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Negative capacitance in island metal films

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Studies of electrical properties of island films were performed. Dependences of differential conductivity of films on temperature and frequency of external electric field are measured. Studies of electrophysical properties of films ensure determination of processes which determine occurrence of negative capacitance in the island films. On one hand, these processes are determined by concentration increasing of excess charge carriers in the film under the action of the electric field, on the other hand, by inertia of current change in the film relative to external variable electric field. The inertia of current change is associated with rate of generation and recombination of concentration of excess charged islands.

Keywords: Island film, thin film, negative capacitance, dielectric permittivity.

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1. Introduction

Effect of negative capacitance (NC) can occur in structures where the effect of delay is observed, when charge accumulation in the system occurs slower than change in the applied voltage. So, at alternative current there is phase shift between the current and voltage, corresponding to negative dielectric permittivity [1] or, so called inductive behavior of dynamic characteristics of the system (negative capacitance).

The negative capacitance is observed in Schottky barrier diodes manufactured from dissimilar materials (NiSi$-n$Si; WNi$-n$GaAs; Pd$-n$GaAs; Pd$-n$Si) [2–4] at direct bias voltages. in heterostructure Ni$-$TiO$_2-p$Si the NC effect is observed at reverse voltage on Schottky barrier Ni$-$TiO$_2$ [5]. There are some papers [1,6,7], where NC is associated with the inertia of current change when direct or alternative voltage is applied to the sample.

In bulk metals in variable electric field the main role belongs to conductivity current, which by many times exceed the bias currents, there is no practical possibility to observe accumulation of charge carriers under action of the variable electric field. But, in island metal films, where the conductivity current and the bias current insignificantly differ by value, we observed the negative capacitance effect. Study of features of structure behavior of island metal film with electrical contacts is task of this paper.

2. Experiment

In study we measured the temperature dependences of the active and reactive conductivities of the island films made of FeNi on frequency of electric field. FeNi thin films were grown on a dielectric substrate made of polycrystalline glass (rutile phase of TiO$_2$ [8]) by method of high-frequency sputtering in argon. See paper [9] for details of method of films production. Film thickness was selected such that to obtain the island films with dielectric nature of conductivity, at that following the results in paper [10]. In this paper we determined the percolation threshold for films FeNi — d^*. The percolation threshold was determined as thickness d when transition metal-dielectric occurs in the film. Films $d < d^*$ thick are island films. For FeNi $d^* \approx 1.8$ nm.

For measurement single-layer films FeNi with different effective thickness were selected: $d = 0.7, 0.9$ and 1.1 nm. On each metal structure the protective layer Al$_2$O$_3$ was sputtered at top with effective thickness $d = 2.0$ nm. The effective thickness of metal dielectric layers was determined by time of films sputtering (deposition rates of metal films and Al$_2$O$_3$ were determined in advance). The effective thickness of film would be equal to actual film thickness if the film would be continuous. From structures rectangular samples with width 1.5$-$3 mm and length 5$-$7 mm were manufactured. Contacts were created by application of narrow strips of indium on surface of island metal film along opposite sides of the rectangular.

When studying frequency dependence of active and reactive differential conductivity of films alternating voltage $U = U_1 \exp(-i\omega t)$ was applied to the sample. The alternating voltage amplitude was $U_1 = 10^{-2}$ V. Active and reactive differential conductivity was measured in range of frequencies of 0.5 to 100 kHz. The reactive component of conductivity of films was determined using phase detection. The unit ensured measurements of alternating current from 10^{-8} A. Minimum measured capacity was 1 pF. The measurement error did not exceed 5%. Measurements were made at temperature of 77 to 300 K.

The dependences of the specific differential conductivity of FeNi films of different thickness on temperature are shown in Figure 1.

Figure 2 shows specific differential capacitance vs. temperature for FeNi films with thickness thick $d = 0.7$ nm and $d = 0.9$ nm (curves *1* and *2* respectively). The attempts to measure capacitance in film 1.1 nm using phase detection were not successful due to weak and unstable signal from capacitance as compared to signal from film conductivity. Note that unstable signal demonstrated negative nature of the capacitance in all range of measured temperatures. Figure 3 shows differential capacitance of island film FeNi $d = 0.7$ nm thick vs. frequency of electric field.

