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Electrical and Dielectric Properties of Nanoisland Systems Below Percolation Threshold

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Electric and dielectric properties of nanoisland metal films were studied in this work. Permittivity of these films was determined from the analysis of the dependences of the susceptance of the films. It is found that the effective permittivity of the researched structures has abnormally high positive value (107-108). The temperature dependence of the permittivity of the films practically does not differ from the temperature dependence of the conductivity of these films. Such behavior of these dependences is possible, if the reasons for the change of permittivity and conductivity of the films with the temperature are of the same nature. With the growth of the electric field effective permittivity of the films decreases and the conductivity increases. The cause behind the high effective permittivity of the films is the island nature of these metal films, and it is associated with the polarization of the dipoles, formed by couples negatively and positively charged metal islands.

Keywords: Nanoislands, Multilayer, Tunneling, Permittivity.

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1. INTRODUCTION

Nanostructures formed by granulated or discontinuous metal films offer a number of unique properties. The emission of light in an external electric field [1], photoconductivity in the visible and infrared regions of the spectrum [2], and a significant change in the differential conductivity of discontinuous metal films from the weak electric field at temperatures up to 300K [3] were found in these structures. In the structures with not too high density of metal islands the thermally activated conductivity were implemented. However, the mechanism of transfer of charge carriers in nanoisland and granular systems are still under discussion. In this work electrical and dielectric properties of nanoisland metal systems were studied, to do so, the measurements of the temperature dependences of the active and reactive differential conductance of the structures on the basis of metal nanoislands Co, W, FeNi were taken. These measurements were taken in combination with the study of the dependences of the active and reactive conductivity of the film from the electric field. Permittivity of the films was determined from the analysis of the dependences of the susceptance of the films.

2. SAMPLE PREPARATION

The island films of Co, W, FeNi were grown on D_{sub} = 0,5 mm thick dielectric substrate (glass-ceramics) by RF-sputtering. Measurement were taken on both single-layer and multilayer films.

1. FeNi film consisted of ten island metallic layers. Each FeNi layer was d = 7 Å thick, between FeNi layers there was Al_2O_3 layer d = 20 Å thick.
2. Co film consisted of five island layers d = 9 Å thick each. Between Co layers there was Al_2O_3 layer d = 20 Å thick.
3. Island W film was d = 12 Å thick. After deposition film was covered with layer of Al_2O_3 d = 8 Å thick.

The effective thickness of the metal and dielectric layers was determined from the time of deposition with the well-known deposition rate. Earlier in [2] by the method of atomic-force microscopy studies of the topography of the single-layer films FeNi, Co, W, which were specially made for this purpose, were conducted. It is shown that the single-layer metal film of FeNi with the effective thickness of d = 7 Å has island structure, with the size of the islands ~15-30 nm. The islands are apart from each on the average at the distance up to 1.5-5 nm. If you take the average distance between the islands d = 2 nm and the average diameter of the islands equal to r = 18 nm, the number of islands per unit area will be equal to $N = 2.5 \cdot 10^{11}$ cm^{-2}. Part of the volume occupied by metal islands is equal to $V/V_0 = N\pi r^2 = 0.64$. Metal film of Co (d = 9 Å) and W (d = 12 Å) had such island structure but with the larger size of the islands. Structures (1-3) were sputtered on rectangular samples of 0.5-1.5 mm width and 2-3.5 mm length. Ohmic contacts were created by the application of narrow strips of indium to the surface of the film along the opposite sides of the rectangle. In the study of active and reactive differential conductance voltage $U = U_0 + U_1\cos(\omega t)$ were applied to the sample. The amplitude of the alternating voltage was equal to $U_1 = 10^{-2}$ V. Constant voltage U_0 changed from U_0 = 0 V to $U_0 \approx$ 15 V (electric field in the samples varied between F = 0 V/cm up to $F \approx$ 60 V/cm). Measurements of differential conductance were made at the frequency of the external electric field of $f = \omega/2\pi$ = 12 kHz. It should be emphasized that the voltage in these structures lagged the current by phase, i.e. the imaginary part of the impedance was capacitive in nature. Resistance and capacitance of the film FeNi per unit area at a temperature of T = 300K were equal to $R = 7.7 \cdot 10^4$ Ohms and $C = 3.3 \cdot 10^{-11}$ F, the resistance of the film Co $R = 4.45 \cdot 10^7$ Ohm, and capacitance $C = 4.3 \cdot 10^{-13}$ F. The capacity of dielectric substrate of the area S = 1 cm^2 in air is approximately equal to the $C_{sub} \approx 5 \cdot 10^{-14}$ F. The measurement error of conductivity and capacity estimated about 5%. Reactive component of the conductivity of films was determined from measurements of the phase shift of the current relatively to the phase of AC voltage applied to a sample. It is estab-

