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Dynamic Conductivity of Amorphous Nanogranular Films in the Microwave Frequency Range

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Abstract—The electric conductivity of amorphous nanogranular composite films has been experimentally studied on direct current and under microwave reflection conditions. It is established that the dynamic conductivity exceeds the static value by two to four orders of magnitude. Suggestions concerning the nature of this difference are formulated.

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In recent years, increasing use of various metamaterials–composites, the properties of which are determined by a special structure consisting of separate elements, has been an important trend in microwave technology [1]. Photonic and magnonic crystals [2, 3] possessing left-handed waveguide properties [4] are important examples of these metamaterials. A special position is occupied by nanostructured metal–dielectric films comprising metal granules chaotically distributed in a dielectric matrix [5]. The use of a ferromagnetic metal in this nanostructure provides, in addition to manifestations of the colossal magnetoresistance, a high level of microwave absorption [6], which is of considerable practical interest.

The microwave properties of granular nanocomposites are yet insufficiently studied, all the more so in view of the rapid development of their technology. Quite recently, nanocomposites containing an amorphous metal phase were created [7], but their properties have not been studied so far. On the other hand, no data have been reported on the practically important relationship between static and dynamic properties of nanogranular composite films, a significant difference of which was originally pointed out in [8].

The present work was devoted to studying the microwave properties of amorphous nanogranular composites—in particular, the characteristics of the reflection of electromagnetic waves from composite films—in order to gain information concerning the relationship between their static and dynamic parameters that is necessary for developing data processing devices based on these materials.

We have studied the films of nanogranular composites with a general formula $(Co_{45}Fe_{45}Zr_{10})_x(Zr_2O_3)_{1-x}$, where $x \sim 0.54$–0.78 is somewhat higher than the percolation threshold ($x \sim 0.45$). The films were prepared on Lavsan (Dacron) polymer substrates with a thickness of 0.02 mm and lateral dimensions 21×28 cm by ion-beam deposition with screening [9], which allowed a composite layer with gradually varying thickness to be obtained on the extended substrate in a single deposition cycle. Then, the coated substrate was cut into 12 samples of the same composition with the film thickness varying from 70 to 550 nm, as measured by a scanning electron microscope at 15 points on the edge. The granular character of the film and the amorphous structure of metal grains have been confirmed by electron-microscopic examinations [9–12]. The dimensions of grains fell within 2–7 mm.

The surface of films also exhibited a granular structure, which was studied using an atomic-force microscope with a resolution of about 10 nm. The grain sizes varied from 30 to 100 nm and increased with the film thickness. The grains were spaced by depressed troughs with widths amounting to 20–50% of grain diameters.

The electric conductivity of the composite films was measured by a contact method using the substitution scheme [13, 14]. The microwave power reflection coefficient was measured in a frequency range of 14–18 GHz using a panoramic network analyzer [13–15].

Figure 1 shows a plot of specific conductivity σ versus film thickness d, where points present the results of static (dc) conductivity measurements and the solid curve is constructed according to the empirical formula

$$\sigma(\Omega^{-1}\,\mathrm{m}^{-1}) = \exp\left\{10\tanh\left(\frac{d(\mathrm{nm}) - 300\ \mathrm{nm}}{190\ \mathrm{nm}}\right) + 1\right\}. \quad (1)$$

Figure 2 shows the dependence of microwave reflection coefficient R on film thickness d, where points also present the experimental data. Calculations of the reflection coefficient were based on the formula [16, 17]

$$R = \left|\frac{(\varepsilon_1\mu_2 - \varepsilon_2\mu_1)[\exp(ik_2d) - \exp(-ik_2s)]}{(\sqrt{\varepsilon_2\mu_1} + \sqrt{\varepsilon_1\mu_2})^2\exp(ik_2d) - (\sqrt{\varepsilon_2\mu_1} - \sqrt{\varepsilon_1\mu_2})^2\exp(-ik_2d)}\right|^2, \quad (2)$$

where ε_1, μ_1, and k_1 are the dielectric permittivity, magnetic permeability, and wavenumber in the free space, respectively, and ε_2, μ_2, and k_2 are the analogous quantities in the film; and d is the film thickness. Conductivity σ was taken into account in the form of an imaginary additive to the real dielectric permittivity in the following equation:

$$\varepsilon = \varepsilon_r + i\varepsilon_\sigma = \varepsilon_r - \frac{i\sigma}{\varepsilon_0\omega}, \quad (3)$$

where ε_r is the permittivity in the absence of conductivity, ω is the frequency, and ε_0 is the dielectric constant. In these calculations, it was assumed that, for $\omega = 1.0053 \times 10^{11}\ \mathrm{s}^{-1}$ ($f = 16$ GHz), the free space has $\varepsilon_1 = 1$, $\mu_1 = 1$, and $k_1 = 293\ \mathrm{m}^{-1}$ and the film material in the absence of conductivity is characterized by $\varepsilon_{r2} = 2.5$, $\mu_2 = 1$, and $k_1 = 463\ \mathrm{m}^{-1}$.

