Relief, magnetic structure and microwave properties of composite films

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Abstract. Relief, magnetic structure and microwave properties of the composite (metal-dielectric) films: (CoFeZr)\textsubscript{x}(Al\textsubscript{2}O\textsubscript{3})\textsubscript{y}, (A series) and (CoTaNb)\textsubscript{x}(SiO\textsubscript{2})\textsubscript{y}, (B series) were investigated. Experimental data on the microwave magnetic characteristics were also calculated on the basis Dubowik model.

Introduction
In present time the magnetic, magnetooptical and magnetoacoustic properties of thin composite films are widely investigated. The attention in this area is caused by opportunity of new information processing devices building (spintronics), rf tuning filters. Also we have the interest to unusual behavior of the magnetization, the domain structure formation, propagation of the electromagnetic waves in these nanosized objects.

The metal-dielectric composite films describing in the work have essential nonhomogeneous structure. The parameters of the structure depend on the metal/dielectric ratio. We can reveal the regularities of the average particles size changes in the atomic force microscopy topographic images, the regularities of the particle size distribution and the films thickness changes with varying metal/dielectric ratio.

The metal phase of the films is super-paramagnetic or ferromagnetic depending on the films thickness. This fact makes difficult the analysis of the microwave properties. We develop the known model for describe the composite structures consisting of separate granules and bigger labyrinth formations. These formations appear at big metal concentrations by convergence of the small particles.

Object and investigation methods
The two series of the films (Co\textsubscript{0.4}Fe\textsubscript{0.6}Zr\textsubscript{0.1})\textsubscript{x}(Al\textsubscript{2}O\textsubscript{3})\textsubscript{y}, where 0.25<x<0.65 (A series) and (Co\textsubscript{86}Ta\textsubscript{12}Nb\textsubscript{2})\textsubscript{x}(SiO\textsubscript{2})\textsubscript{y}, where 0.3<x<0.6 (B series) were investigated. The percolation threshold for these series was about x=0.45. The investigated films were prepared by ion beam sputtering method on the polycrystalline glass substrate. For preparing of the films the alloyed metal target with size 270×70×14 mm\textsuperscript{3} were applied. The dielectric plates with size 70×9×2 mm\textsuperscript{3} were fixed with changing interval on the targets. It gives us in one technological process to obtain the samples with continuous changes of the metal/dielectric ratio. We used the argon in the chamber with pressure 8×10\textsuperscript{-5} Torr.
The films were amorphous. The ferromagnetic phase in the films is localized in the granules and doesn’t form compounds with dielectric. The chemical composition and thickness of the films were determined using a scanning electron microscope JSM-6400. For investigation of the films structure the scanning of the films surface was performed by the atomic force microscope (AFM) ARIS-3500 (Burleigh Instrument Co, USA) with maximal scanning area 70×70 μm². The standard pyramidal shape silicon cantilevers with nose rounded radius ≈ 10 nm and stiffness 0.1 N/m were used. We made the calibration of the image scale by certificated test specimens.

**Thopography and phase contrast of amorphous composite films**

Let’s consider topographic imaging of the films (Co₄₅Fe₄₅Zr₁₀(Al₂O₃)₁₋ₓ) at the different metal phase concentrations. The typical imaging shown on the fig. 1.

**Figure 1.** Thopographical and phase imaging of the A series of the composite films: x = 0.3 (a, c) and x = 0.6 (b, d)

The Fig. 1 shows essential nonhomogeneous structure of the films. We can’t select separate granules in this structure. It is right for the small and big metal concentrations. But the relief of the film is less nonhomogeneous at big metal concentration and we can extract arbitrary big rolls-and-swells. This
The fact shows more homogeneous growth of the film after percolation threshold [3]. The bright white blobs on the figures correspond to film defects and we can exclude the blobs from consideration.

![Figure 2](image)

**Figure 2.** Topographical and phase imaging of the B series of the composite films: \( x = 0.35 \) (a, c) and \( x = 0.55 \) (b, d)

We can see that average particles size increases with growth of metal concentration comparing the topographical images before and after percolation threshold. This is due to increase of the common homogeneity of the chemical films compound.

