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Spectra of impedance and dielectric loss tangent in the frequency range 10 Hz–10 MHz and in the temperature range 120–420 K and magnetic structure of composite films (CoFeB+SiO_2)

© M.P. Lasek[1], L.N. Kotov[1], Yu.E. Kalinin[2], A.V. Sitnikov[2]

[1] Syktyvkar State University,
Syktyvkar, Russia
[2] Voronezh State Technical University,
Voronezh, Russia
E-mail: mplasek@yandex.ru

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In this work, temperature and frequency dependences of real and imaginary parts of impedance, relative permittivity and dielectric loss tangent of composite films (CoFeB+SiO_2) were obtained by impedance spectroscopy. The composite films with the metal alloy concentrations $x = 0.62-0.92$ were on a polyethylene terephthalate substrate. The studies were carried out in the frequency range 10 Hz–10 MHz and in the temperature range 120–420 K. Images of surface topography and magnetic phase contrast of the composite films with the concentrations were obtained using an atomic force microscope. Maximum loss tangent was observed at frequencies of 70-120 kHz throughout the entire temperature range for the films with the concentrations from 0.62–0.92, for which a stripe magnetic structure was observed. The real part of the impedance of the composite films with the concentrations of 0.62–0.92 decreases exponentially with increasing the frequency from 10 Hz to 10 MHz throughout the entire temperature range studied.

Keywords: composite metal-dielectric films, magnetic structure, impedance spectroscopy, dielectric loss tangent.

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Composite metal-dielectric films command great attention of scientific researchers due to the wide spectrum of their use, such as magneto-resistive sensors for devices of magnetic data recording and reading, as materials for electromagnet screening and non-linear filters, microelectronics and spintronics [1–8]. Such films have been actively studied for several decades, which is related to their various magnetic, transport, optical and magnetooptical properties. The examples may include giant magnetoresistance, abnormal Hall effect, amplified magneto-optic Kerr effect etc. [2–6]. One of the relevant problems for development of new composite materials with unique properties remains the objective to determine the connection between the structural statistical parameters of the films and their electrodynamic and micromagnetic properties. This circumstance is due to their complex and diverse structure depending on the composition, thickness and method of film manufacturing. It is known that the method of impedance spectroscopy is very informative for the research of various materials [9,10]. At the same time the metal-dielectric composite films have hardly been studied by the method of impedance spectroscopy in a wide range of temperatures.

Composite films studied in this paper were produced by method of ion-beam sputtering of targets from metal plates of alloy $Co_{41}Fe_{39}B_{20}$ and dielectric SiO_2 on a lavsan film with thickness of 30 μm in argon atmosphere at pressure of 26.7 mPa [2]. Alloy $Co_{41}Fe_{39}B_{20}$ is known as material with high magnetic transport and soft magnetic properties [3,4]. Moreover, CoFeB alloys demonstrate higher spin polarization compared to Co, Fe and Ni metals (up to 65% vs. approximately 45%) [5]. The scanning electron microscope TESCAN MIRA 3 (SEM) was used to define shares of atoms of chemical elements and film thicknesses. Using a sharp blade, microscopic particles of the film with the substrate were scraped off the film end on the object table with a double-sided carbon adhesive tape. Using SEM, electronic images of film ends were obtained on the basis of such prepared specimens, and film thicknesses were further measured with the precision of up to the units of nanometers. The analysis of the elemental composition showed that the ratio of Co, Fe atoms in the composite films is close to the ratio of the same atoms in plates made of alloy $Co_{41}Fe_{39}B_{20}$ installed on the targets. The ratio of Si, O atoms is close to the ratio of atoms in SiO_2 plates. The percentage of B boron atoms in composite films, relative to the number of metal alloy atoms varied from 10 to 25%. For the films studied in this paper the concentration of metal alloy atoms varied from 0.62 to 0.92, where x defined the total share of Co, Fe, B atoms contained in composite films. Film thicknesses were in the range from 0.6 to 0.85 μm. Images of the surface relief and magnetic phase contrast were obtained using atomic-force microscope (AFM) INTEGRA Prima (NT-MDT, Russia). The microscope used a silicon probe