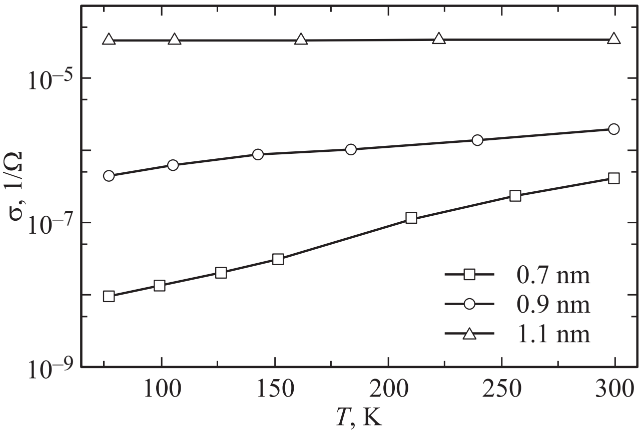

Figure 1. Dependence of specific differential conductivity of FeNi films of different thickness.

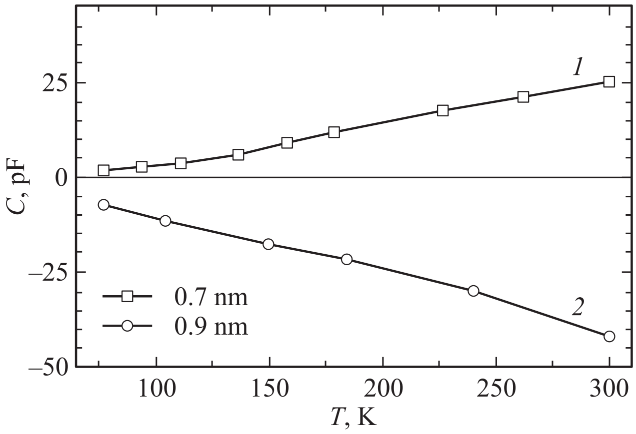

Figure 2. Dependence of specific differential capacitance of FeNi films of different thickness.

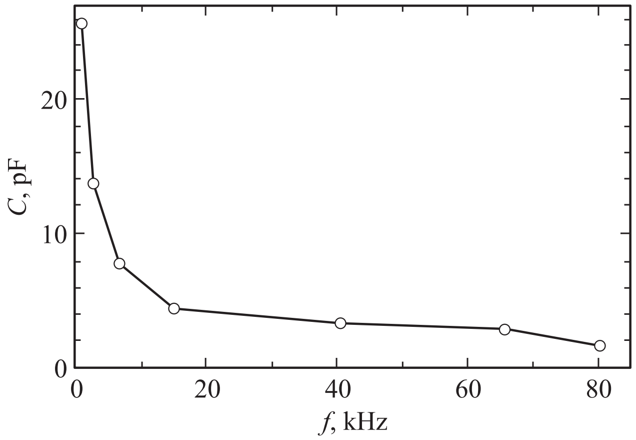

Figure 3. Specific differential capacitance of FeNi films of thickness $d = 0.7$ nm vs. frequency.

3. Discussion of results

From Figure 2 we see that capacitances of samples with thickness of films $d = 0.7$ nm and $d = 0.9$ nm qualitatively differ from each other. Capacitance of film $d = 0.7$ nm thick in entire measured range of temperatures is positive. Capacitance of film $d = 0.9$ nm thick is negative. General feature is their dependence on temperature, which by absolute value increases with temperature increasing. Increase in capacitance of samples by absolute value with temperature is practically similar to the temperature dependence of conductivity (Figure 1). The main reason of conductivity increasing with temperature increasing is associated with concentration increasing of charged islands [11]. It is obvious that the value of the sample capacitance also depends on the concentration of charged islands.

The conductivity of island films FeNi with thickness below $d = 1.1$ nm increases with temperature increasing (Figure 1). In these films the dielectric nature of conductivity is observed. The conductivity of island films is associated with the electrons tunneling between islands. During the electron tunneling from one neutral island to another neutral island two charged islands are created. One with excess electron — island with negative charge, another — island with positive charge. Paper [11] showed that conductivity in island metal film with dielectric nature of conductivity can be represented as dependence:

$$\sigma = 2\beta N_S \exp(-L/\lambda - E_S/kT), \qquad (1)$$

where E_S is reduced total activation energy, approximately equal to averaged charge energy ($E_S \approx q^2/\varepsilon D_S$, where q is electron charge, ε is dielectric permittivity of dielectric between islands, D_S is average size of charged islands [11]), k is Boltzmann constant, T is film temperature, L is hop length, λ is length of decreasing of wave function of electron in dielectric, which separates the metal islands ($\lambda = \hbar/(mW)^{0.5}$, where m is electron mass, W is height of tunnel barrier, practically coinciding with half-width of band gap of dielectric), N_S is concentration of islands where charge energy is $E_S \leq kT$, β is coefficient of proportionality.

