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Structure, conductive and reflective properties of amorphous nanogranular composites $(CoFeB)_x+(SiO_2)_{1-x}$

© I.V. Antonets, R.I. Korolev, L.N. Kotov

Syktyvkar State University,
Syktyvkar, Russia

E-mail: aiv@mail.ru

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The results of experimental studies of the structure, conductive and microwave reflective properties in the frequency ranges of 8–12 GHz and 25–36 GHz of amorphous nanogranular composites $(CoFeB)_x+(SiO_2)_{1-x}$, $32.79 < x < 52.00$ at.%, with a thickness of 1.11–2.54 μm, deposited on a lavsan substrate with a thickness of about 20 μm, are presented. The influence of the content of the CoFeB ferromagnetic alloy on the conductive and reflective properties of the films is estimated. The results of experimental effects of magnetic fields with induction up to 0.3 T on the reflection coefficient of microwave waves of composite films are presented.

Keywords: composite films, reflection coefficient, conductivity.

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Nanostructured composites containing metal and dielectric particles, and regions are actively studied in recent decades in connection with the development of the latest small-sized microwave devices [1–3]. Amorphous composites containing metal granules in the alloy have a number of unique properties. The ferromagnetic metal in such composites causes a giant magnetoresistance [4] and a high level of microwave radiation absorption [5]. Zirconium-containing composites can have microwave conductivity by two to four orders of magnitude higher than static conductivity at direct current long before the percolation threshold [6]. Such composites have high specific conductivity 10^2-10^5 S/m and a microwave reflection coefficient varying in a wide range of 0.01–0.96 at a metal phase content of 30–70 at.% and thickness of hundreds of nanometers — units of micrometers [7–9].

In this paper the amorphous nanogranular composites $(CoFeB)_x+(SiO_2)_{1-x}$ are studied, in which the alloy contains, along with metal granules (iron and cobalt), semimetal (boron) granules. Based on a soft ferromagnetic alloy $Co_{40}Fe_{40}B_{20}$ with perpendicular anisotropy the spin valves are in wide demand in electronics [10,11]. Island films of magnetic nanocomposites containing nanoparticles $Co_{41}Fe_{39}B_{20}$ in matrix SiO_2 have gigantic magnetoresistance [12], while the magnetic properties of the nanocomposite are determined by the concentration of ferromagnetic granules x [11].

In this paper we studied composite films $(CoFeB)_x+(SiO_2)_{1-x}$ of 1.11–2.54 μm thick. The films were made at the Voronezh State Technical University by ion-beam sputtering in a nitrogen atmosphere in a low vacuum 10^{-5} Torr on lavsan substrates about 20 μm thick with a ferromagnetic alloy content below percolation threshold ($32.79 < x < 52.00$ at.%). At such concentrations of the ferromagnet, the granules are interconnected by a significant exchange interaction, providing a contribution to the effective anisotropy of the composite [11].

The elemental composition and film thickness were determined using Tescan MIRA 3 LMH SEM electron microscope. The surface topography was studied using Integra Prima atomic force microscope (AFM) (NT-MDT, Russia) in semi-contact mode. The microwave reflection coefficient at normal wave incidence was determined in the frequency ranges 8–12 GHz and 25–36 GHz using the method of measuring the voltage standing-wave ratio (VSWR) in a rectangular waveguide, described in the paper [7]. The dependence of VSWR on frequency was taken from the indicator at least 300 points and entered into the computer memory. Next, the reflection coefficient of microwave waves by power was calculated. To evaluate the magnetic field effect on the reflection coefficient, part of the waveguide with a composite film was placed in the gap of the electromagnet with magnetic field induction of up to 0.3 T. Conductivity was calculated from the reciprocal value of electrical resistance measured at direct current by a two-probe method using a potentiometric method of substitution and taking into account the geometric dimensions of the samples. The main characteristics of composite films are presented in the Table. The sputtering and measurement methods are described in more detail in the paper [7].