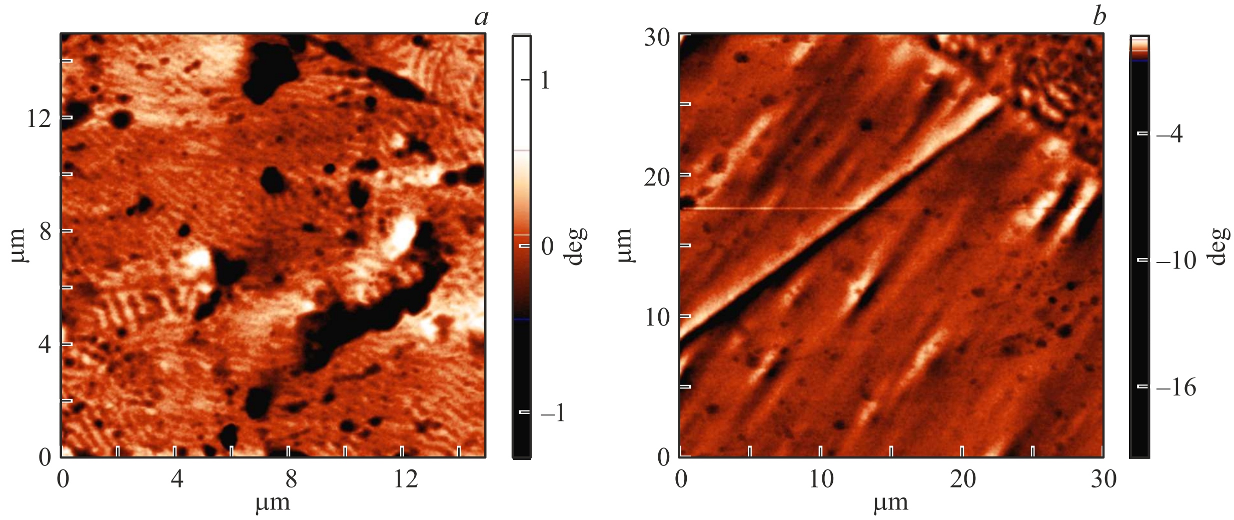

Figure 1. Images of magnetic-phase contrast of the surface of composite films ($CoFeB+SiO_2$) with concentrations of metal alloy $x = 0.64$ (*a*) and 0.90 (*b*). The phase shift values are shown in degrees in color on the right along the vertical scale.

coated with CoCr magnetic alloy with thickness of 40 nm. The probe tip rounding radius was 20 nm. The vibration frequency of the external force that the cantilever with the probe is exposed to, was selected in the frequency range of 47–90 kHz. The microscope recorded the change in the probe vibration phase, which varied depending on the force of the probe interaction with the film surfaces. Temperature and frequency studies of the impedance and loss-angle tangent for the films were carried out in the Center of Diagnostics of Functional Materials for Medicine, Pharmacology and Nanoelectronics of the Scientific Park of the St. Petersburg State University using impedance analyzer ALPHA-AT and cryosystem Quatro in the frequency range from 10 Hz to 10 MHz and in the temperature range from 120 to 420 K. Composite films were installed between the electrodes in the form of a disc with diameter of 8 mm. One electrode was pressed to the surface of the composite layer of the film, and the second one - to the substrate surface.

As a result of the relief and magnetic structure studies using atomic-force microscopy it was found that for the composite films with metal alloy concentrations 0.46–0.49 the granular structure is inherent with granule size 20–50 nm, and the films with concentrations 0.50–0.77 demonstrated the granular-percolation structure [5,6]. For the films with concentration 0.62–0.75 the band magnetic structure is inherent with width of 0.2 μm and band length of over 3 μm (Figure 1, *a*). The structure of composite films with high concentration of metal alloy $x = 0.85{-}0.92$ represented a metal matrix with inclusions of dielectric particles (Figure 1, *b*).