The conductivity in Eq. (3) was first calculated using formula (1). The results of these calculations are presented in Fig. 2 by the dashed curve that significantly disagrees with experimental data. The solid curve in Fig. 2, which well approximates the experimental points, was constructed according to the same formula (3) in which the conductivity values were calculated a different formula (instead of (1)):

$$\sigma(\Omega^{-1}\,\mathrm{m}^{-1}) = \exp\left\{7\tanh\left(\frac{d(\mathrm{nm}) - 300\ \mathrm{nm}}{120\ \mathrm{nm}}\right) + 6\right\}. \quad (4)$$

However, the use of this formula to plot conductivity versus film thickness leads to the dashed curve in Fig. 1, which is situated significantly above and shifted leftward from the solid curve (approximating the experimental points of dc conductivity) so that the excess reaches two to four orders of magnitude.

Thus, if measurements of the reflection coefficient are considered as a method of determining a dynamic conductivity (in the microwave range), then one may conclude that this conductivity is two to four orders of magnitude greater than the static value.

It should be noted that some (about twofold) excess of the dynamic conductivity relative to the static (dc) value was previously reported for metal films with a granular structure [8]. However, the excess of several orders of magnitude observed in the present study

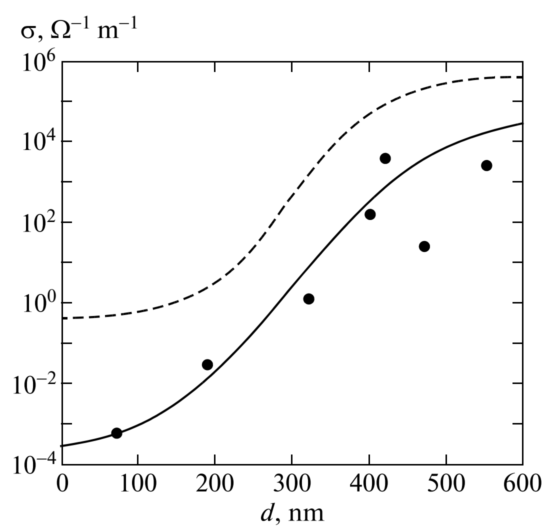

Fig. 1. Dependence of specific conductivity σ on film thickness d. Points present experimental data; the solid curve is constructed in accordance with empirical formula (1), and the dashed curve is constructed in accordance with formula (4).

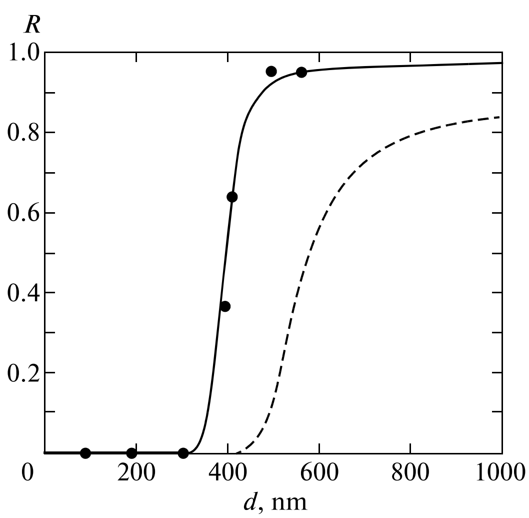

Fig. 2. Dependence of microwave reflection coefficient R on film thickness d. Points present experimental data; the solid curve is constructed according to formulas (3) and (4), while the dashed curve corresponds to formulas (3) and (1).

seems to be related particularly to the granular structure of the composite films studied. This conclusion is suggested by the mechanism of capacitive shunting of intercluster gaps [18], which is especially important in the microwave range.

On the other hand, since the conductivity of the granular composite films under consideration is seven to eight orders of magnitude lower than that of a pure metal (i.e., the conducting percolation channels are rather weak and curved), a significant role in the conductivity can be played by the mechanism of electron tunneling via dielectric gaps between grains [19]. Another consequence of the observed sharp excess of the dynamic conductivity as compared to the static value in these films can be manifested by broadening of the ferromagnetic resonance line in addition to that caused by the mechanism of polarization relaxation [20]. Checking for this issue requires special experiments.

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