lished that the electric field applied to the film is quasistationary. Conditions for quasistationarity of electric field implies that the electric field is concentrated inside the sample, and outside the sample value of the electric field is much less than the electric field inside a sample. In the case of quasistationarity of the electric field in the sample for the estimation of the values of the effective permittivity film can be considered as a flat capacitor ($C = \varepsilon\varepsilon_0/d$, where ε – effective permittivity of the structure, ε_0 – vacuum permittivity, d – film thickness). By measuring the reactive component of the conductivity of the film one can estimate the value of the effective permittivity of a structure.

3. RESULTS AND DISCUSSION

Figures 1 and 2 show dependences of differential conductivity ($\sigma(0)$) and permittivity of permalloy (FeNi) film on the temperature, where $\sigma(0)$ – differential conductivity of the film per unit area, with $U_0 = 0$ V. The dependence of the dielectric permittivity on the temperature practically does not differ from the temperature dependence of the conductivity, which points to a common cause of changes of capacity and conductivity from the temperature. On the figure it is seen that the higher the temperature the higher the value of the effective permittivity. What most draws attention to itself – it is the anomaly high value of the effective permittivity of discontinuous films in the whole range of measured temperatures – 10^7-10^8. For comparison, low-frequency permittivity of ferroelectric materials, characterized by high values of permittivity has a value of about 10^4.

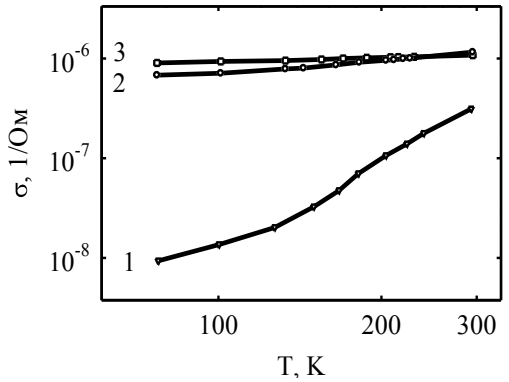

Fig. 1

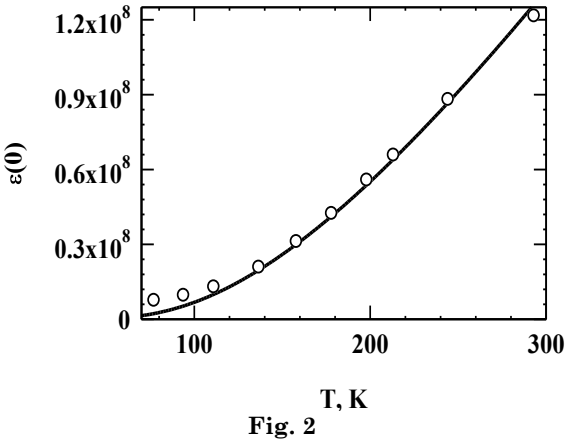

Fig. 2

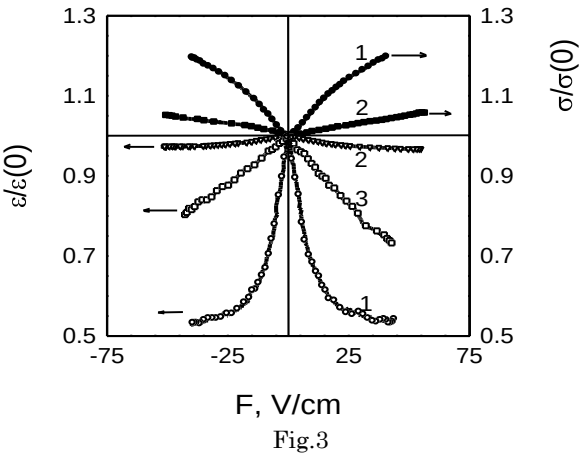

Fig.3

In these same structures it has been studied the influence of the electric field (F) on electrical and dielectric properties. We measured the dependence of the active and reactive components of conductance of the structures on the electric field. Figure 3 shows the dependence of the relative permittivity and relative conductivity on the electric field. Where ε and σ — permittivity and conductivity of films when $F \neq 0$ V/cm, $\varepsilon(0)$ and $\sigma(0)$ — permittivity and conductivity of the film when $F = 0$ V/cm. FeNi and Co structures were measured at $T = 77$ K temperature, and the W structure was measured at a temperature of T = 300K.