We can see also that metal and dielectric don't form the granules, the components have homogeneous distribution in the film volume.

Let's consider the peculiarities of the topographical imaging for B series of the films (fig. 2). The surface of the B series of the films is similar to the surface of the A series of the films at small metal concentration and show the rolls-and-swells matrix. However, at big metal concentration we can see granular objects with elliptic shape and 1.5 nm size. We connect this fact with more homogeneous compound for the B series of the films. The basic component of the B series of the films is Co with adding of small amount of the other components which give amorphous condition.

We can find the building blocks with high and low densities on the phase image. These building blocks corresponds metal and dielectric. It is possible to see the big 3 μm granules with metal phase...
concentration increase. We can see that areas of the topographic elevation correspond to areas with low density if we compare phase images with topographical images.

**Investigation of magnetic domains of the composite films**

For investigation of the magnetic properties of the A series we used cantilever MFM10 (NT-MDT). The thickness of the magnetic covering of the AFM probe was 30–50nm, rounded radius was about 30nm. To exclude the influence of surface relief to magnetic force imaging firstly we define the surface relief in traditional regime of AFM. During the next passage on the scanning place the AFM probe move on the surface on the trajectory duplicate the relief of the specimens for excluding of the Van der Waals forces interactions with surface. In this case the AFM probe is in the field of specimens magnetic fields. We got the magnetic domain structure image as result of these forces registration.

The investigations showed that at metal phase concentration lower than 38% the magnetic structure in the films specimen it is not seen. We can see the sharp stripe or net domain structure at higher concentrations. Fig. 3 shows the topographical images of A series of the films with various metal phase concentration with superimposed images of the magnetic domains contour lines. The areas with uniform magnetization (domains) denoted by translucent filling inside the contour lines.

![Figure 3](image-url)

**Figure 3.** Topographical images of the A series of the films with superimposed images of the magnetic domains for the specimens with metal phase concentration a) 38%; b) 45%; c) 48%; d) 55%
We can see the stripe domain structure with the period about 0.4 μm at 38% metal phase concentrations. The length of the visible on the image domains is 5 μm and more. We observe the areas of uniform magnetization coincide with the areas of the topographic elevation usually but sometimes contrary coincide with throats. This occurs due to loose structure of the films before the percolation threshold. That is why there is no strict correspondence between topographic elements and compound materials.

We can see the sharp stripe domain structure associated with topographic elements reaching the percolation threshold. The magnetic domains have eccentric shape and thickness until 0.5 μm. Besides the stripes of domains can join in the more complicate formations. It seems it’s connected with join of the metal granules in the film volume with increase of the metal concentration.

The domain structure has the net shape with weak connection with topographic nonuniformity at 55% metal phase concentration. This is due to the granular structure weakly appearing when a lot of materials carry the substrate and on the film surface we can observe evenness [4]. The magnetic structure in this case exact appears due to the high concentration of the ferromagnetic. The average thickness of the magnetic net wall is 0.4 μm. The average size of the cell in the net is 0.5×1.0 μm² and the cells have the elongate shape.

**Model of the films for magnetic parameters calculations**

Let the plane of the composite film coincides with the plane XY, magnetic bias field is directed along the axis Z. The particles included in the dielectric matrix is approximated by ellipsoids of rotation with the demagnetization factor

For the theoretical calculations of the microwave characteristics we present film as ensemble of particles with elongated ellipsoidal shape. According to the Dubowik model the dipole-dipole interaction energy we can write following way:

\[ F_D = \frac{1}{2} \sum_{i=1}^{3} \hat{N}_{ii}^{M} (\langle M_i \rangle^2 V_M + \frac{1}{2} \sum_{i=1}^{3} \hat{N}_{ii}^{P} (\langle M_i^2 \rangle - \langle M_i \rangle^2) V_M , \]

where \( V_M \) - ensemble volume, \( \hat{N}_{ii}^{M} \) - the macroscopic demagnetizing ensemble tensor, \( \hat{N}_{ii}^{P} \) - demagnetizing tensor of separated granule. If the ensemble consists of rotation ellipsoids or cylinders then demagnetizing factors tensor we can present in following way:

\[ \hat{N}_{ii}^{P} = 4\pi \cdot diag(\epsilon; \epsilon; 1 - 2\epsilon) , \]

where \( \epsilon = \frac{N_1}{4\pi} \) - elliptic coefficient.