As a result of the completed studies of the films using the impedance spectroscopy method, temperature and frequency dependences were obtained for the real and imaginary parts of impedance, and the loss-angle tangent in the frequency range of 10 Hz–10 MHz in the temperature range of 120–420 K of composite films ($CoFeB+SiO_2$) with concentrations $x = 0.62{-}0.92$ (Figure 2, 3). In the entire studied frequency range the real part of impedance of the composite films with $x = 0.62{-}0.92$ decreased with the growth of frequency in the wide temperature range 120–420 K (Figure 2). The frequency range 10 Hz–1 kHz demonstrated minima of the real part of impedance at temperatures 120 and 300 K for the films with x from 0.62 to 0.92, accordingly. The difference in the positions of temperature minima of the real part of impedance for this films is apparently related to their different microstructure (Figure 1). Frequency dependence of the real part of impedance may be related to the heterogeneous distribution of capacities and resistances in the film volume on the lavsan substrate located between the electrodes [10]. In this case one may use a frequency-dependent element in the equivalent electric scheme of the film between the electrodes, and this element may be compared to the frequency-dependent capacity formed by charge storage and relaxation for the half-period of electric field at the boundaries of the conducting conglomerates separated with a dielectric. Such accumulation of charges will screen the external electric field that will manifest in the experiment as the apparent low-frequency increase in capacity and dielectric permittivity using Maxwell–Wagner mechanism [9]. The absolute value of the imaginary part of impedance in the frequency range from 10 Hz to 1 kHz monotonously rises with the temperature rise and decreases exponentially as the frequency increases for the composite films with $x = 0.62{-}0.92$.

For the films with concentrations of metal alloy from 0.62 to 0.72 in the frequency range from 10 kHz and above, for which the band magnetic structure was observed, a wide maximum was seen at frequency dependences of loss-angle tangent in the entire studied temperature range 120–420 K

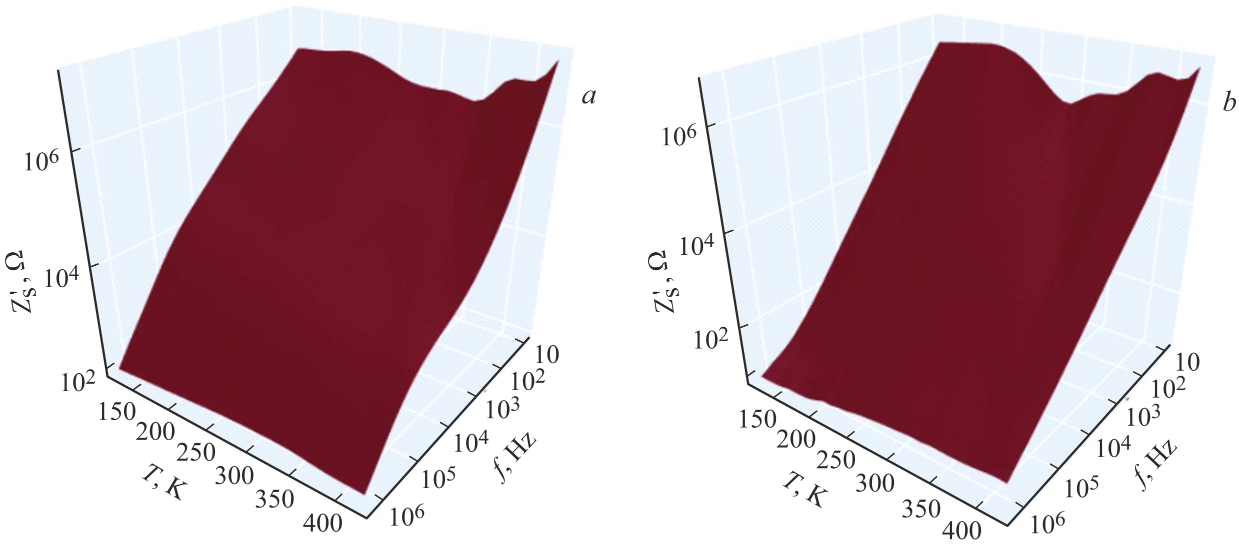

Figure 2. Temperature and frequency dependences of the real part of impedance in composite films ($CoFeB+SiO_2$) with concentrations $x = 0.62$ (a) and 0.88 (b).

