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EFFECT OF NONPARABOLICITY DESCRIBED BY THE FIVAZ MODEL ON THE ELECTRICAL RESISTANCE OF THERMOELECTRIC MATERIAL-METAL CONTACT

The temperature dependences of thermoelectric material-metal electrical contact resistance were investigated in the case when a band spectrum of free charge carriers in material is described by the Fivaz model. A transient contact layer formed by the deviation of the surface of superlattice semiconductor thermoelectric material (SL TEM) from the ideal plane and transient contact layers with and without clusters formed in the process of steady-state diffusion of metal particles in SL TEM were considered. It was established that contact resistance drastically decreases with increase in the degree of nonparabolicity of SL TEM band spectrum, which is determined as the ratio of the Fermi energy of ideal two-dimensional electron (hole) gas with a quadratic dispersion law to the miniband width describing translation motion of charge carriers in the direction perpendicular to the plane of layers. This decrease is explained by blocking of free carrier scattering in the direction perpendicular to the plane of layers. It is shown that in the range of degrees of nonparabolicity K from 0.1 to 10, transient layer thicknesses from 20 to 150 μm, dimensionless intensities of metal atoms entering the volume of transient layer A from 0 to 1 and temperatures from 200 to 400 K, the electrical contact resistance of transient layer due to the deviation of SL TEM surface from the ideal plane varies from $8 \cdot 10^{-9}$ to $1.9 \cdot 10^{-7} Ohm \cdot cm^2$, transient layer due to steady-state diffusion of metal into SL TEM without formation of clusters – from $8 \cdot 10^{-9}$ to $4 \cdot 10^{-7} Ohm \cdot cm^2$, transient layer due to steady-state diffusion of metal in SL TEM with formation of clusters – from $8 \cdot 10^{-9}$ to $4.5 \cdot 10^{-7} Ohm \cdot cm^2$.

Key words: Fivaz model, superlattice, Fermi energy, miniband, degree of nonparabolicity, thermoelectric material–metal contact, electrical contact resistance of transient layer, deviation of thermoelectric material surface from the ideal plane, steady-state diffusion, intensity of metal particles entering semiconductor, clusters.

Introduction

The thermoelectric material (TEM)–metal electrical contact resistance, all other conditions being equal, essentially depends on the resistivities of metal and TEM. In turn, the resistivity of TEM depends not only on the concentration and scattering mechanisms of free charge carriers in it, but also on the nature of the TEM band structure, because the mobility of free charge carriers depends not least on it.

Layered TEM, which, in particular, include bismuth telluride and alloys on its basis, are more or less prone to the formation of superlattices. In turn, thermoelectric converters from these materials are usually made so that the planes of the contact electrodes are perpendicular to the

planes of layers. Therefore, the TEM-metal electrical contact resistance in this case depends essentially on the resistivity of TEM in the plane of layers. But it is known that the formation of a superlattice, that is, a gradual transformation of a material with a three-dimensional parabolic band spectrum into a material with a quasi-two-dimensional substantially non-parabolic band spectrum, reduces the TEM resistivity in the plane of layers. The study of the influence of the degree of quasi-two-dimensional TEM with a superlattice (SL TEM) on the electrical resistance of TEM-metal electrical contact resistance under different conditions is the purpose of this article.

The resistivity of TEM described by the Fivaz model

The energy spectrum of charge carriers in SL TEM is rather often described by the Fivaz model [1]. Within this model, the motion of electrons and holes along the layers is described by the effective mass approximation, and across – by the tight-binding method. It can be presented as follows:

$$\varepsilon\left(k_x, k_y, k_z\right) = \frac{\hbar^2}{2m^*}\left(k_x^2 + k_y^2\right) + \Delta\left(1 - \cos ak_z\right), \tag{1}$$

where k_x, k_y, k_z – the quasi-momentum components of electron (hole), m^* – the effective mass of electron (hole) in the plane of layers, Δ – the half-width of the mini-band that describes the motion of electrons (holes) in the direction perpendicular to the layers, a – the distance between the translation equivalent layers.

Therefore, the electrical resistivity of "superlattice" thermoelectric material (SL TEM) in the plane of layers as determined as follows [2]:

$$\sigma_s = \sigma_{0l} \int\limits_0^\infty \int\limits_0^\pi \frac{y \exp\left\{\left[y + K^{-1}(1 - \cos x) - \gamma^*\right]/t_{2D}\right\}}{\left\{\exp\left\{\left[y + K^{-1}(1 - \cos x) - \gamma^*\right]/t_{2D}\right\} + 1\right\}^2 \sqrt{2y + 4\pi K^{-2} n_0 a^3 \sin^2 x}} dx dy, \tag{2}$$

where $\sigma_{0l} = 8\pi^{5/2} e^2 l \sqrt{n_0 a}/(aht_{2D})$, l – mean free path of electrons (holes), n_0 – concentration of electrons (holes), $t_{2D} = kT/\zeta_{02D}$, $\zeta_{02D} = h^2 n_0 a/4\pi m^*$ – the Fermi energy of an ideal two-dimensional Fermi-gas with a quadratic law of dispersion at the absolute zero temperature, $K = \zeta_{02D}/\Delta$, $\gamma^* = \zeta/\zeta_{02D}$, ζ – chemical potential of electron (hole) gas in SL TEM. Parameter K characterizes the degree of quasi-dimensionality of SL TEM, or, in other words, the degree of openness of its electron (hole) Fermi surface.

