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Conductance of island and granular metal films

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ABSTRACT

Results of measurements of the specific surface conductance of island metal films at different temperatures are presented. The study of conduction allowed us to establish processes, which determine the transfer of charge carriers in granular and island metal structures. These processes determine the excess charge carriers concentration in film and, on the other hand, characterize the transfer speed of excess charge carriers from one island to another (i.e. mobility). Moreover, these processes occur independently from each other.

1. Introduction

Conductance of island and granular metal films in insulating state (conductance increases with the temperature growth) is widely discussed in the literature [1–16]. It is established that transfer of charge carriers in granular and island films is caused by tunnelling of electrons between islands. Tunnelling of electrons from one neutral island to another changes energy of the system by the value approximately equal to the island charge energy $E \approx e^2/2C$, where e is the electron charge, C is the island capacitance. Under such charge transfer conditions the conductance should be described by activation dependence (Arrhenius law) $\sigma = \sigma_0 \exp(-E/kT)$, where k is the Boltzmann constant, and T is the temperature [15]. However, it has been experimentally found that conductance of the island metal structures and nanocomposites often depends on the temperature according to the expression [1,7]:

$$\sigma = \sigma_0 \exp[-(T_0/T)^{0.5}], \quad (1)$$

where σ_0 is the film conductance at high temperatures and T_0 is material dependent parameter.

Thus, most of the experimental data are described by the "1/2 law" [1–3]. Different theoretical models have been used to explain the "1/2 law" [4–8,15]. In general, these models are a modification of the theory of hopping conduction in semiconductors, where the "1/2 law" is interpreted as the appearance of Coulomb gap in the density of electronic states near the Fermi level. Papers [4,5] point to the important role of the Coulomb interaction between charged particles. There are also other models explaining the "1/2 law", for example, in the paper [7] conductance of granular structures is associated with the variation in sizes of metal granules.

Several studies have shown that degree (x) in expression describing nanocomposite conductance is not always equal to 1/2. In the paper [8] it is shown that, when the hopping length is less than size of islands and close to the distance between them, the standard theory of hopping conduction with the variable hopping length is not applicable. In the paper [9] it was found that $x=0.75$, in the paper [10] $x=0.72$, in the paper [11] $x=1$. In the paper [11], where the mechanism of conduction in island metal films of Au, Ni, and Pt was studied, authors experimentally have shown that conductance of 1 nm thick film changes with temperature according to the Arrhenius law. Also, it was noted that with the increase of film thickness up to 3 nm deviation from the activation dependence has been observed. The reasons of such deviations from the dependence with $x=1/2$ still remain the subject of debate. The nature itself of the conductance dependence of island and granular films in accordance to $\sigma = \sigma_0 \exp[-(T_0/T)^{0.5}]$ in wide temperature range ($kT < E \approx e^2/2C$) remains mystery [17].

In this paper we will show that change of conductance in granular and island films in wide temperature range is defined by generally accepted activation and tunnel processes, but, what is very important, these processes take place independently. This approach to the problem of charge carriers transfer in granular and island films allows us to explain the conductance dependence on the temperature with any degree (x) in the expression.

2. Problem formulation and experiment

We shall consider, as in the paper [11], that current flow in the island metal films and granular systems is caused by two

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consecutive processes. The first process determines the excess charge carriers concentration in the island or granular film and it is associated with the excitation of electrons from the traps to the neutral islands with the charged island formation or with the tunnelling of electrons from one neutral island to another with the formation of positively and negatively charged islands. This process occurs with system energy change on the value $E \approx e^2/2C$. At low temperatures positively and negatively charged islands can be separated by neutral islands. Type of electron transfer process from one neutral island to another (e.g. virtual hopping process [15,16]) has an impact only on the relaxation time of the equilibrium concentration of charged islands at given temperature. This first process determines the concentration of islands with excess charge carriers. The greater the concentration of charged islands, the higher the system conductance.

Second process characterizes the transition speed of the excess charge carriers between the islands (i.e. mobility) and it is caused by tunnelling of charge carriers from charged island to the neutral one. This process adds term in conductance that takes barrier properties dependence into account $\sigma \propto \exp(-L/\lambda)$, where L is the hopping length, $\lambda = \hbar/(mW)^{0.5}$ is the length of electron wave function decay in dielectric that separates metal islands, m is the electron mass, W is the tunnelling barrier height (practically equal to a half-width of the dielectric band gap). This kind of tunnelling changes the energy of the system by $\Delta E \approx e^2(1/C_1 - 1/C_2)$, where C_1 and C_2 are the capacitances of different size islands. If islands are identical then the energy of the system does not change. We must note that charge energy $E \approx e^2/2C \gg \Delta E \approx e^2(1/C_1 - 1/C_2)$, so later we neglect ΔE.

Therefore the conductance of island film in insulator state can be defined as follows:

$$\sigma = \sigma_0 \exp(-L/\lambda - E/kT), \tag{2}$$

We assume that this equation can describe any kind of experimental conductance dependence in island structures, including Eq. (1).

We assume that experimental dependence of the granular and island structures conductance, which is described by Eq. (1), is, firstly, a consequence of activation energy E magnitude variation. Secondly, the dependence can be caused by change of the hopping length L with the structure temperature change. Moreover, these two processes occur independently from each other.

