08

Concentration dependences of ferromagnetic resonance parameters and structure of composite films ($CoFeB+SiO_2+N_2$), obtained in nitrogen atmosphere

© V.A. Ustyugov[1], L.N. Kotov[1], P.D. Kovalev[1], Yu.E. Kalinin[2], A.V. Sitnikov[2]

[1] Syktyvkar State University,
Syktyvkar, Russia
[2] Voronezh State Technical University,
Voronezh, Russia

E-mail: ustyugovva@gmail.com

Received April 30, 2024
Revised October 28, 2024
Accepted October 30, 2024

The paper presents the results of statistical analysis of magneto-phase contrast images of [$CoFeB+SiO_2+N_2$] composite films sputtered in nitrogen atmosphere. The relationship of structural features (distribution of granules by effective size) with the concentration of metallic phase is shown. The data on concentration dependences of the position and width of the ferromagnetic resonance line are presented. Using a mathematical model, the relationship between the structural and magnetic characteristics of the investigated composite films is shown.

Keywords: sputtering of films in nitrogen atmosphere, granular and granular-percolation structure, ferromagnetic resonance.

DOI: 10.61011/PSS.2024.12.60188.6516PA

Pelleted composite films command great attention of scientific researchers due to a wide spectrum of their applications (non-linear filters, spintronics etc.) and wide capabilities in the field of mathematical and computer modeling of electromagnet and structural properties within approximation of micromagnetism [1–5]. However, building the structural models of pelleted metal-dielectric composite films making it possible to describe them as solid media [3], is usually coupled with difficulties in definition of statistical distributions of geometric parameters of pellets within a film, especially at high concentrations of a metal alloy. The critical issue in the field of searching for new compositions and possible applications of magnetic composite films is the research of the interconnection in the structural statistical parameters of the films and their microwave magnetic characteristics. This paper studies the parameters of the structure of composite pelleted films with composition of ($CoFeB+SiO_2+N_2$) and thicknesses $1.0-2.1\,\mu$m and concentrations of metal alloy $x = 0.39-0.59$, made by method of ion-beam sputtering in argon atmosphere with pressure of 0.24 Pa with addition of nitrogen at pressure of 0.035 Pa. When argon ions bombard targets of metal alloy $Co_{41}Fe_{39}B_{20}$ and dielectric SiO_2, composite films were sputtered on lavsan sheets with size of $210 \times 297\,\text{mm}^2$. Thicknesses and chemical composition of specimens were defined using scanning electron microscope TESCAN MIRA3. Research of the surface relief and magnetic structure of film surface was carried out using atomic-force microscope NTEGRA PRIMA (NT-MDT, Russia). Ferro-magnetic resonance (FMR) spectra of the films were produced on radiospectrometer RE-1306 at AC field frequency of 9.4 GHz. DC magnetic field was directed perpendicularly to the AC magnetic field and lied in the plane of the film.

The paper [6] shows that the presence of nitrogen used in process of sputtering causes appearance of non-magnetic nitrides in the film volume and isolation of magnetic pellets due to nitrogen pore formation. The last circumstance causes higher resistance of the granular structure of the films in the wide interval of concentrations x, which simplifies the internal structure of the films and accordingly the development of the theoretical model for the relation between the structural and magnetic characteristics for this chemical composition. The chemical composition of the studied specimens is given in the table. All specimens maintain close ratio of cobalt and iron concentrations in the same manner as for the target, which means high-quality manufacturing of the films. Also note that the concentration of nitrogen atoms will not increase with the growth of metal alloy concentration, which may indicate the absence of nitrides in the film substance. Adsorbed nitrogen provides for pores between metal particles and supports granular structure formation.

The study of the images of the relief and magnet-phase contrast of composite films produced with the help of atomic-force microscope (AFM) shows that the selected chemical composition in a combination with the sputtering of films in the atmosphere of nitrogen makes it possible to produce a pelleted structure in the higher volume of the film and for larger intervals of concentrations, compared to the previously studied composite films [3,7]. Figure 1 shows the imposition of black contours of topographic elevations of the