From the expression (1) the conductivity of island film is determined by two processes. One of them determines the concentration of excess charge carriers in the island film and is associated with the electron tunneling from one neutral island to another neutral island with the formation of positively and negatively charged islands. Coefficient „2“ in equation (1) indicates that during one act of electron tunneling from one neutral island to another one two charged islands are formed (positive and negative). This process occurs with change in system energy by value E_S, at that the concentration, for example, of negatively charged islands is:

$$n = N_S \exp(-E_S/kT). \tag{2}$$

The second process determines the transfer of charge carriers under the action of the electric field and is carried out due to tunnel transitions between charged and neutral islands, which are characterized by the hop length (L).

As the process of tunneling between the neutral islands occurs with change in system energy by value E_S, the external electric field (F) decreases the potential barrier in the direction of opposite to the electric field by value $U = -qFh$, where h is mean distance between islands. This energy decreasing increases the probability of thermal excitation of tunnel transitions between the islands and facilitates the concentration increasing of charged islands

$$\begin{aligned} n + \Delta n &= N_S \exp\big(-(E_S - U)/kT\big) \\ &= (1 + qhF/kT) N_S \exp(-E_S/kT), \end{aligned}$$

$$\Delta n = \frac{qFhN_S \exp(-E_S/kT)}{kT}. \tag{3}$$

Taking into account the change in the concentration of charged islands and in accordance with the objectives of the article, we will consider the electric field effect on the conductivity and capacitance of the film. The behavior of charge carriers under the action of the external electric field is determined by the equation for the current density and by the continuity equation. In one-dimension case for the island film the equation for current density and continuity equation have view:

$$J = (\sigma + \Delta\sigma) F, \tag{4}$$

$$\partial\Delta n/\partial t = G - \Delta n/\tau, \tag{5}$$

where $\Delta\sigma$ is change in film conductivity in external electric field, F is strength of external electric field, G is rate of generation of charged islands, $\Delta n/\tau$ is rate of recombination of excess concentration of charged islands, τ is life time of charged islands.

Let $F = F_0 \exp(i\omega t)$, then the generation rate for positively charged and negatively charged islands, considering (3), can be represented using expression:

$$G = \frac{qhF_0 \exp(i\omega t)}{\tau kT} N_S \exp(-E_S/kT). \tag{6}$$

In this case, continuity equation will be:

$$\frac{\partial\Delta n}{\partial t} + \frac{\Delta n}{\tau} = \frac{qhF_0 \exp(i\omega t) N_S \exp(-E_S/kT)}{\tau kT}. \tag{7}$$

We will look for solution to this equation in the form:

$$\Delta n = \Delta n_0 \exp(i\omega t). \tag{8}$$

Substituting Δn in equation (7), we obtain:

$$\Delta n_0 = \frac{qF_0 hN_S \exp(-E_S/kT)}{kT} \frac{1 - i\omega\tau}{1 + \omega^2\tau^2}. \tag{9}$$

Equation for current density of island film in variable electric field is sum comprising conductivity current and Maxwell bias current, which density, as per Maxwell equations, is:

$$J_M = \frac{\varepsilon}{4\pi} \frac{\partial F_0 \exp(i\omega t)}{\partial t},$$

where ε is macroscopic dielectric permittivity of film. Equation for full current is:

$$J = (\sigma + \Delta\sigma) F_0 \exp(i\omega t) + \frac{\varepsilon}{4\pi} \frac{\partial F_0 \exp(i\omega t)}{\partial t}. \tag{10}$$

Change in film conductivity in external variable electric field $(\Delta\sigma)$ considering equations (1), (3) and (9) is:

$$\begin{aligned} \Delta\sigma &= \big[2\beta hqN_S F_0 \exp(i\omega t) \exp(-L/\lambda - E_S/kT)\big] \\ &\times \frac{1 - i\omega t}{kT(1 + \omega^2\tau^2)} = \sigma hqF_0 \exp(i\omega t) \frac{1 - i\omega t}{kT(1 + \omega^2\tau^2)}. \end{aligned} \tag{11}$$

