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Concentration dependences of ferromagnetic resonance parameters of CoFeB/SiO$_2$ composite films for various angles between the direction of the magnetic field and the plane of the films

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The concentration dependences of the resonant field and the width of the ferromagnetic resonance line (FMR) are obtained at different angles φ between the direction of the magnetic field and the plane of the films for two series of composite films CoFeB/SiO$_2$ obtained under different spraying conditions. The films of the first series had a mainly granular structure, while the films of the second series had a granular-percolation structure, which, at high concentrations of metals x, included a strip magneto-metal structure. It is shown that the resonance field for films of both series decreases with increasing concentrations of x. The resonant field and the width of the FMR line increase with increasing angle φ for films of both series with different structures. It is demonstrated that the FMR method is a structurally sensitive method for studying composite films.

Keywords: Composite metal-dielectric films, ferromagnetic resonance, structure.

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Composite metal-dielectric films are of great interest to researchers, since their electrical, magnetic, and electromagnetic properties can vary very widely with changes in composition and deposition conditions [1–5]. Composite films can be used as a basis for the creation of sensors and biosensors, due to their high sensitivity to electromagnetic fields [6–10]. In this paper, ultrahigh-frequency (microwave) electromagnetic properties and the effect of film structure on them have been studied using the ferromagnetic resonance (FMR) method. In this work, the concentration dependences of the resonant field Br and the width of the FMR line ΔB were studied at different angles φ between the direction of the permanent magnetic field and the plane of composite films (CoFeB+SiO$_2$) with different structures.

Composite films were obtained by ion bombardment of targets consisting of a metal alloy $Co_{0.44}Fe_{0.36}B_{0.2}$ and a dielectric SiO$_2$, followed by deposition of dislodged particles onto a dacron sheet with dimensions $210\times290\,mm^2$ [1–3]. Films of two series were obtained under different spraying conditions. Films of the series I with total concentrations of Co, Fe metals of $x = 31-48.7$ at.% were obtained in an argon atmosphere with a pressure of $1.35\,\mu$Pa and in a nitrogen atmosphere with a pressure of $0.2\,\mu$Pa. Films of the series II with concentrations of $x = 44.3-83.2$ at.% were obtained in an argon atmosphere with a pressure of $1.4\,\mu$Pa. The targets were bombarded with lighter argon ions for sputtering the films of both series. The addition of nitrogen to the chamber during sputtering of I series films made it possible to obtain granular composite films in a wide range of concentrations x, even beyond the percolation range for II series films. The structure of the studied composite films is divided into granular (G), granular-percolation (GP) with the inclusion of magneto-metal strips (GPMP) [2]. Tables 1, 2 show thicknesses d and concentrations of elements, the total concentration of metals Co, Fe x in at.% and the structure of composite films of two series I and II. To study films by the FMR method, film pieces with dimensions $5\times3\,mm^2$ were cut from a dacron sheet with a sprayed composite layer.

To study the surface of films using an atomic force microscope (AFM), samples with dimensions $2\times2\,mm^2$ were cut from the same sheet [1,2]. Studies of the relief and magnetic phase contrast of the film surface were carried out using an NTEGRA Prima atomic force microscope (NT-MDT, Russia) with a cantilever and an MFM10 magnetic probe. The silicon probe was coated with a magnetic CoCr alloy with a thickness of 40 nm and a radius of curvature of the probe tip up to 20 nm. Magnetic cantilevers of the MFM10 series had a force constant of $1-5$ N/m, and the spatial resolution of AFM with such cantilevers was 30 nm. The oscillation frequency of the external force acting on the cantilever with the probe was selected in the range $47-90$ kHz [2]. The phase difference $\Delta\beta$ of the oscillations of the cantilever with the probe was recorded in a microscope, which varied depending on the strength of the interaction of the magnetic probe with the magnetic field arising above the surface of the films. The attraction and repulsion of the probe from the film surface

Table 1. Compositions, thicknesses and structure of composite films of the series I

d, μm	x	Co	Fe	B	O	Si	N	Structure	№
1.51	31.0	17.3	13.7	1.7	44.2	14.7	8.9	G	1
2.15	32.2	18.0	14.2	2.7	43.3	14.3	8.5	G	2
1.85	38.0	21.4	16.6	4.3	37.8	12.4	8.4	GP	3
1.22	43.9	24.5	19.4	1.7	38.7	9.2	7.4	GP	4
1.55	46.2	26.2	20.0	5.8	33.1	9.7	7.7	GP	5
1.53	48.7	27.4	21.3	3.0	32.6	9.4	8.4	GP	6