In the case of granules magnetized perpendicularly to ensemble plane the density of the anisotropy energy we can write following way [5]:

\[ F_D = 2\pi M_s^2 f^2 + 2\pi M_s^2 f (f - 1) N_{\perp} , \]

where \( f \) - volume concentration of the particles in ensemble. For investigated in the work composite films the concentration \( f \) we can present in following way:

\[ f = \frac{V_m}{V_m + V_d} , \]

where \( V_m \) - the volume of metal phase, \( V_d \) - the volume of dielectric phase in composite material.

The density of the free energy of the composite we can write:

\[ w = -M \cdot H + \frac{1}{2} f^2 M N_{film} M + \frac{1}{2} f (1 - f) M N_{part} M , \]

where \( N_{film} \) - demagnetizing factors tensor of the film, \( N_{part} \) - demagnetizing tensor of separated particle. The demagnetizing factors of the particle we can find by Osborn’s formula.
The particle sizes were obtained by AFM images.

Let’s involve notation \( Q = \left( f - (1 - f)(N_\parallel - N_\perp) \right) \).

![Figure 4. Geometry of the problem](image)

For obtain the resonance condition at perpendicular relative to film plane dc field (fig. 4) we should select the axes \( x \) as the polar axes. We can write the free energy of the ensemble in the following way:

\[
w = -H_0 M_s f \sin \varphi \sin \vartheta + \frac{1}{2} M_s^2 f^2 \sin^2 \varphi \sin^2 \vartheta + \frac{1}{2} M_s^2 f (1 - f)(N_\parallel (\sin^2 \vartheta \cos^2 \varphi + \cos^2 \vartheta) + N_\perp \sin^2 \varphi \sin^2 \vartheta)\]

The equilibrium conditions for the magnetization vector we can present in following way:

\[
\varphi_0 = \frac{\pi}{2}, \quad \sin \vartheta_0 = \frac{H_0}{4 \pi M_s Q}, \quad \text{если} \quad H_0 < 4 \pi M_s Q \quad \text{и} \\
\varphi_0 = \frac{\pi}{2}, \quad \vartheta_0 = 0 \quad \text{при} \quad H_0 \geq 4 \pi M_s Q.
\]

Using Smith-Beljers procedure we can obtain the resonance equation:

\[
\left( \frac{\omega_{res}}{\gamma} \right)^2 = H_0^2 (2f^2 + 3f + 1) - (4\pi M_s)^2 Q^2 (f^2 + f).
\]

The calculation result shown in fig. 5. We can see that the model have limited range of applicability. This range doesn’t include area of the high metal concentrations. The reason is the join of the metal particles and forming of the complicated volume aggregates at high metal concentrations. The calculation of demagnetizing factors of these aggregates calls for the special methods take into account the exchange interaction among the magnetic particles [6].
Figure 5. The ferromagnetic resonance field for the A series of the films (a) and (b) for the B series of the films. Circles - the experimental values, solid line - the result for the model calculations.

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References

[1] Kotov, L. 2008 Advanced Materials Research, 47, p. 706.
[2] Kotov, L. et al. 2012 Journal of Nanoscience and Nanotechnology, 12, p. 1696.
[3] Turkov, V. et al. 2013 Bulletin of the Russian Academy of Sciences. Physics, Vol. 77, 10, pp. 1223–1227.
[4] Antonets, I. et al. 2016 Technical Physics, Vol. 61, 3, pp. 416–423.
[5] Butera A. et al., 1999 Ferromagnetic resonance in as-deposited and annealed Fe-SiO₂ heterogeneous thin films, Phys. Rev. B.60, pp. 12270–12278.
[6] Golov A. 2015 Solid State Phenomena 233-234 pp. 485-489.