Chemical potential is determined from the equation:

$$\frac{t_{2D}}{\pi} \int\limits_0^\pi \ln\left[1 + \exp\left(\frac{\gamma^* - K^{-1}(1 - \cos x)}{t_{2D}}\right)\right] dx - 1 = 0. \tag{3}$$

It is believed that scattering of charge carriers mainly occurs on the deformation potential of acoustic phonons, and, hence, the mean free path of charge carriers is inversely proportional to temperature and does not depend on the energy of charge carriers. To calculate the temperature

dependences of SL TEM resistivity at different values of the degree of nonparabolicity K, the following parameters of SL TEM were taken: $n_0 = 3 \cdot 10^{19}\,\text{cm}^{-3}$, $a = 3\,\text{nm}$, $m^* = m_0$, $T_0 = 300\,\text{K}$, $l_0 = 20\,\text{nm}$.

Based on these dependences, the temperature dependences of the SL TEM-metal electrical contact resistance were calculated for two cases: when transient contact layer is due to the deviation of the SL TEM surface from the ideal plane and when it is due to steady-state diffusion of the metal in SL TEM without the formation of intermetallic compounds.

SL TEM-metal electrical contact resistance due to a deviation of SL TEM surface from the ideal plane

The calculations were performed on the assumption that "hollows" and "humps", caused by the deviation of SL TEM surface from the ideal plane, are distributed evenly over it. Therefore, for the thickness h of the damaged layer which is recognized at the vertical distance between the deepest "hollow" and the highest "hump", the TEM-metal electrical contact resistance caused by this deviation was determined through the resistivities of semiconductor ρ_s and metal ρ_m as follows [3]:

$$r_{ce} \equiv r_c = \frac{h(\rho_s - \rho_m)}{\ln(\rho_s/\rho_m)},$$
(4)

and the resistivity of metal was considered to be directly proportional to temperature. Nickel was taken as the metal whose resistivity at 300 K is $8 \cdot 10^{-6}$ Ohm·cm.

The results of calculations of the temperature dependences of thermoelectric material-metal contact resistance caused by the deviation of TEM surface from the ideal plane are shown in Figs. 1 and 2.
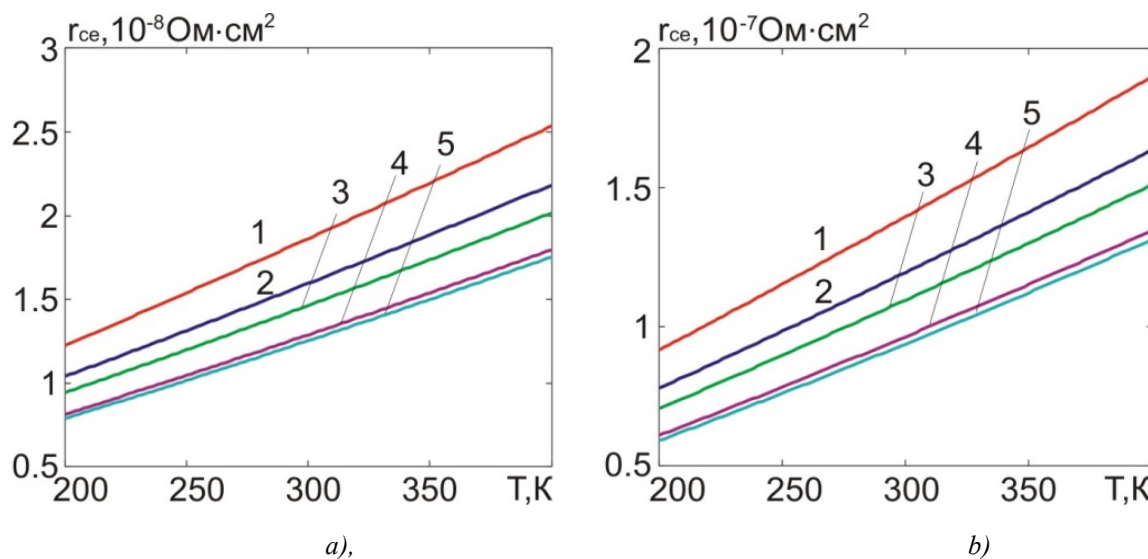

a),   *b)*

Fig.1.Temperature dependences of TEM-metal electrical contact resistance at damaged layer thickness: a) h=20 µm, п) h=150 µm. 1 – K=0.1; 2 – K=0.5; 3 – K=1; 4 – K=5; 5 – K=10.