The first process is responsible for the excess charge carriers concentration in the film. Variation of the activation energy magnitude in granular and island systems may be associated with the presence of charged defects in dielectric matrix of the structure [16] or with the variation of granule size. For example, in the structures with insulating conductance the island size can vary from $D = 10$ nm to $D = 200$ nm. Charge energy of islands ($E \approx e^2/\varepsilon D$) in its turn can vary from 0.005 eV to 0.1 eV [1,11,12]. In this case, the conductance dependence on the temperature differs from the activation dependence $\sigma = \sigma_0 \exp(-E/kT)$, in which the activation energy is constant.

For example, let us review island metal film, which contains two types of islands. The first type with D_1 island size and N_1 island concentration, and the second type with D_2 island size and N_2 island concentration. Charge energy of these islands equals, $E_1 \approx e^2/\varepsilon D_1$ and $E_2 \approx e^2/\varepsilon D_2$ ($E_1 \ll E_2$ or $D_1 \gg D_2$). At low temperatures ($T \approx E_1/k$) due to tunnelling transitions of electrons between neutral islands or due to electron excitation from the traps to the neutral islands part of the islands becomes positively or negatively charged. The excess electrons or holes are captured by islands with minimum charge energy in the first place. In our case, it is the islands with the size D_1, charge energy E_1, and charged islands concentration $n_1 = N_1 \exp(-E_1/kT)$ [7,11].

Charge carriers transfer in the electric field will be carried out due to tunnelling transitions between charged and neutral islands of size D_1. Energy of the system does not change, because islands have the same size, hence, the tunnelling probability does not depend on the temperature. Conductance dependence on the temperature in the proximity of $T \approx E_1/k$ will be of the activation type with the activation energy close to E_1.

With the temperature growth the excess charge carriers will be excited on smaller islands (of D_2 size) with charge energy $E_2 \approx e^2/\varepsilon D_2$. Charged islands of size D_2 will now participate in tunnelling transition of charge carriers. Activation type of conductance still remains with temperature growth. As two types of islands take part in the charge carriers transfer, the process will be characterized by the aggregate activation energy (E_S). The E_S value will depend on the temperature and concentration of charged islands $n_1 = N_1 \exp(-E_1/kT)$ and $n_2 = N_2 \exp(-E_2/kT)$. Activation energy E_S in this case can be represented by the following expression:

$$E_S = \frac{E_1 N_1 \exp(-E_1/kT) + E_2 N_2 \exp(-E_2/kT)}{N_1 \exp(-E_1/kT) + N_2 \exp(-E_2/kT)},$$

where $E_1 N_1 \exp(-E_1/kT)$ and $E_2 N_2 \exp(-E_2/kT)$ are changes in energy of the systems, in which n_1 and n_2 charged islands are exited. The activation energy E_S will increase with the temperature growth from $E_S \approx E_1$ to $E_S \to E_2$ ($E_1 \ll E_2$). The dependence of energy E_S on the temperature and, consequently, the conductance dependence on the temperature is determined by the distribution of island sizes. If islands are identical then the activation energy is constant and the conductance dependence on the temperature obeys the Arrhenius law $\sigma = \sigma_0 \exp(-E/kT)$ [11]. On the other hand, in paper [16] authors report that they have created periodic granulated structures and the island size was controlled with an accuracy of few percent. However, the "1/2 law" has been observed in these structures. As suggested by the authors, such conductance dependence on the temperature in the periodic granular systems can be connected with the presence of charged defects in dielectric matrix [16], which creates a random potential in the structure and leads to the disorder of charge energy. In general case of arbitrary island size distribution the activation energy E_S can be presented as follows:

$$E_S = \frac{\sum_1^n E_i N_i \exp(-E_i/kT)}{\sum_1^n N_i \exp(-E_i/kT)}, \tag{3}$$

where $i = 1, 2, 3, \ldots, n$ characterizes specific size of the islands and their concentration. It should be emphasized that with the temperature growth it is becoming possible for one, two, three, and more excess electrons to transit on larger islands. The charge energy, for example, of size D island will have a value of $E \approx e^2/\varepsilon D$, $E \approx (2e)^2/\varepsilon D$, $E \approx (3e)^2/\varepsilon D$, etc.

The second process determines the charge carriers transfer in the electric field and caused by tunnelling transitions between charged and neutral islands, which are characterized by the hopping length L. Obviously, any kind of conductance dependencies for the island and granular films $\sigma = \sigma_0 \exp[-(T_0/T)^x]$, where $x = 0.5$ [1–3], $x = 0.75$ [9], $x = 0.72$ [10], $x = 1$ [11], can be explained by the dependence $\sigma = \sigma_0 \exp(-L/\lambda - E_S/kT)$, where the activation energy E_S and the hopping length L depend on the temperature, but do not depend on each other.

To verify these assumptions we have created island metal films of Tungsten (W) from 0.63 to 2 nm thick. The topography of the film surface was studied by the atomic force microscopy (AFM) method. The differential conductance dependence of metallic films on the temperature was measured.

Tungsten films were grown by the RF-sputtering method. At the beginning of the process a vacuum chamber with the sample was kept at 2×10^{-6} mbar pressure. Then Tungsten was sputtered