Table 2. Compositions, thicknesses and structure of composite films of the series II

d, nm	x	Co	Fe	O	Si	Structure	№
514	44.3	23.6	20.7	38.2	17.5	GP	1
504	45.5	22.2	23.3	37.1	17.4	GP	2
524	49.3	26.3	23.0	34.3	16.4	GP	3
529	56.2	29.0	27.2	28.15	15.57	GPMP	4
534	63.7	34.0	29.7	24.1	12.3	GPMP	5
634	64.0	34.1	29.9	23.7	12.3	GPMP	6
733	67.3	36.3	32.0	21.0	11.2	GPMP	7
796	76.7	41.0	35.7	14.9	8.4	GPMP	8
859	80.7	43.2	37.5	12.2	7.2	GPMP	9
774	83.2	44.9	38.3	10.4	6.4	–	10

area corresponded to different signs $\Delta\beta$. The dependence of $\Delta\beta$ on the coordinates of the film surface, obtained during the first penetration of the magnetic probe over the film surface, was transformed into a relief image showing the sizes and heights of homogeneous regions. The magnetic phase contrast (MPC) image, characterizing the induction of a magnetic field over the film surface, was created after the second penetration of a magnetic probe over the film surface and with the values $\Delta\beta$ subtracted, displaying the relief of the film surface. Figure 1 shows images of the magnetic phase contrast of composite films of series I and II. As can be seen from Figure 1,a the film of the series I at $x = 31$ at.% has a granular structure, and the film with $x = 48.7$ at.% has granular-percolation structure. Films of the series II with $x = 44.1$ at.% have a granular percolation structure (Figure 1,c), and a film with a high concentration of metals $x = 67.3$ at.% has magneto-metal bands with a length of 3 μm and a width of 0.2–0.3 μm, which included a granular structure.

The FMR parameters of composite films were studied using a RE-1306 radio spectrometer at an electromagnetic field frequency of 9.36 GHz at different angles φ between the direction of the alternating magnetic field and the film plane, which varied in the range from 0 to 50° in increments of 10° [2]. The maximum error in determining the angle of φ in experiments did not exceed 10 %. At angles greater than 50°, conducting experiments for composite films of the series II was difficult because there were no signals from the sample, most likely due to the strong absorption of the electromagnetic field by the films. In the process of measuring FMR spectra, the radio spectrometer receiver recorded a signal proportional to the derivative of the magnetic field of the power absorbed by the sample. Based on the resonant lines recorded on the recorder, the values of the line position (resonant field B_r) and the width of the resonant line ΔB were determined. The maximum error in determining the resonant field B_r and the width of the FMR line ΔB did not exceed 5 %. At the initial angle $\varphi = 0°$, the constant and alternating magnetic fields were perpendicular to each other and lay in the plane of the film.

Let us analyze the behavior of the concentration dependences of the FMR parameters of the studied samples at small angles $\varphi = 0$–$30°$. The resonance field B_r for composite films of both series, as can be seen from Figure 2, decreases with increasing metal concentration x. Taking into account the rate of decrease of B_r from the concentration of metals x, the entire concentration range can be divided into two regions. The first region with a rapid decrease in the resonance field B_r with an increase in concentration x for films of the series I is the range of $x = 32$–44 at.%, and this is a range of $x = 44$–49 at.% for films of series II. Films of the series I and II with such x are characterized by a granular structure with a small volume of percolation or metallic regions (Figure 1,a, c). The second concentration range x with a weaker change in function $B_r(x)$ for films of the series I has a range of 44–49 at.%, and this is a range of $x = 60$–80 at.% for films of series II. It should be noted that composite films of the series I with such x are characterized by a granular percolation structure with a large volume of extended metal regions (Figure 1,b), and the metal areas are long metal bands for films of the series II with $x > 64$ at.% (Figure 1,d). A maximum is observed in the range of $x = 37$–43 at.% at large angles $\varphi = 40°, 50°$ on the dependencies $B_r(x)$ for films of the series I. In composite films with such x, the size of percolation (metallic) regions increases with an increase of x. As the angle φ increases from 0° to 50°, the resonant field B_r increases for films of both series for the entire range of x, but the field B_r grows especially rapidly at $x = 38$ at.% for films of the series I, and at $x = 58$ at.% for films of the series II series.