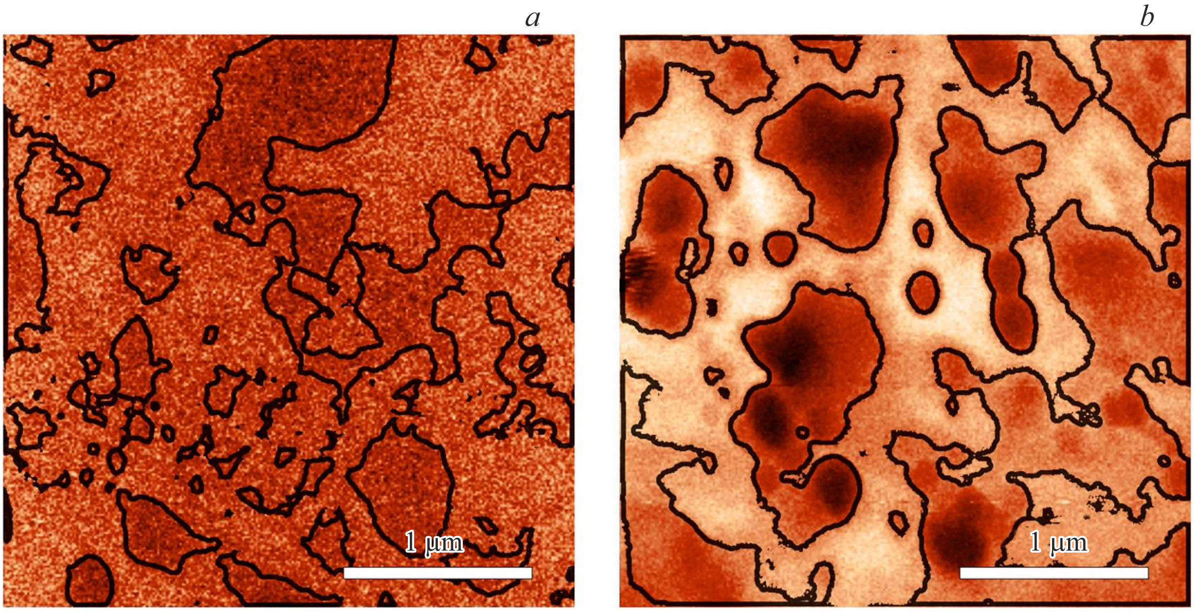

Figure 1. Topographic and magnetic structure (contours of topographic elevations are marked with a black line) of the films [$CoFeB+SiO_2+N_2$] at concentration of metal alloy $x = 0.4$ (*a*) and 0.6 (*b*).

Chemical composition of composite films [$CoFeB+SiO_2+N_2$]

	Co, at.%	Fe, at.%	B, at.%	Si, at.%	O, at.%	N, at.%	x, at.%
1	31.58	24.48	3.53	11.15	22.31	6.94	59.60
2	29.80	23.50	4.12	11.02	22.05	9.50	57.43
3	27.67	21.52	6.18	11.74	23.48	9.41	55.37
4	23.82	19.11	4.14	14.48	28.96	9.48	47.07
5	21.91	17.50	3.11	16.04	32.08	9.37	42.52
6	21.18	16.71	2.13	16.65	33.30	10.04	40.01
7	20.58	16.32	2.05	16.86	33.72	10.47	38.95

film surface upon the image of magnetic contrast areas for metal alloy concentrations $x = 0.4$ and $x = 0.6$. One can see that in both cases there is a clear granular structure on the surface, the pellet dimensions are comparable, but at higher metal concentration the pellets have more regular shape with predominance of the large size. Using magnetic contrast images, one may state that at high metal concentration the topographic elevations are matched with specifically metal areas with high magnetization. At low concentration x the magnetic structure is hardly seen.

Automated analysis of atomic-force microscopy images of composite films at various scales was conducted using free software Gwyddion. The error of detection of average radii for the pellets automatically identified on the image (pellets were approximated by ellipsoids) was $\sim 5\%$. The study showed that the specific feature of the specimens is the preservation of the parameters of statistical distributions of pellets at various metal concentrations. Small pellets grow together into large aggregates, but their melting into complex labyrinth-like structures is prevented.

Figure 2, *a* shows distribution of relative density of surface point heights that characterizes roughness. From the curve one can see that specimens with metal alloy concentration $x = 0.4$ are characterized by high homogeneity of the surface (narrow distribution peak), besides, the most probable height of the point on the film surface is $h = 20$ nm above the selected zero level. For the same metal concentration Figure 2, *b* shows distribution of pellets on the film surface by dimensions (shapes of pellets are approximated by ellipsoids), besides, the most probable value of particle diameter was $d = 18$ nm. Therefore, pellets on the film surface may be successfully described as rotation ellipsoids, close by the ratio of semi-axes to the sphere. As metal concentration increases ($x = 0.6$), as one can see also from Figure 2, minor displacement of distributions occurs towards larger particles, points with heights appear on the surface that have not been observed previously. However, no radical changes are traced in the granular structure of the films visually or by results of automated analysis. This makes the films of this composition a convenient tool to build and verify the mathematical models of relation between structural and magnetic properties.