It can be seen from the figures that with an increase in the degree of nonparabolicity of the band spectrum of free charge carriers, and, consequently, in the degree of openness of the Fermi surface (FS) of TEM, the TEM-metal electrical contact resistance, due to the deviation of TEM surface from the ideal plane, decreases significantly, and with a rise in temperature grows in conformity with the temperature dependences of the resistivities of both metal and SL TEM. In the whole temperature range of $200 - 400$ K at the degree of openness $0.1 \le K \le 10$ the specific electrical resistance of TEM – metal contact varies from $8 \cdot 10^{-9}$ to $2.5 \cdot 10^{-8}$ Ohm·cm^2 at the thickness of damaged layer 20 μm, and from $6 \cdot 10^{-8}$ to $1.9 \cdot 10^{-7}$ Ohm·cm^2 at the thickness of damaged layer 150 μm, which is not only in qualitative, but also in quantitative agreement with the experimental data. The larger contact resistances observed by the authors can be explained by the lower degree of nonparabolicity of the band spectrum of bismuth telluride-based alloys used for the manufacture of thermoelectric legs.

SL TEM-metal electrical contact resistance due to steady-state diffusion of metal in SL TEM without formation of clusters

If transient contact layer is formed in the process of steady-state diffusion of metal in SL TEM, then the distribution of metal atoms over the depth of this layer is determined as [4]:

$$n(x) = n_0 \left[1 - (1 - A)x - Ax^2 \right],$$ (5)

where x – depth normalized to layer thickness, n_0 – concentration of metal atoms close to metal-transient layer boundary, A – dimensionless parameter which characterizes the mode of contact creation as is determined as:

$$A = Qh^2 / 2Dn_0 ,$$ (6)

where Q – the intensity of metal entering TEM, D – coefficient of metal diffusion in TEM. From formula (5) follows the distribution of the relative volumetric fraction of metal in the transient layer:

$$v(x) = \frac{(A_m / \gamma_m) \left[1 - (1 - A)x - Ax^2 \right]}{(A_m / \gamma_m) \left[1 - (1 - A)x - Ax^2 \right] + (A_s / \gamma_s) \left[(1 - A)x + Ax^2 \right]},$$ (7)

where A_m, A_s, γ_m, γ_s – atomic (molecular) masses and densities of metal and TEM, respectively. So, if there are no clusters in transient layer, the dependence of its electrical conductivity on the depth is determined as:

$$\sigma_l(x) = \sigma_s + (\sigma_m - \sigma_s)v(x),$$ (8)

and, thus, the SL TEM-metal electrical contact resistance with the uneven distribution of metal atoms in transient layer is determined as:

$$r_{ce} = h \int_0^1 \frac{dx}{\sigma_l(x)}.$$

(9)

If, however, the distribution of metal atoms in transient layer becomes uniform, for instance, due to annealing of contact structure, its electrical conductivity is determined as:

$$\sigma_0 = \sigma_s + (\sigma_m - \sigma_s) v_0,$$

(10)

where

$$v_0 = \int_0^1 \frac{(A_m/\gamma_m)\left[1 - (1-A)x - Ax^2\right]}{(A_m/\gamma_m)\left[1 - (1-A)x - Ax^2\right] + (A_s/\gamma_s)\left[(1-A)x + Ax^2\right]}.$$

(11)

Hence, in this case

$$r_{ce} = h/\sigma_0.$$

(12)

The results of calculation of the electrical contact resistance of transient layer without clusters are shown in Figs.2 – 5.
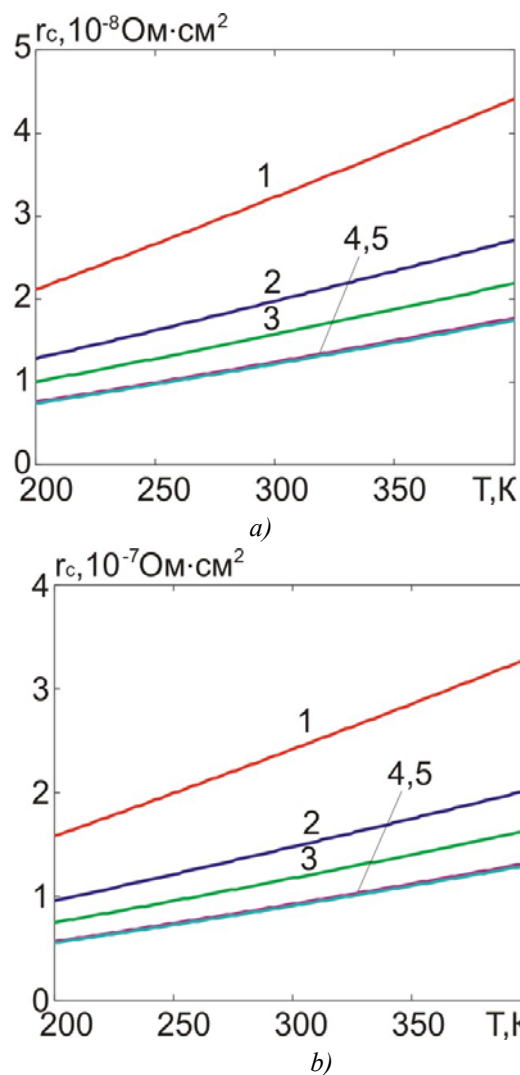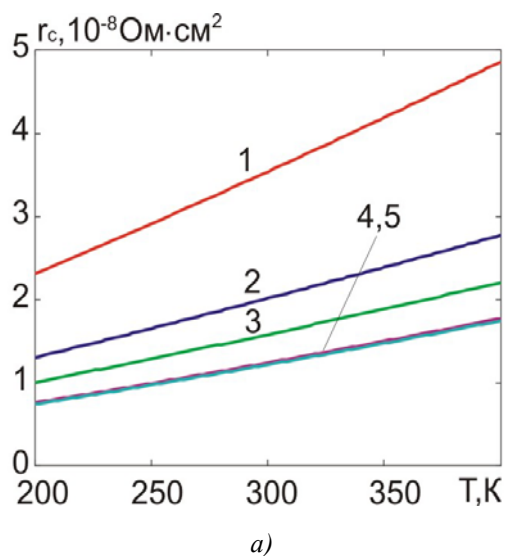