Figure 1 shows the surface morphology of the film 4. As can be seen from Figure 1, on the surface there are isolated islands of ferromagnetic alloy of various shapes (the lightest grains) with sizes of hundreds (less often tens) nanometers, along with which there are numerous grains with sizes of several tens of nanometers of the matrix SiO_2, covering the surface evenly. The grains of the metal alloy are located mainly individually, the distances between them are often comparable or greater than their sizes. The spreading resistance microscopy method did not allow us to

Content of the ferromagnetic component of the alloy, layer thickness, electrical resistance and conductivity of composite films $(CoFeB)_x+(SiO_2)_{1-x}$

№	Ferromagnetic component content x, at.%	Layer thickness d (μm), substrate thickness 20 μm	Electrical resistance r (Ω)	Conductivity σ (S/m)
1	32.79	1.73	$8.08 \cdot 10^6$	$3.73 \cdot 10^{-8}$
2	34.02	1.86	$5.50 \cdot 10^6$	$5.03 \cdot 10^{-8}$
3	36.84	1.73	$5.35 \cdot 10^5$	$4.74 \cdot 10^{-7}$
4	41.13	1.83	$1.45 \cdot 10^5$	$1.17 \cdot 10^{-6}$
5	44.42	1.11	$1.17 \cdot 10^4$	$1.13 \cdot 10^{-5}$
6	47.88	2.54	6503	$5.72 \cdot 10^{-5}$
7	49.42	1.73	2864	$7.55 \cdot 10^{-5}$
8	49.84	2.24	2436	$1.18 \cdot 10^{-4}$
9	51.56	2.20	1991	$1.06 \cdot 10^{-4}$
10	52.00	1.59	1659	$2.07 \cdot 10^{-4}$

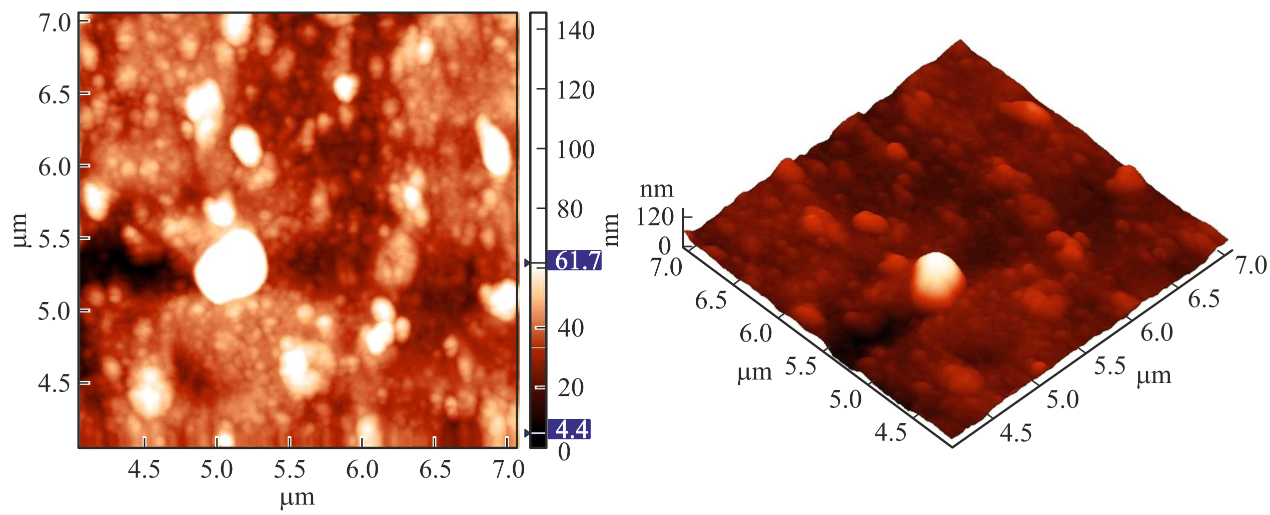

Figure 1. AFM-image of film surface 4 ($x = 41.13$ at.%, $d = 1.83\,\mu$m).

find significant conductive areas on the surface of the films. Point nanoscale areas of conductivity are localized at the tops of the largest particles of the ferromagnetic alloy. These structural features are responsible for the low conductivity of the films (Table).