The study of magnetic characteristics of the composite films found (Figure 3) that width of the ferro-magnetic resonance line at room temperature hardly depends on metal concentration, since the main effect on the line width is provided by the spread of demagnetizing factors of individual pellets, which, as shown above, hardly depends on concentration. However, as the concentration x increases,

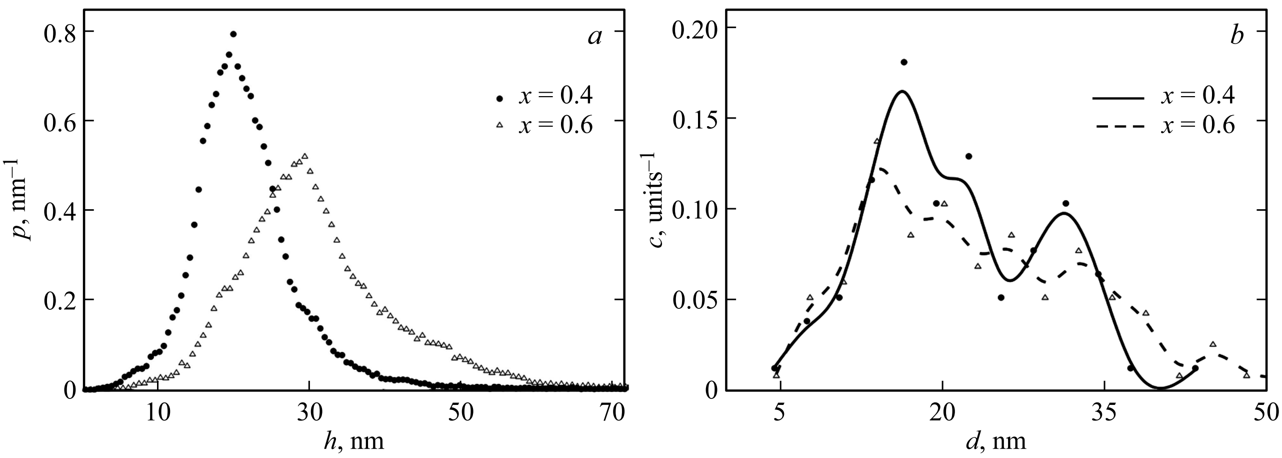

Figure 2. Distribution of relative density of surface point height density (a) and effective pellet sizes (b) on surface of films [$CoFeB+SiO_2+N_2$] at different concentrations of metal alloy.

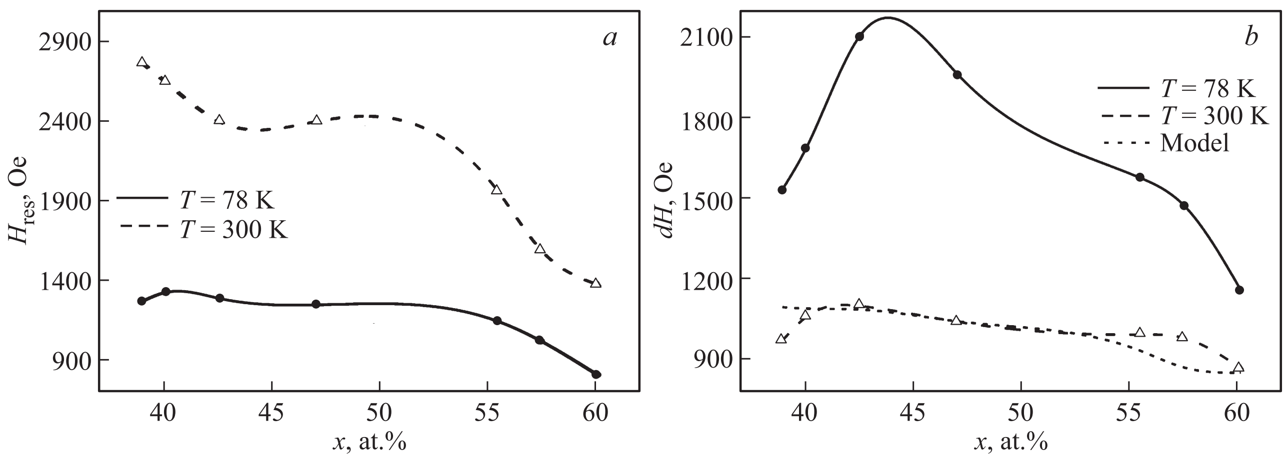

Figure 3. Dependence of position (a) and width (b) of FMR line in films [$CoFeB+SiO_2+N_2$] on concentration of metal alloy at temperatures $T = 300$, 77 K.