a)

b)
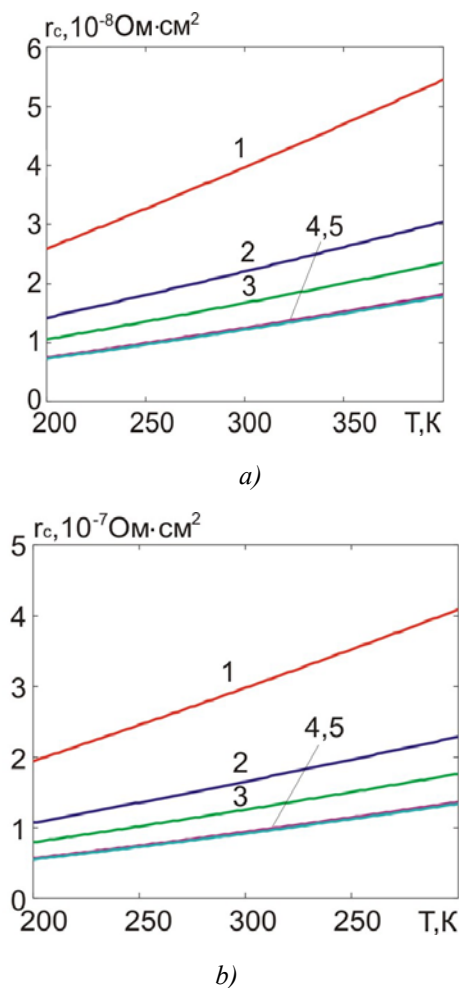
Fig.2. Temperature dependences of TEM-metal electrical contact resistance at uneven distribution of metal particles in transient layer without clusters at the value of A=0: a) h=20 μm, b) h=150 μm. 1 – K=0.1; 2 – K=0.5; 3 – K=1; 4 – K=5; 5 – K=10.

Fig.3. Temperature dependences of TEM-metal electrical contact resistance at uneven
distribution of metal particles in transient layer without clusters at the value of A=1:
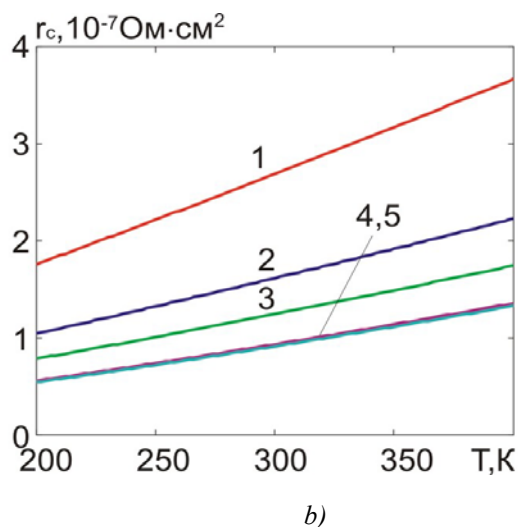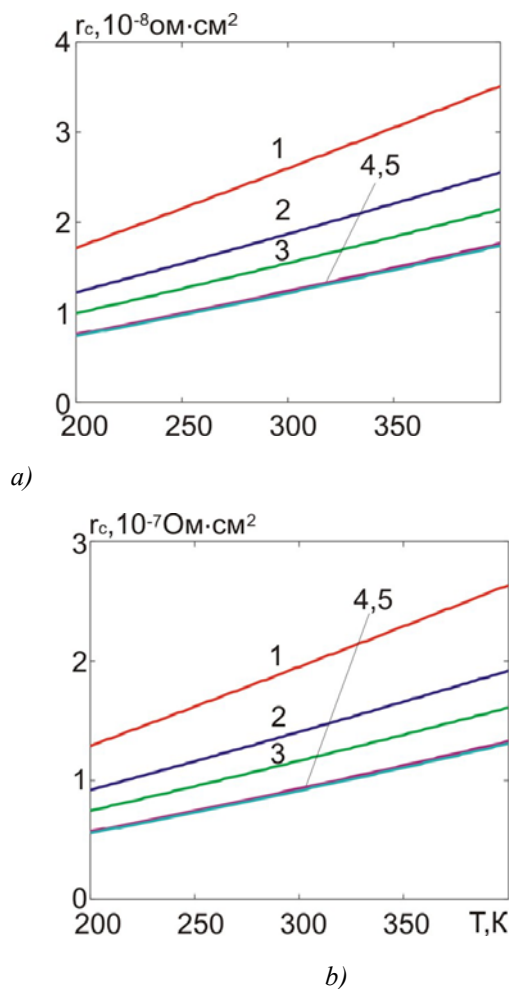*a) h=20 μm, b) h=150 μm. 1 – K=0.1; 2 – K=0.5; 3 – K=1; 4 – K=5; 5 – K=10.*

b)

