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Formation of Noncoil-Like Inductance in Nanocomposites $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{1-x}$ Manufactured by Ion-Beam Sputtering of Complex Targets in $Ar+O_2$ Atmosphere

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This paper investigates the inductive contribution to AC conductance in the granular nanocomposites $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{1-x}$. The initial nanocomposites studied were manufactured in $Ar+O_2$ atmosphere by ion-beam sputtering of the target containing $Fe_{0.45}Co_{0.45}Zr_{0.10}$ and alumina stripes and then subjected to the annealing procedure in air over the temperature range 373 K $< T_a <$ 873 K. These samples, before and after annealing, were studied using the temperature 77 K $< T_p <$ 300 K and frequency 50 Hz $< f <$ 1 MHz dependences of a real part of the admittance $\sigma(T,f)$. Analysis of the observed $\sigma(f,T_p)$ dependences for $x < 0.5$ demonstrated that in the studied samples the equivalent circuits with the capacitive and noncoil-like inductive contributions can be accomplished. Just in this case, the capacitive properties of *RLC* circuit with the phase angle $-90^\circ \le \theta_L < 0^\circ$ are exhibited at low frequencies and the inductive properties with $0^\circ \le \theta_H < 90^\circ$ become apparent at high frequencies. A value of the critical frequency f_R, where θ_H changes sign, depends on the metallic phase concentration x, measuring temperature T_p, and annealing temperature T_a.

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1. Introduction

In our previous papers we studied a real part of the admittance σ of the $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{1-x}$ nanocomposite samples containing FeCoZr nanoparticles in the alumina matrix and deposited in $Ar+O_2$ gas mixture by ion-beam sputtering of complex targets containing metallic alloy and alumina stripes [1, 2]. It was established that below the percolation threshold the studied composites were characterized by low values of σ. As follows from [3], the observed negative sign of $d\sigma/dT$ points to the dielectric regime of the electron transport at $x < x_C$. Our previous experiments [1, 2, 4] showed that this is due to the realization of the thermally stimulated tunnelling of electrons over the dielectric barriers created by the alumina matrix and FeCoZr-oxide shells surrounding the metallic nanoparticles.

The main goal of this paper is to study the AC phase angle θ and capacitance C_p in the nanocomposite samples depending on the CoFeZr content, annealing temperature T_a, and measuring temperature T_p.

2. Experimental

In our experiments we used $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{1-x}$ films with the thickness from 3 to 6 μm prepared by ion-beam sputtering of a complex target in a chamber evacuated with a mixture of argon under the pressure P_{Ar} = 0.667 mPa and oxygen with the partial pressure P_{O2} = 2.4 mPa. All the technological procedures and experimental techniques are described in [1, 2, 4–9]. The method used allows for manufacturing of samples with different metallic phase concentrations $0.313 < x < 0.621$ under the identical deposition conditions.

The as-deposited films contained extra oxygen and hence consisted of the metallic alloy nanoparticles covered with complex FeCoZr-oxide "shells" and embedded into the alumina matrix.

The as-deposited films were cut to give rectangular samples, and silver contacts were deposited nearby the film sample butt-ends using a special silver paste. Measurements of AC conductance σ of the studied samples were performed for the frequencies 50 Hz $< f <$ 1 MHz at the measuring temperature T_p from 77 K to 373 K by 5 K steps. After isochronous (15 min) annealing of the samples in a tubular furnace over the temperature range from 398 K to 873 K by 25 K steps the measuring cycle was repeated. This procedure was described in more detail in [10].

3. Results and discussion

During the study of AC conductivity for $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{1-x}$ nanocomposites we have found that after the 15 min step annealing at the temperatures $T_a >$ 373 K the film samples with x between 0.382 and 0.495 display negative values of the phase angle shift $-90^\circ < \Theta_L < 0^\circ$ at low frequencies (see Fig. 1). At the same time, in the high-frequency region the phase angle shift was positive $0^\circ < \Theta_H < +90^\circ$.