minor monotonous decrease of FMR line width is noted, which may be related to increase in homogeneity of the substance and approximation of pellet shape to spherical shape, which is also fixed by the images of film AFM relief. The study of the FMR line width in the specimens at liquid nitrogen temperature $T = 77$ K (Figure 3, b) makes it possible to record the structural phase transition at metal alloy concentration $x = 0.45$. At this temperature the length of free path of electrons in the substance between film pellets increases substantially, as a result of which the exchange interaction between pellets increases. This provides for decrease of FMR line width, starting from the specified metal concentration.

The position of the FMR resonant field peak (Figure 3, a) also shows the presence of structural transition at concentration of around $x = 0.45$. Note that in virtue of the fact that metal pellets in the case of the studied chemical composition would not unite, this structural transition may not be treated as percolation transition, since it manifests itself only in respect to statistical characteristics of pellets on the film surface.

Let us estimate the contribution of the random structure of particle (pellet) ensemble on the film surface along the FMR line width. According to the known method [3], this contribution is

$$\Delta H_{\text{por}} = 8\pi M_s \big(\lambda/(1+\lambda)\big),$$

where M_s — material saturation magnetization, $\lambda = V_d/(V_d + V_m)$ — porosity, V_d and V_m — volume of dielectric and metal alloy, accordingly. The FMR line width calculated according to this model is shown in Figure 3, b as a fine dotted line. One can see that there is good compliance with the result of the experiment in the studied interval of metal concentrations. At lower concentrations of metal alloy x the model will not yield a positive result due to significant spread of distances between the particles on the film surface and their dimensions. At high metal concentrations the number of pellets in the film volume becomes so high that many of them start contacting with each other. At the same time the so called granular-percolation structure arises, when no melting of pellets into a single metal matrix occurs, however, the electron energy is such that they may overcome the potential barrier between pellets.

To conclude, one can note that the paper studied structural statistical characteristics of composite metal-dielectric films with composition [$CoFeB+SiO_2+N_2$], sputtered in nitrogen atmosphere. It was found that in the studied interval of metal phase concentrations x the distributions of specimen pellets by effective dimensions hardly depend on x. The relation was identified between microwave magnetic characteristics of the films and their granular and granular-percolation structure. A structural phase transition was found when specimens were cooled down to 77 K for the films with metal alloy concentration $x = 0.45$.

Funding

The study was supported by a grant from the Russian Science Foundation (project No. 21-72-20048).

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] P.G. Baranov, A.M. Kalashnikov, V.I. Kozub, V.L. Korenev, Yu.G. Kusraev, R.V. Pisarev, V.F. Sapega, I.A. Akimov, M. Bayer, A.V. Scherbakov, D.R. Yakovlev. UFN **189**, *8*, 849–880 (2019). (in Russian).

[2] S. Bandyopadhyay, J. Atulasimha, A. Barman. Appl. Phys. Rev. **8**, *4*, 041323 (2021).

[3] L.N. Kotov, V.A. Ustyugov, V.S. Vlasov, A.A. Utkin, Yu.E. Kalinin, A.V. Sitnikov. Bull. Russ. Acad. Sci.: Phys. **87**, *3*, 385–388 (2023). DOI: 10.3103/S1062873822701283

[4] T. Parpiiev, A. Hillion, V. Vlasov, V. Gusev, K. Dumesnil, T. Hauet, S. Andrieu, A. Anane, T. Pezeril. Phys. Rev. B **104**, *22*, 224426 (2021). DOI: 10.1103/PhysRevB.104.224426

[5] V.V. Rylkov, A.V. Emelyanov, S.N. Nikolaev, K.E. Nikiruy, A.V. Sitnikov, E.A. Fadeev, V.A. Demin, A.B. Granovsky, J. Exp. Theor. Phys. **131**, 160–176 (2020). DOI: 10.1134/S1063776120070109

[6] B.A. Belyaev, V.V. Tyurnev. J. Exp. Theor. Phys. **127**, *4*, 608–619 (2018). DOI: 10.1134/S1063776118100114

[7] L.N. Kotov, V.S. Vlasov P.D. Kovalev, A.A. Utkin, Yu.E. Kalinin, A.V. Sitnikov, V.A. Ustyugov. 2023 International Conference on Next Generation Electronics (NEleX), Vellore, India, 2023, pp. 1–5. DOI: 10.1109/NEleX59773.2023.10420863

Translated by M.Verenikina