*Fig.4. Temperature dependences of TEM-metal electrical contact resistance after levelling
the distribution of metal particles in transient layer without clusters at the value of
A=0: a) h=20 µm,b) h=150 µm.1 – K=0.1; 2 – K=0.5; 3 – K=1; 4 – K=5; 5 – K=10.*

a)

b)

*Fig. 5. Temperature dependences of TEM-metal electrical contact resistance after
levelling the distribution of metal particles in transient layer without clusters
at the value of A=1: a) h=20 µm, b) h=150 µm. 1 – K=0.1;
2 – K=0.5; 3 – K=1; 4 – K=5; 5 – K=10.*

It can be seen from the figures that the electrical contact resistance decreases with an increase in the degree of nonparabolicity of SL TEM band spectrum and the intensity of metal entering transient layer during its creation, and increases with a rise in temperature. Moreover, it also decreases after levelling the distribution of metal particles in transient layer. On the whole, in the considered range of degrees of nonparabolicity (openness of SL TEM FS), the intensities of metal particles entering transient layer, the thicknesses of contact layers and temperatures, the electrical contact resistance due to steady-state diffusion of metal particles without formation of clusters varies in the range of $8 \cdot 10^{-9}$ to $4 \cdot 10^{-7}$ Ohm·cm^2. This interval is broader than in the case when transient contact layer is formed due to the deviations of SL TEM surface from the ideal plane.

SL TEM-metal electrical contact resistance due to steady-state diffusion of metal in SL TEM with formation of clusters

Due to the large number of defects in layered SL TEM, clusters of atoms can form in the interlayer space. In this case, the conductivity calculation should be performed using percolation theory. In accordance with this theory, taking into account the depth dependence of the concentration of metal atoms in transient layer, the electrical conductivity of transient layer is determined as [5]:

$$\sigma_l(x) = 0.25\left\{\sigma_s[2 - 3v(x)] + \sigma_m[3v(x) - 1] + \sqrt{\{\sigma_s[2 - 3v(x)] + \sigma_m[3v(x) - 1]\}^2 + 8\sigma_m\sigma_s}\right\}, \quad (13)$$

and in the case when the distribution of atoms over the depth of transient layer becomes uniform, as:

$$\sigma_0 = 0.25\left\{\sigma_s(2 - 3v_0) + \sigma_m(3v_0 - 1) + \sqrt{[\sigma_s(2 - 3v_0) + \sigma_m(3v_0 - 1)]^2 + 8\sigma_m\sigma_s}\right\}. \quad (14)$$

Further, the calculation of the electrical contact resistance is performed in the same order as in the case of transient layer without clusters. The results of calculating the temperature dependences of the SL TEM-metal electrical contact resistance in the case of transient contact layer with clusters are shown in Figs. 6 - 9.

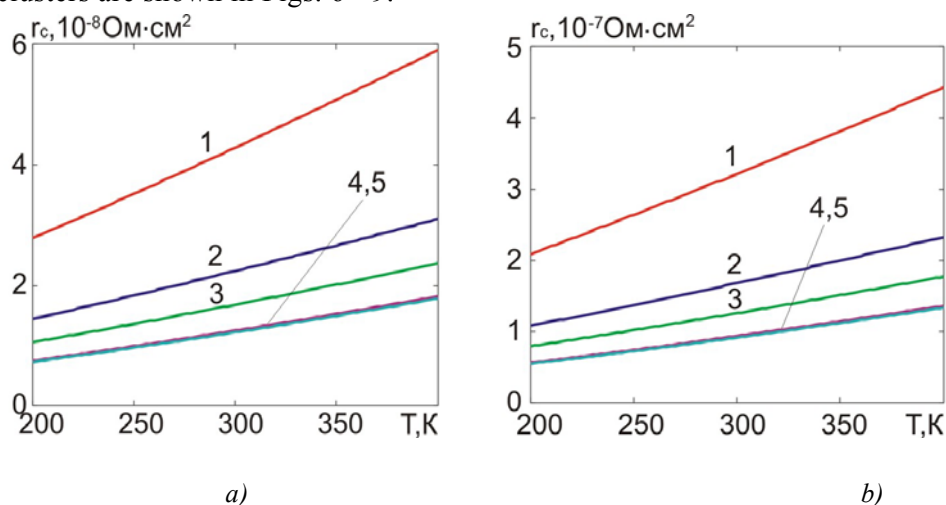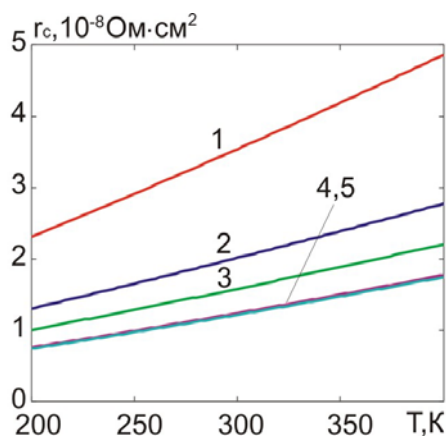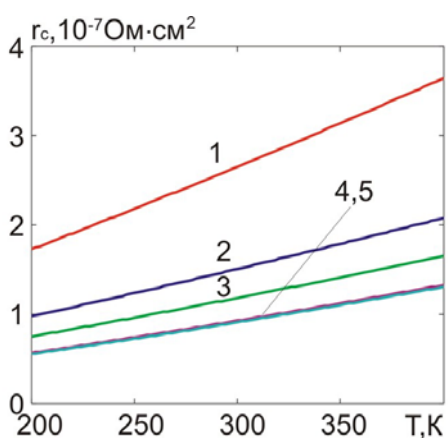*a)*   *b)*