Let us discuss the behavior of the concentration dependences of the resonance line width ΔB for the studied composite films. As can be seen from Figure 3, for $\varphi = 0°$ composite films of the series I with a granular structure, the line width of ΔB increases with an increase of x from 31 to 35 at.%. At high concentrations of metals, the line width ΔB with increasing x decreases for films of series I at $x = 36$–49 at.% and at $x = 44$–64 at.% for films of the series II. For films of the series II with a metal strip structure, a section of the line width ΔB increases with an increase of x from 65 to 80 at.%. Two wide maxima are observed for composite films of both series on the concentration dependences $\Delta B(x)$ at large angles $\varphi = 40°, 50°$. In addition, another maximum is observed at low $x = 46$ at.% at $\varphi = 40°, 50°$ on the concentration

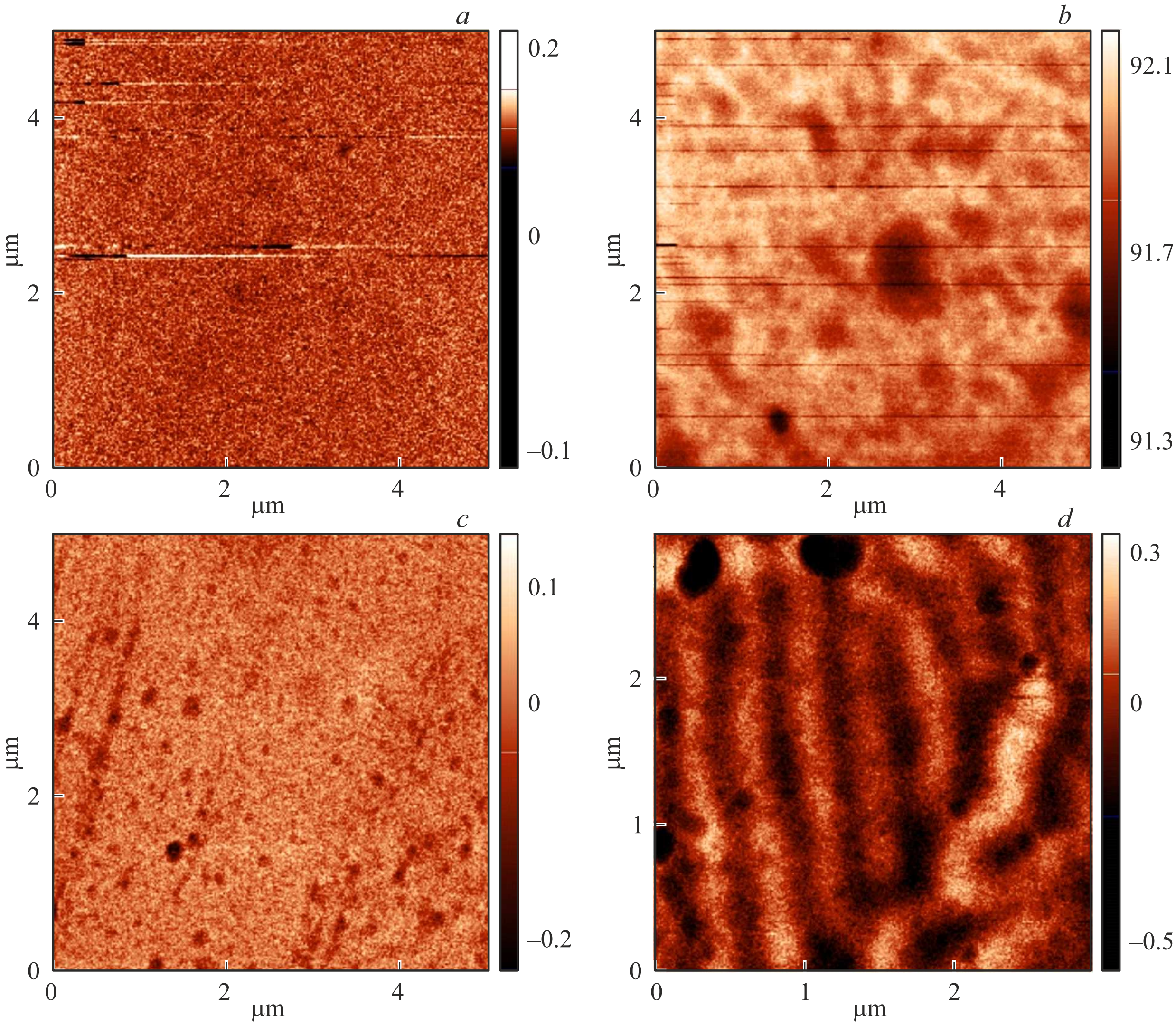

Figure 1. Magnetic phase contrast (MPC) images of composite films of the series I at $x = 31$ at.%) (a), 48.7 (b) and series II at $x = 44.3$ at.% (c), 67.3 (d). The values of the phase difference $\Delta\beta$ of the cantilever oscillations are shown on the color scale to the right of the MPC image.

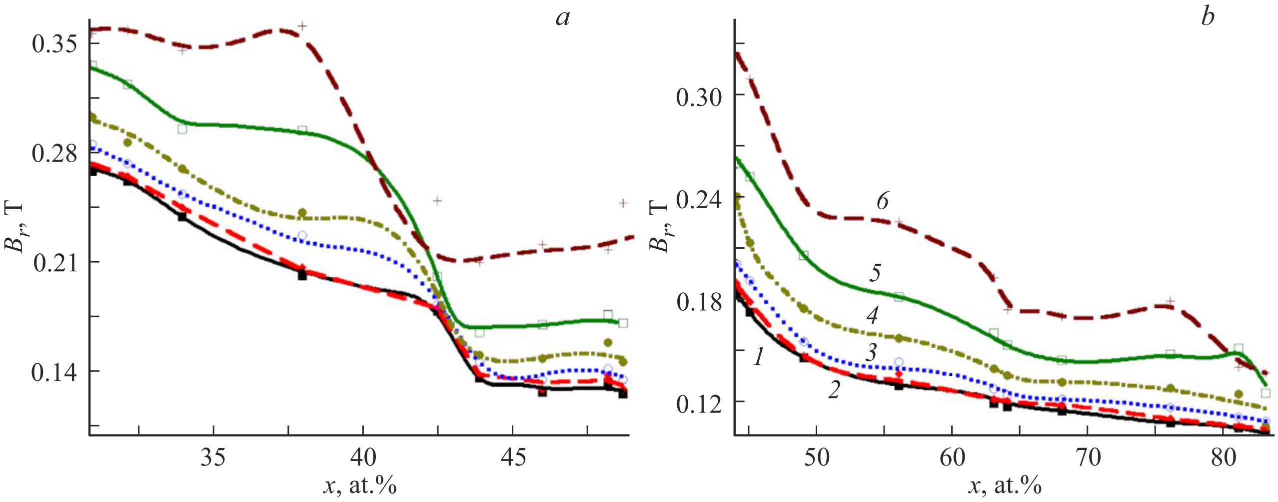

Figure 2. Concentration dependences of the resonance field B_r for composite films of the series I (a) and II (b) for angles $\varphi = 0°$ — *1*, 10° — *2*, 20° — *3*, 30° — *4*, 40° — *5*, 50° — *6*. $T = 300$ K.