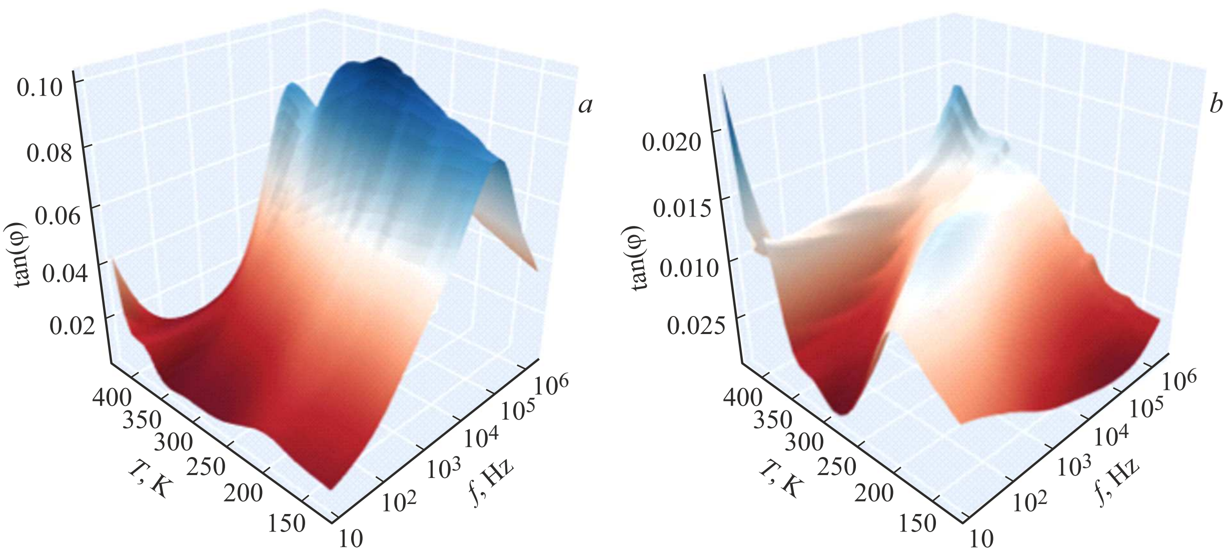

Figure 3. Dependence of loss-angle tangent of composite films ($CoFeB+SiO_2$) on temperature and frequency at concentrations of metal alloy $x = 0.62$ (a) and 0.88 (b).

(Figure 3, a). This temperature maximum of loss-angle tangent moved by frequency from 70 kHz towards high frequencies at temperature rise from 120 to 420 K and increase in concentration x. Additional maxima of loss-angle tangent were observed at temperature dependences of loss-angle tangent at concentrations x from 0.62 to 0.92 at current frequency up to 10 kHz at temperatures 250 and 370 K (Figure 3, b). As frequency increases, these maxima moved towards high temperatures and blended into one peak in the megahertz range of frequencies.

Therefore, this paper used the impedance spectroscopy method to obtain temperature and frequency dependences of loss-angle tangent, real and imaginary parts of impedance of composite films ($CoFeB+SiO_2$) on a lavsan substrate with metal alloy concentrations 0.62–0.92 in the range of frequencies 10 Hz–10 MHz and in the temperature range 120–420 K. Analysis of MPC images shows that for composite metal-dielectric films with low concentrations $x < 0.62$ the granular-percolation structure is specific, and for $x = 0.62–0.75$ - band magnetic structure, and a metal matrix with dielectric inclusions is observed for films with large $x > 0.85$. Analysis of temperature and frequency dependences of the real part of impedance and loss-angle tangent shows that the behavior of these dependences differs greatly for the composite films with various microstructure. The composite film study results in this paper show that the impedance spectroscopy is rather informative for research of the electric properties and detection of the correlation between these properties and the structure of composite metal-dielectric films on a polymer substrate, prior to range expansion to the microwave range, would help to improve the quality of research.

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Conflict of interest

The authors declare that they have no conflict of interest.

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