Fig.6. Temperature dependences of TEM-metal electrical contact resistance at uneven distribution of metal particles in transient layer with clusters at the value of A=0: a) h=20 μm, b) h=150 μm. 1 – K=0.1; 2 – K=0.5; 3 – K=1; 4 – K=5; 5 – K=10.
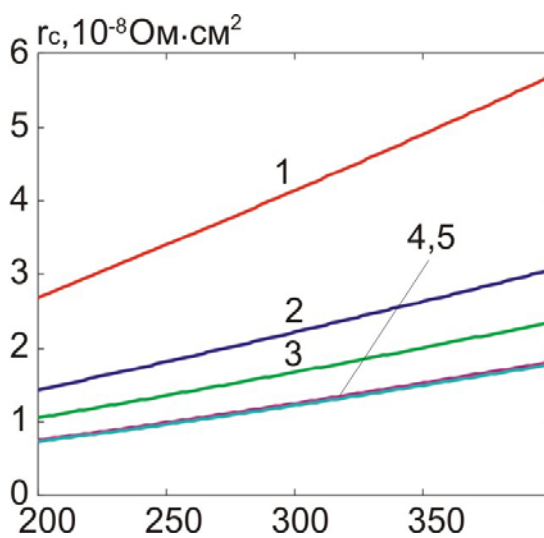
a)

b)

*Fig.7. Temperature dependences of TEM-metal electrical contact resistance at uneven
distribution of metal particles in transient layer with clusters at the value of
A=1: a) h=20 μm, b) h=150 μm. 1 – K=0.1; 2 – K=0.5;
3 – K=1; 4 – K=5; 5 – K=10.*

a)

b)

*Fig.8. Temperature dependences of TEM-metal specific electrical
contact resistance after levelling the distribution of metal particles in
transient layer with clusters at the value of
A=0: a) h=20 μm, b) h=150 μm.
1 – K=0.1; 2 – K=0.5; 3 – K=1;
4 – K=5; 5 – K=10.*

a)

b)