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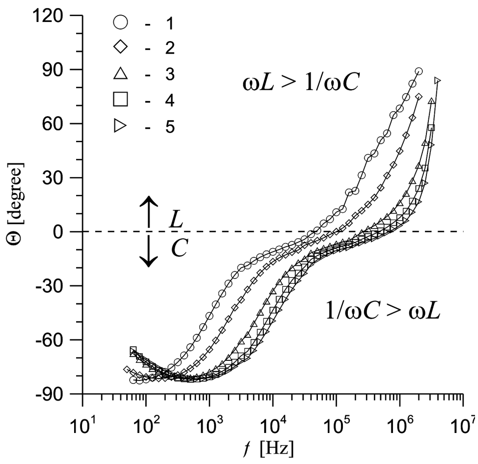

Fig. 1. Dependences of the phase angle shift Θ on the frequency f for the nanocomposite sample $(Fe_{0.45}Co_{0.45}Zr_{0.10})_{0.426}(Al_2O_3)_{0.574}$ after annealing at the temperature T_a = 448 K. Measuring temperatures T_p: 1 — 77 K, 2 — 123 K, 3 — 173 K, 4 — 233 K, 5 — 273 K.

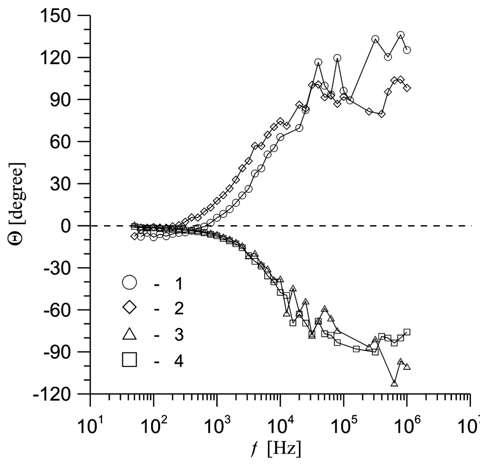

Fig. 2. Dependences of the phase angle shift Θ on the frequency f for the nanocomposite sample $(Fe_{0.45}Co_{0.45}Zr_{0.10})_{0.495}(Al_2O_3)_{0.505}$ after annealing at the temperature T_a = 873 K. Measuring temperatures T_p: 1 — 233 K, 2 — 248 K, 3 — 328 K, 4 — 368 K.

According to the analysis of the observed $\Theta(f)$ dependences, they indicate the presence of the inductive--like contribution to the AC carrier transport at high frequencies. Such a behaviour was mentioned earlier in our work [11] for the nanocomposites deposited in the atmosphere containing reactive gases (oxygen or nitrogen). This was explained by bending of the current-conducting routes having the nanocoil-like shape in nanocomposites with $x < x_\mathrm{C}$ due to the electron tunnelling over the dielectric strata between metallic nanoparticles. It is easy to show that such a behaviour electrotechnically corresponds to an equivalent circuit with the series connected discrete RLC elements having the resonance frequency

$$f_\mathrm{R} = 1/(2\pi\sqrt{LC}) \tag{1}$$

for the phase angle shift $\Theta = 0°$.

However, such a model for the inductive-like behaviour of the studied composites can be considered only qualitatively for four reasons. Firstly, our films contained no real coils (turns) that are specific for the traditional inductive elements L. Secondly, as can be seen from Fig. 1, the observed $\Theta(f)$ curves have a clearly marked minimum in the low-frequency range that is impossible for the circuit with series connected RLC elements. Thirdly, in some cases an increase of the measuring temperatures in our experiments caused switching-off of the inductive--like contribution with $\Theta > 0$ and its replacement by the capacitive-like one with $\Theta < 0$ (see Fig. 2). Fourthly, the resonance frequency f_R in (1), when Θ changes its sign, increases with temperature (see Fig. 1). The latter means that one or both of L and C elements in the proposed equivalent circuit should decrease with a temperature growth.

Figure 3 presents the frequency dependence of capacitance for the sample with a low content of the metallic phase $x = 0.426$ after annealing at the temperature $T_\mathrm{a} = 488$ K, whose $\Theta(f)$ curves are given in Fig. 1. As shown by Fig. 3, $C_p(f)$ dependence in the low-frequency range falls rapidly with the increase in f, approaching its minimum at some critical frequency f_min that depends on the measuring temperature T_p.

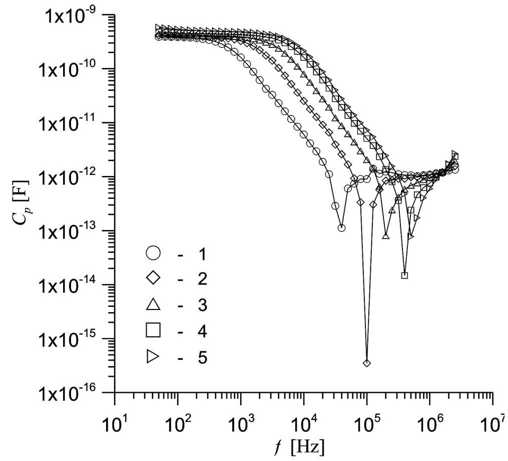

Fig. 3. Frequency dependence of the capacitance C_p for the nanocomposite $(Fe_{0.45}Co_{0.45}Zr_{0.10})_{0.426}(Al_2O_3)_{0.574}$ after annealing at the temperature T_a = 448 K. Measuring temperatures T_p: 1 — 77 K, 2 — 123 K, 3 — 173 K, 4 — 233 K, 5 — 273 K.