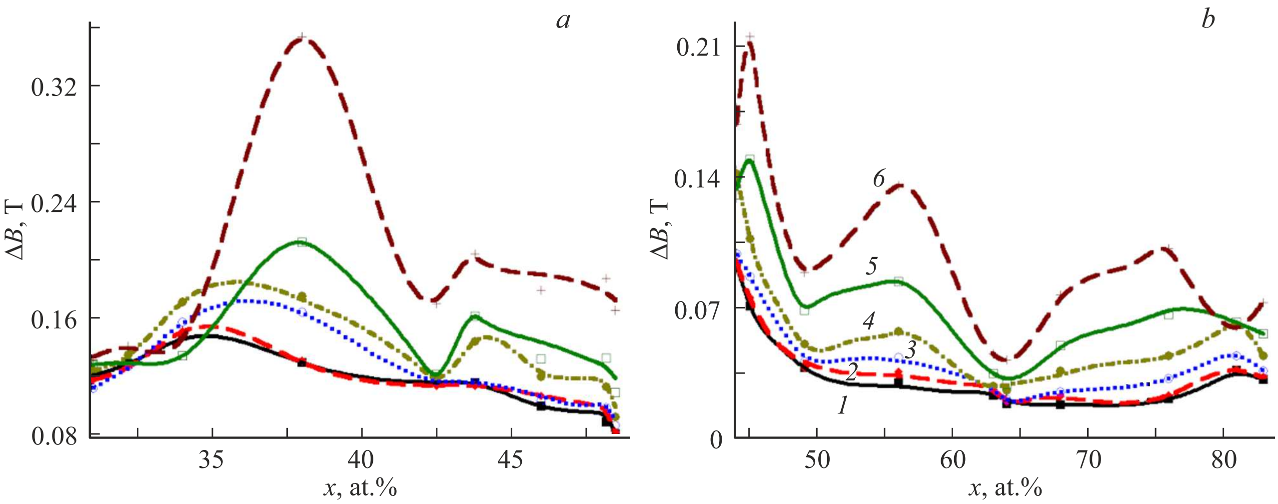

Figure 3. Concentration dependences of the line width ΔB for composite films of the series *I* (*a*) and *II* (*b*) for angles $\varphi = 0^\circ$ — 1, 10° — *2*, 20° — *3*, 30° — *4*, 40° — *5*, 50° — *6*. $T = 300$ K.

dependences $\Delta B(x)$ for composite films of the series *II*. As can be seen from Figures 2, 3, as the angle φ increases from 0° to 50°, the values of the resonant field B_r and the line width ΔB increase for all composite films with different metal concentrations x and with different structures, but the growth rate of the FMR parameters from the angle φ strongly depends on the concentration x and the structure of the films. For example, for films of the series *I* with a granular structure at $x = 38$ at.%, the resonant field of B_r increases from 0.2 to 0.36 T, and the line width of ΔB increases from 0.12 to 0.34 T as the angle increases φ from 0° to 50°. At the same time, the value of B_r increases from 0.11 to 0.2 T with an increase in the angle of φ from 0° to 50° for films of the series *II* with a band magneto-metal structure at $x = 75$ at.%, and the line width ΔB increases from 0.025 to 0.1 T. Such different changes in the resonance field B_r and line width ΔB for films with different structures under the same change in angle φ from 0° to 50° indicate that, along with the demagnetizing fields due to the shape and dimensions of the films, the demagnetizing field arising from the internal structure of the films plays a significant role [3]. In the case where the alternating magnetic field lies in the plane of the film ($\varphi = 0^\circ$), the main contribution to the magnitude of the resonant field B_r and the line width ΔB is provided by the demagnetizing field due to the internal structure of the films. At large angles $\varphi = 30^\circ - 50^\circ$, the contribution of the demagnetizing field due to the shape and size of composite films begins to prevail in B_r and ΔB.

Conclusion

In the course of the conducted studies, patterns of behavior of the resonant field B_r and the width of the ferromagnetic resonance line (FMR) ΔB were revealed depending on the total concentration of Co, Fe metals and the film structure, as well as on the angle between the external permanent magnetic field and the film plane. The research results obtained in this paper show that the FMR parameters of composite metal-dielectric films are significantly influenced by the structure and size of percolation (metallic) regions. The composition of films also affects the FMR parameters, but indirectly, mainly through the film structure, since the composition and deposition conditions determine the structure of composite films, including the concentration range x, at which the phenomenon of percolation occurs during film deposition. As the analysis of the results of studies of FMR parameters and film structure shows, the FMR method is a structurally sensitive method for studying the electromagnetic properties of composite films. Further more detailed experimental studies of the FMR parameters of magnetic composite films using a radio spectrometer and finding a functional relationship between the FMR parameters and the film structure (the size and shape of granules and percolation regions) will make it possible to develop a technique for obtaining images of micro- and nanostructure films.

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Conflict of interest

The authors declare that they have no conflict of interest.

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