Figure 2 shows the dependence of the microwave reflection coefficient in the ranges 8–12 GHz and 25–36 GHz on frequency for the most conductive films with $x > 41$ at.%. As can be seen from Figure 2, films in the studied frequency ranges maximum 7–8% of the incident radiation is reflected. The dependences in the frequency range 8–12 GHz (Figure 2, a) are quite smooth, compared to the range 25–36 GHz (Figure 2, b), without pronounced reflection extrema up to $x > 49$ at.%. In the frequency range 25–36 GHz (Figure 2, b), the changes in the reflection coefficient are sharper, and extremes are present for any metal alloy content.

The decrease in reflection coefficient with frequency increasing, observed for both frequency ranges (Figure 2), may be associated with an increase in losses caused by granular currents in the plane of conducting composite films. The presence of deep minima in the reflectance spectra (Figure 2, b) indicates the interference of two electromagnetic waves in antiphase, reflected from the composite film and from the lavsan substrate. This is also confirmed by the fact that the greatest minima are observed for composite films with the maximum metal alloy content (Table).

Figure 3 shows the reflection coefficient vs. metal alloy content (Figure 3, a, in the insert — conductivity vs. metal

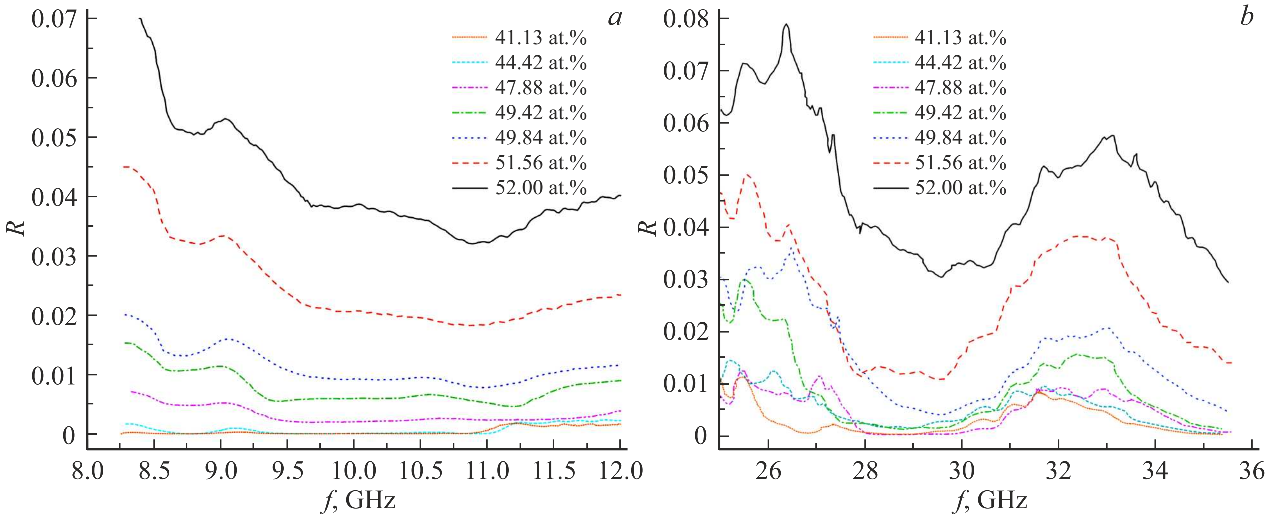

Figure 2. Reflection coefficient vs. frequency: *a*) 8–12 GHz, *b*) 25–36 GHz.