Permittivity and conductivity of the films of FeNi when F = 0 V/cm are equal to $\varepsilon(0) = 8.5 \cdot 10^6$, $\sigma(0) = 6.1 \cdot 10^{-7}$ 1/Ω at $T = 77$ K, of the films of Co – $\varepsilon(0) = 4 \cdot 10^6$, $\sigma(0) = 2.25 \cdot 10^{-8}$ 1/Ω at $T = 77$ K, of the films of W – $\varepsilon(0) = 1.2 \cdot 10^7$ at $T = 300$ K. Figure 3 shows that the effective permittivity of the films with the growth of the electric field decreases, and the conductivity increases. The most dramatic increase of conductivity and the most dramatic decrease of the permittivity is observed in electric fields from $F = 0$ V/cm up to $F \approx 25$ V/cm (for example, figure 3 (curves 1)). A further increase in the electric field leads to a slower growth of conductivity and, practically, to the saturation of the values of the permittivity.

The observed high values of the effective permittivity of the structures of metal nanoislands, in which a dielectric character of the conductivity been established, and temperature dependence of the permittivity show, that the nature of the permittivity in conducting systems with metal nanoparticles is fundamentally different from the nature of the permittivity both in the continuous metal films, and in nonconductive systems with nanoislands. It was noted above that the temperature dependence of the permittivity of the films practically does not differ from the temperature dependence of the conductivity of these films (Fig. 1 and Fig. 2). Such behavior of these relations is possible, if the cause for the change of permittivity and conductivity of the films with the temperature are of the same nature. It is known, that the mechanism of activated conductivity in films with a system of metal nanoislands caused by two consecutive processes. Firstly, at a nonzero temperature due to the tunneling transitions of electrons between metal islands the thermodynamic equilibrium is established, in which part of the islands becomes posi-

tive or negative charged. The energy of the system is changed to the value $E \sim e^2/\varepsilon R$, where R – longitudinal size of the metal island. The number of such charged islands proportional to $N_{+-} \sim T \cdot \exp(-E/kT)$. Secondly, as a result of a presence of the charged islands the conditions for the conductivity due to tunneling between charged and neutral islands without a significant change in the energy of the system $\Delta E \sim (e^2/\varepsilon)(1/R_1 - 1/R_2)$ appear. The value of energy ΔE is determined by the disparity of the geometrical sizes of islands. It was noted above that the reason for high positive permittivity and mechanism of transfer of charge carriers in nanoisland structures are determined by the same phenomenon. As shown, the conductivity of nanoisland structures connected with the appearance of excess charges on individual islands (positive and negative), the number of which ($N_+ + N_-$) increases with the temperature. These excess charges of the islands are responsible for tunnel transfer of charge carriers in nanoisland structure. On the other hand, since the current in nanoisland structures is determined by tunneling, the excess charge carriers on the islands can be considered as temporarily localized. The localization time for the excessive charges on the islands is determined by the probability of their tunneling. In the period of time when the excessive charges located on the nanoislands their movement is limited by the size of the metal island and, consequently, a pair of positively and negatively charged islands can be seen as dipoles. The number of dipoles is proportional to $(N_+ + N_-)/2$. Obviously, the capacity and positive permittivity of the nanoisland structure largely determined by the concentration of these dipoles. Thus, the temperature dependences of the permittivity $\varepsilon(T)$ and conductivity $\sigma(T)$ are determined by the temperature dependence of the concentration of charged islands ($N_+ + N_-$).

The electric field has a significant impact on the magnitude and sign of the effective permittivity of discontinuous metal films. With the growth of the electric field the conductivity of discontinuous metal films is increasing. When the conductivity of the film is increasing, the effective permittivity should decrease. Indeed, full permittivity in conducting systems with the account of the attenuation and bias currents in an alternating electric field with a frequency ω is determined by the expression:

$$\varepsilon = \varepsilon_0 - 4\pi\sigma_0 \frac{\tau_0}{1+\omega^2\tau_0^2}, \quad (1)$$

where ε_0 – the macroscopic permittivity of the film, σ_0 – DC conductivity, τ_0 – electron relaxation time (in this case this time is determined by the inverse value of the probability of tunneling of charge carriers from the charged island to the neutral one τ_r). It is obvious, as it follows from the expression (1), that with the growth of the conductivity the permittivity should reduce as it is observed in Figure 3.

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