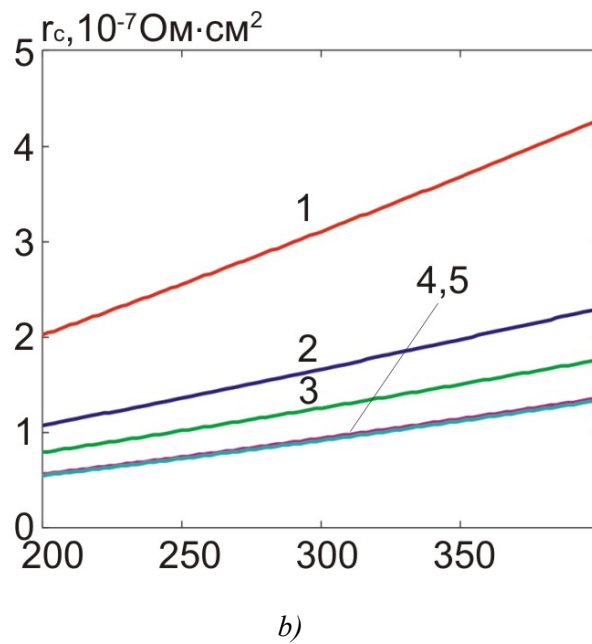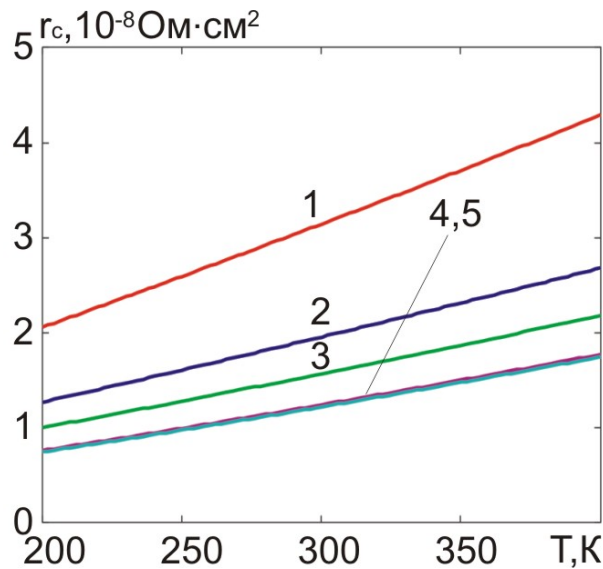*Puc.9.Temperature dependences of TEM-metal specific electrical contact resistance*
after levelling the distribution of metal particles in transient layer with clusters at
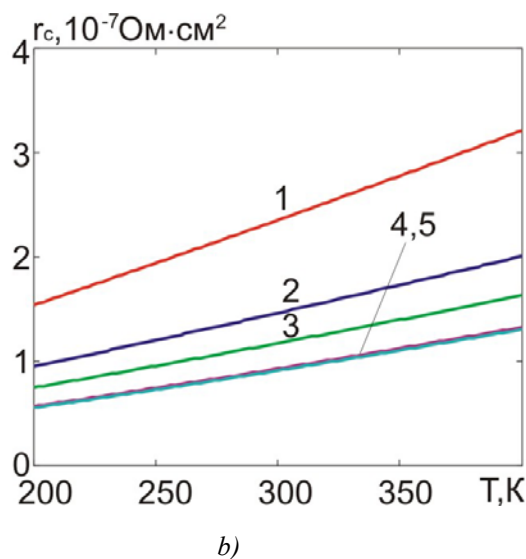*the value of A=1: a) h=20 μm, b) h=150 μm. 1 – K=0.1; 2 – K=0.5;*
3 – K=1; 4 – K=5; 5 – K=10.

From the figures it is seen that just as in the case of transient contact layer without clusters, the electrical contact resistance decreases with increase in the nonparabolicity degree of the band spectrum of SL TEM and the intensity of metal entering transient layer in the process of its creation, and increases with a rise in temperature. In so doing, it also decreases after levelling the distribution of metal particles in transient layer. On the whole, in the considered range of degrees of nonparabolicity (openness of SL TEM FS), the intensities of metal particles entering transient layer, the thicknesses of contact layers and temperatures, the electrical contact resistance due to steady-state diffusion of metal particles with formation of clusters varies in the range from $8 \cdot 10^{-9}$ to $4.5 \cdot 10^{-7}$ Ohm·cm^2. This range is somewhat broader than in the case when transient contact layer formed in the process of steady-state diffusion of metal atoms in SL TEM has no clusters.

Conclusions

1. It was established that, just as in the case of formation of SL TEM-metal transient contact layer due to the deviation of SL TEM surface from the ideal plane, in the cases of said transient contact layer formation due to steady-state diffusion of metal in SL TEM with or without formation of clusters, SL TEM-metal electrical contact resistance decreases essentially with increase in the degree of nonparabolicity of SL TEM band spectrum, or, which is the same, the degree of openness of SL TEM FS.
2. As in the case of TEM with a parabolic band spectrum, the TEM-metal electrical contact resistance decreases with increasing intensity of metal entering transient layer and as a result of subsequent levelling the distribution of metal atoms in transient layer, for instance, due to annealing.
3. At the thickness of transient layer 20 μm and the degree of nonparabolicity of SL TEM band spectrum $K=10$, which corresponds to strongly open SL TEM FS, the electrical contact resistance at a temperature of 200 K tends to the asymptotic value equal to $8 \cdot 10^{-9}$ Ohm·cm^2. This value can be considered as minimum for this temperature. Though the character of

temperature dependence of the electrical contact resistance even at such degree of nonparabolicity depends on contact creation conditions, at 400 K this contact resistance does not exceed $2 \cdot 10^{-8}$ Ohm·cm^2.

4. On the whole, in the range of degrees of nonparabolicity K from 0.1 to 10, the thicknesses of contact layers from 20 to 150 µm and temperatures from 200 to 400 K, the electrical contact resistance of transient contact layer due to the deviation of SL TEM surface from the ideal plane varies from $8 \cdot 10^{-9}$ to $1.9 \cdot 10^{-7}$ Ohm·cm^2, the electrical contact resistance of transient contact layer due to steady-state diffusion of metal in SL TEM without formation of clusters – from $8 \cdot 10^{-9}$ to $4 \cdot 10^{-7}$ Ohm·cm^2, the electrical contact resistance of transient contact layer due to steady-state diffusion of metal in SL TEM with formation of clusters – from $8 \cdot 10^{-9}$ to $4.5 \cdot 10^{-7}$ Ohm·cm^2. Therefore, in the case of SL TEM the formation of clusters affects the SL TEM-metal electrical contact resistance significantly less than in the case of SL TEM with a parabolic band spectrum.