As was established in [12], at the AC hopping carrier transport between neutral potential wells (metallic nanoparticles) the additional thermally activated dielectric permeability is induced. It arises due to the electric dipole occurrence owing to jump of the electron from one well to another. When the electron returns back to the first well in the time τ, the induced dipole disappears. The frequency region, where this additional polarization is observed, depends on the time τ involved in the expression

$$f \leq 1/(2\tau)\,. \tag{2}$$

Analysis of Fig. 3 shows that the position of f_{min} closely corresponds to the frequency f_{R}, where $\Theta(f)$ curves have their zero crossings (see Fig. 1).

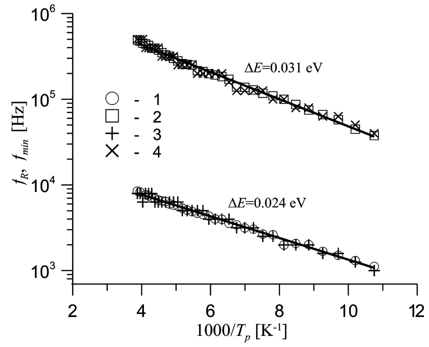

Fig. 4. Dependences of f_{R} (1, 2) and f_{min} (3, 4) on the reversal temperature for the samples with $x = 0.382$ (1, 3) and $x = 0.426$ (2, 4) after annealing at $T_{\mathrm{a}} = 823$ K.

Plotting of the Arrhenius curves $f_{\mathrm{R}}(1000/T_p)$ and $f_{\mathrm{min}}(1000/T_p)$ in Fig. 4 for the sample with $x = 0.382$ after annealing at $T_{\mathrm{a}} = 823$ K and for the sample with $x = 0.426$ after annealing at $T_{\mathrm{a}} = 448$ K demonstrates that the frequencies f_{R} and f_{min} are coincident over the whole region of the measuring temperatures T_p. From these straight lines we can extract the activation energies of the polarization process (dipole inducing) which are equal to 24 meV and 31 meV for the samples with the metallic phase content $x = 0.382$ and 0.426, respectively.

The performed analysis confirms our previous qualitative suggestions in [12] that positive angles of the phase shifts observed in the samples studied, which are specific *RLC* equivalent circuits, are due to the presence of a hopping mechanism in the carrier transport. As follows from the model presented in [12], every jump of the electron from one neutral well (metallic nanoparticle) to another occurs during a period of time τ of the order of 10^{-13} s, and its return to the first well during $\tau \approx 10^{-3}$–10^{-6} s (see Fig. 4). For the frequencies

$$f > 1/(2\tau) \tag{3}$$

the delay $\omega\tau$ can be greater than 2π, that creates the possibility for positive angles of the phase shifts.

4. Conclusion

An analysis of the observed $\sigma(T, f)$ dependences for the granular nanocomposites $(\mathrm{Fe}_{0.45}\mathrm{Co}_{0.45}\mathrm{Zr}_{0.10})_x(\mathrm{Al}_2\mathrm{O}_3)_{1-x}$ with $0.38 < x < 0.50$ manufactured in the $\mathrm{Ar}+\mathrm{O}_2$ atmosphere by ion-beam sputtering of a complex target has revealed that in these samples equivalent circuits corresponding to the series connection of a capacitive and noncoil-like inductive elements can be accomplished. So just for this case, the capacitive properties of an *RLC* circuit with the phase angle $-90° \leq \Theta_{\mathrm{L}} < 0°$ are exhibited at low frequencies ($f < f_{\mathrm{R}}$) and the inductive properties with $0° \leq \Theta_{\mathrm{H}} < 90°$ become apparent at high frequencies ($f > f_{\mathrm{R}}$). A value of the resonance frequency f_{R}, where $\Theta_{\mathrm{R}} = 0°$, depends on the metallic phase concentration x, measuring temperature T_p, and annealing temperature T_{a}. The dependence of $f_{\mathrm{R}}(T_p)$ plotted to the Arrhenius scale has an activation character with the activation energy around (0.024–0.031) eV for the samples with different content x of metallic elements.

Acknowledgments

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