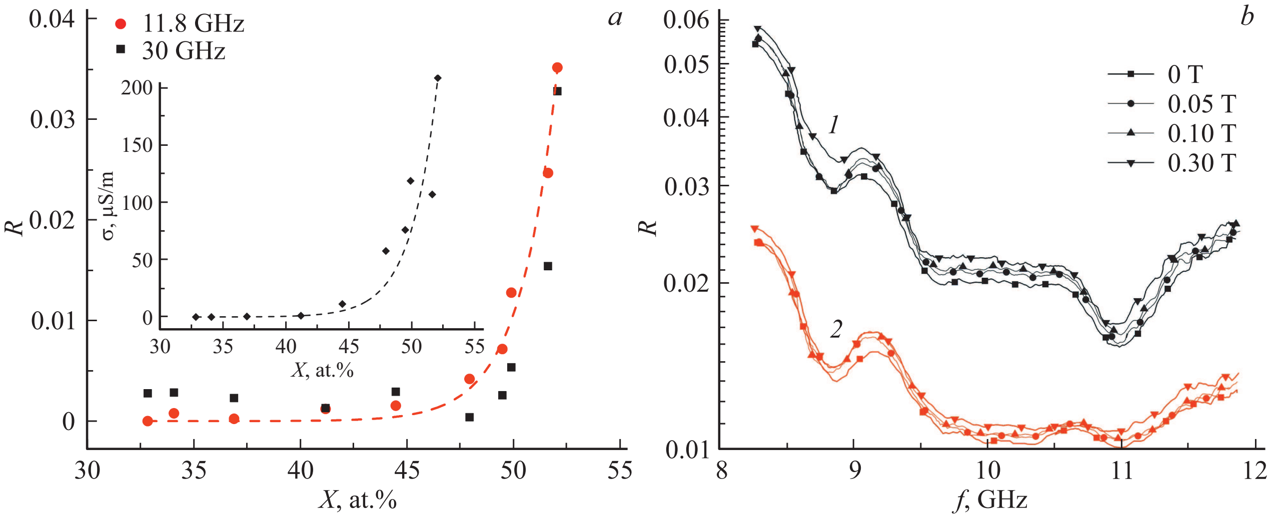

Figure 3. Reflection coefficient vs. the alloy content (in the insert — conductivity vs. alloy content) and vs. frequency in the range 8–12 GHz when exposed to magnetic field with induction up to 0.3 T on films 9 (curves *1*) and 8 (curves *2*).

alloy content) and vs. frequency (Figure 3, *a*,*b*) under the magnetic fields effect on films 9 and 8.

From Figure 3, *a* it is clear that σ and R vs. the alloy content x are qualitatively described by the same law: $y = y_0 + A\exp(x/b)$, where y_0 characterizes the minimum value of conductivity and reflection coefficient, respectively. For the dependence of conductivity: $A = 2.88 \cdot 10^{-15}$, $b = 2.08$, for the dependence of the microwave reflection coefficient at frequency of 11.8 GHz: $A = 3.02 \cdot 10^{-15}$, $b = 1.73$.

A significant increase in conductivity and reflection coefficient in the concentration range of the metal alloy 47–52 at.% can be explained by the percolation process (or metal flow), which is accompanied by a sharp increase in the size of metallic regions [7,9].

From Figure 3, *b* it is clear that, magnetic field effect on the reflection coefficient of microwave waves results in R general increasing as B increases from 0 up to 0.3 T in the entire frequency range under study. The increase in the reflection coefficient may be associated with decrease in resistance with increase in the magnetic field for composite films of the given composition. In this case, the maximum increase R (up to 13% for film 9 and up to 9% for film 8) appears at the frequencies of the first extremes (about 9 GHz) at $B = 0.3$ T. Subsequently, with frequency increasing the influence of the magnetic field on the reflection coefficient becomes less noticeable, and the maximum increase of R does not exceed 10% and 7%, respectively (observed at the minimum reflection at frequency of 11 GHz at field induction 0.3 T).

In this paper the conductive and reflective properties of amorphous nanogranular composites $(\mathrm{CoFeB})_x+(\mathrm{SiO_2})_{1-x}$ are studied. The influence of the magnetic field on the microwave reflection of waves was discovered. For the practical application of this effect it is necessary to carry out further studies on the microwave reflection of composite films at high magnetic fields with induction up to 1 T and with other magnetic metal alloys.

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Conflict of interest

The authors declare that they have no conflict of interest.

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