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ВЛИЯНИЕ НЕПАРАБОЛИЧНОСТИ, ОПИСЫВАЕМОЙ МОДЕЛЬЮ ФИВАЗА НА ЭЛЕКТРИЧЕСКОЕ КОНТАКТНОЕ СОПРОТИВЛЕНИЕ ТЕРМОЭЛЕКТРИЧЕСКИЙ МАТЕРИАЛ - МЕТАЛЛ

Исследованы температурные зависимости электрического контактного сопротивления термоэлектрический материал - металл в случае, когда зонный спектр свободных носителей заряда в материале описывается моделью Фиваза. Рассмотрены переходный контактный слой, образованный отклонением поверхности полупроводникового термоэлектрического материала со сверхрешеткой (СРТЭМ) от идеальной плоскости и переходные контактные слои без кластеров и с кластерами, образованные в процессе стационарной диффузии частиц металла в СРТЭМ. Установлено, что контактное сопротивление резко снижается с ростом степени непараболичности зонного спектра СРТЭМ, которая определяется как отношение энергии Ферми идеального двумерного электронного (дырочного) газа с квадратичным законом дисперсии к ширине минизоны, описывающей трансляционное движение носителей заряда в направлении, перпендикулярном плоскости слоев. Такое снижение объясняется блокированием рассеяния свободных носителей заряда в направлении, перпендикулярном плоскости слоев. Показано, что в интервале степеней непараболичности K от 0.1 до 10, толщин переходного слоя от 20 до 150 мкм, безразмерных интенсивностей поступления атомов металла в объем переходного слоя A от 0 до 1 и температур от 200 до 400 K электрическое контактное сопротивление переходного слоя, обусловленного отклонением поверхности СРТЭМ от идеальной плоскости меняется от $8 \cdot 10^{-9}$ до $1.9 \cdot 10^{-7}$ Ом·см^2, переходного слоя, обусловленного стационарной диффузией металла в СРТЭМ без образования кластеров - от $8 \cdot 10^{-9}$ до $4 \cdot 10^{-7}$ Ом·см^2, переходного слоя, обусловленного стационарной диффузией металла в СРТЭМ с образованием кластеров - от $8 \cdot 10^{-9}$ до $4.5 \cdot 10^{-7}$ Ом· см^2.

Ключевые слова: модель Фиваза, сверхрешетки, энергия Ферми, минизоны, степень непараболичности, контакт термоэлектрический материал - металл, электрическое контактное сопротивление переходного слоя, отклонения поверхности термоэлектрического материала от идеальной плоскости, стационарная диффузия, интенсивность поступления частиц металла в полупроводник, кластеры.

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ВПЛИВ НЕПАРАБОЛІЧНОСТІ, ОПИСУВАНОЇ МОДЕЛЛЮ ФІВАЗА НА ЕЛЕКТРИЧНИЙ КОНТАКТНИЙ ОПІР ТЕРМОЕЛЕКТРИЧНИЙ МАТЕРІАЛ – МЕТАЛ

Досліджено температурні залежності електричного контактного опору термоелектричний матеріал – метал у випадку, коли зонний спектр вільних носіїв заряду у матеріалі описується моделлю Фіваза. Розглянуто перехідний контактний шар, утворений відхиленням поверхні напівпровідникового термоелектричного матеріалу з надґраткою (НГТЕМ) від ідеальної площини та перехідні контактні шари без кластерів і з кластерами, утворені у процесі стаціонарної дифузії частинок металу у НГТЕМ. Встановлено, що контактний опір різко знижується зі зростанням ступеня непараболічності зонного спектру НГТЕМ, який визначається як відношення енергії Фермі ідеального двовимірного електронного (діркового) газу з квадратичним законом дисперсії до ширини мінізони, яка описує трансляційний рух носіїв заряду у напрямку, перпендикулярному до площини шарів. Таке зниження пояснюється блокуванням розсіювання вільних носіїв заряду у напрямку, перпендикулярному до площини шарів. Показано, що в інтервалі ступенів непараболічності K від 0.1 до 10, товщин перехідного шару від 20 до 150 мкм, безрозмірних інтенсивностей надходження атомів металу в об'єм перехідного шару A від 0 до 1 та температур від 200 до 400 К електричний контактний опір перехідного шару, зумовленого відхиленням поверхні НГТЕМ від ідеальної площини змінюється від 8·10⁻⁹ до 1.9·10⁻⁷Ом·см², перехідного шару,зумовленого стаціонарною дифузією металу у НГТЕМ без утворення кластерів – від 8·10⁻⁹ до 4·10⁻⁷ Ом·см², перехідного шару,зумовленого стаціонарною дифузією металу у НГТЕМ з утворенням кластерів – від 8·10⁻⁹ до 4.5·10⁻⁷ Ом·см².

Ключові слова: модель Фіваза, надґратка, енергія Фермі, мінізона, ступінь непараболічності, контакт термоелектричний матеріал – метал, електричний контактний опір перехідного шару, відхилення поверхні термоелектричного матеріалу від ідеальної площини, стаціонарна дифузія, інтенсивність надходження частинок металу у напівпровідник, кластери.

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