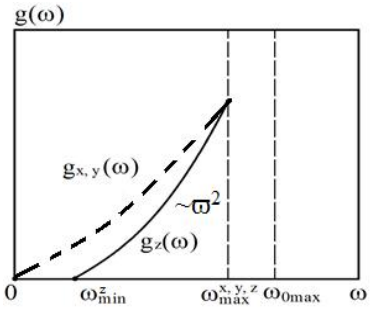


**Fig. 5** – Qualitative frequency dependence of the phonon spectrum density

At the end of the work, it should be also noted that Mott scattering of electrons by phonons, which plays an important role in electrophysical properties, in a slightly different form is manifested in the spin-dependent electron scattering by magnetic moments, when properties of the films are determined by the exchange processes between carriers of the opposite spin polarization (scattering with a spin flip) and scattering time of electrons by phonons, impurities, etc. Two-current model [15] of the conductivity in ferromagnetic films is based on the condition about interaction of electron currents with the directions "spin up" and "spin down" by collisions with the momentum exchange. Phenomenon of mixing of spin states occurs due to the processes of spin flips and, mainly, as a result of scattering by magnons, which, increasing with temperature, partly equalizes the currents of "spin up" and "spin down" channels higher than the room temperature for the majority of ferromagnets. For today, two-current model is the basic model in spintronics, although it is slightly simplified, since does not take into account the mixing of spin states.

## 4. CONCLUSIONS

1. Size dependence of the effective parameter of the electron-phonon interaction is connected with amplification of high-temperature electron-phonon interaction with decreasing film thickness, which is manifested due to the shift of the phonon spectrum from the low-frequency region to the region of higher frequencies.

2. Slope of the experimental temperature dependences of the resistance for Pd, Pt and Sc films, as well as in the case of Mo, Cr, Ag, Au and PdAu films, according to the data of other authors, increases with decreasing their thickness.

3. Phonon spectrum of thin films is more energy compared with bulk samples. With decreasing film thickness, phonon energy in a certain direction increases and, as a consequence, efficiency of electron scattering by phonons increases and, correspondingly, sensitivity of the resistivity to temperature grows.

4. Deformation of the phonon spectrum in film materials occurs from the point of view of both softening and strengthening of its rigidity. These processes takes place simultaneously and do not exclude each other.

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