Substituting in equation for current density of island film (10) the expression for conductivity change (11) we obtain:

$$\begin{aligned} J = \Bigg\{ &\sigma \left[1 + \frac{hqF_0 \exp(i\omega t)}{kT(1 + \omega^2\tau^2)}\right] \\ &+ i\omega \left[\frac{\varepsilon}{4\pi} - \sigma \frac{hqF_0 \exp(i\omega t)}{kT(1 + \omega^2\tau^2)}\right] \Bigg\} F_0 \exp(i\omega t). \end{aligned} \tag{12}$$

Conductivity (expression in brace brackets) is complex value. This expression resembles the conductivity equation for system of connected in parallel resistance, capacitor and inductance.

$$Y = g + i(\omega C - 1/\omega L), \tag{13}$$

where Y is complex conductivity of circuit; g is active conductivity; $1/\omega L$ is inductive conductivity; ωC is capacitive susceptance. The complex conductivity of film (12) comprises two terms:

$$\operatorname{Im}(Y) = \omega \left[\frac{\varepsilon}{4\pi} - \sigma \frac{hqF_0 \exp(i\omega t)}{kT(1 + \omega^2\tau^2)}\right]. \tag{14}$$

The first term, like in (13) is determined as capacitive conductivity. The second term is negative, has inductive nature of conductivity. From (14) we see that with increase in active conductivity the absolute value of inductive term increases. As dimension of the second term of reactive conductivity is capacitive, then it is characterized as negative capacitive conductivity.

Measured capacitances of samples with film thicknesses $d = 0.7$ nm in full measured range of temperature are positive. Value of active conductivity of this film is insignificant (Figure 1), and, obviously, second term in (14) is much lower in comparison with the first term. Positive capacitance of film is determined by polarization of neutral islands and localization of charged islands near contacts under action of electric field. Negatively charged islands are localized near the positive contact, positively charged islands are localized near the negative contact.With temperature increasing the concentration of charged islands increases. With their concentration increasing the number of charged islands near contacts increases, therefore, the positive capacitance of film increases.

Effect of the negative capacitance take place if the second term in expression (14) is larger than the first term. The capacitance dependence on temperature for this case is presented in Figure 2 (curve *2* — FeNi with thickness $d = 0.9\,\text{nm}$). The capacitance of film $d = 0.9\,\text{nm}$ thick in the full range of measured temperatures stays negative. The expression (14) shows that the appearance of negative differential capacitance is associated with the second term in square brackets, which has the dimension of capacitance, and it is clear that the value of the film capacitance is associated with the change in conductivity. Change in conductivity in the external electric field is due to increase in excess concentration of charged islands. It should be noted that the second term in the pseudo component of film conductivity — negative value, therefore, current via the film, determined by only conductivity associated with excess charged islands, lags in phase from the voltage applied to the film and is inductive in nature. Inertia of current change is determined by rates of recombination and generation of concentrations of excess charged islands, its value depends on strength of the electric field.

When frequency of external electric field increases, it is possible that electromagnetic energy absorbed by the structure will be determined by the conductivity inside the metal islands. Then the structure will demonstrate the metal Conductivity, and capacitance of structure can be negative. Therefore, for frequency dependence of capacitance it is possible to implement the range of frequencies, in which transition from low-frequency positive capacitance to negative capacitance at higher frequencies of the electric field. The presented in Figure 3 measured dependence of differential capacitance of island film FeNi $d = 0.7\,\text{nm}$ thick in full measured range of frequencies remains positive. So, we can conclude that the contribution to the formation of film capacitance by electrons inside metal islands is insignificant compared to the contribution of conductivity associated with the increase in excess concentration of charged islands in external electric field.

4. Conclusions

Studies of electrical properties of island films were performed. Dependences of differential conductivity of films on temperature and frequency of external electric field are measured.

Capacitances of samples with thicknesses of films 0.7 nm and 0.9 nm qualitatively differ. Capacitance of film 0.7 nm thick in entire measured range of temperatures has positive values. Capacitance of film 0.9 nm thick is negative. General feature is their dependence on temperature, which by absolute value increases with its increasing.

Increase in capacitance of samples by absolute value with temperature practically does not differ from the temperature dependence of conductivity. The main reason of conductivity increasing with temperature increasing is associated with concentration increasing of charged islands. The value of the samples capacitance also depends on the concentration of charged islands.

Occurrence of negative capacitance in the external electric field is due to increase in excess concentration of charged islands. Current via film determined by only conductivity associated with excess charged islands lags by phase from voltage applied to the film, and has inductive nature. Inertia of current change is determined by rates of recombination and generation of concentrations of excess charged islands, its value depends on strength of the electric field.

Conflict of interest

The authors declare that they have no conflict of interest.

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