

The particle size distribution function in the composite films and microwave magnetic properties

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Abstract. Nano- and microstructure of $\{(\text{Co}_4\text{-Fe}_4\text{-Zr}_{0.7})_x+(\text{Al}_2\text{O}_3)_{1-x}\}$ (A1 series) and $\{(\text{Co}_{1-\text{Nb}_{0.2}\text{-Ta}_{0.05}})_x+(\text{SiO}_2)_{1-x}\}$ (A2 series) films was investigated by atomic force microscopy. The distributions of the metallic granules effective size by various concentrations of the metal phase were obtained. Microwave magnetic characteristics of composite films in respect of obtained distributions were calculated.

1. Introduction

The microwave magnetic and magneto-optical characteristics of magnetic composite films are currently intensively investigated. Such films have unusual properties, such as giant magneto resistance, the anomalous Hall effect, and others [1, 2]. The films have a highly heterogeneous structure defined by the ratio of the concentration of a metal and a dielectric in a film. This fact complicates the explanation of the dependency of the microwave magnetic characteristics of the films from the external magnetic field and structure. Determination of structural and magnetic properties of the film granules makes it possible to construct theoretical models describing its magnetic properties.

2. Preparing of films and investigation methods

The investigated films were prepared by ion beam sputtering on the polycrystalline glass substrate. For film of A1 series used a composite CoFeZr target with size 280x80x10 mm. On the target surface had fixed 12 batched of Al_2O_3 with size 80x10x1 mm. Films of A2 series were obtained with using a composite CoNbTa target and dielectric Zr_2O_3 . The uneven arrangement of the polycrystalline glass plates of silicon oxide on the target surface allow to obtain the continuous concentration change of the metallic and insulating phases of the composite. Deposition time was 120 minutes. Time of cleaning the surface by ion beam was 30 minutes. The total pressure in the chamber before the deposition and after purification of polycrystalline glass substrate was $P = 2.4 \times 10^{-4}$ Torr. Nitrogen pressure in chamber was $P = 7.8 \times 10^{-5}$ Torr.

The chemical composition and thickness of the films were determined using a scanning electron microscope JSM-6400. Used metal alloys have a eutectic composition, and one could expect that the structure of the metallic phases on the film highly likely to be amorphous.

Films were annealed at a temperature of 823 K in a normal atmosphere. Determination of relief and phase contrast surface films before and after annealing was performed on an atomic force microscope (AFM) ARIS-3500.



3. Size distributions of particles

Consider the features of images obtained using an AFM. Topology and phase contrast surface are shown in Fig. 2a-b for A1-series of films with a metal concentration of $x = 0.54$ (Fig. 1a, b) and A2-series with $x = 0.51$ (Fig. 1c, d). It can be seen that the films have a non-uniform surface.

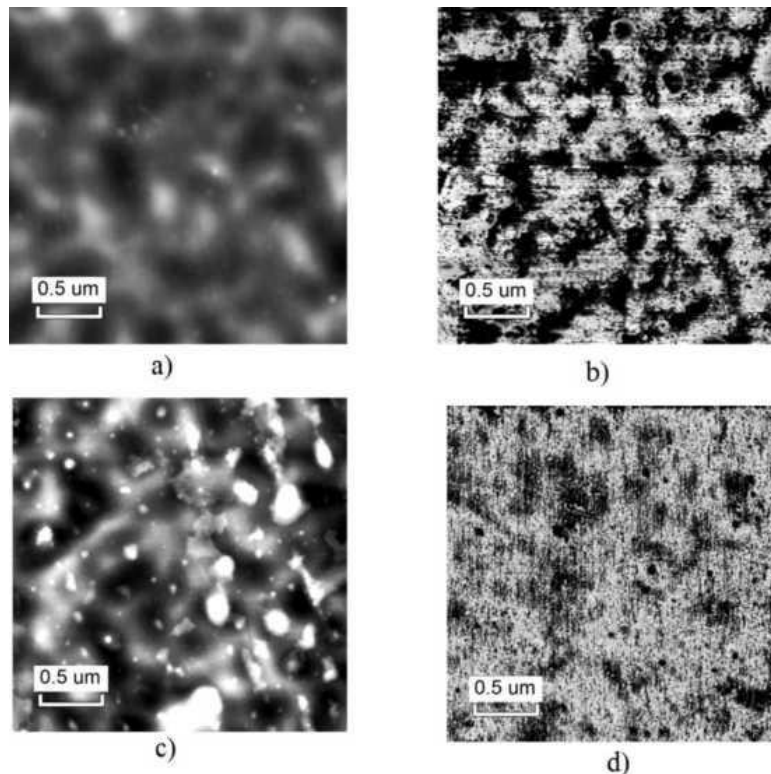


Figure 1. AFM Images of topology and phase contrast of A1 and A2 series composite films

Using the software Gwyddion were built distribution function of metal pellets on the effective size (granules approximated by an ellipsoid of rotation) at different concentrations of the metal phase. Distribution curves of the effective dimensions (radius of ellipsoid on the film plane) of the granules are shown in Fig. 2. On the abscissa in Figure 2 shows effective average size of the granules obtained from the AFM topographic images, and on the ordinate their relative concentrations. It can be seen that increasing of the concentration of the metal alloy the relative amount of small particles size decreases and the number of large particles increases. With increasing concentration of the metal phase x the size and shape of a magnetic granules increases. Changing the shape of the granules leads to the change of the granules demagnetizing fields and respectively, the magnetic characteristics of the films changes.

Annealing of the composite films reduces the heterogeneity of the surface film due to fusion of small areas of metal and dielectric to a larger structures form. This decreases the relative fraction of particles with a small effective size (Figure 3), the particles are formed of a large amount exceeding the maximum particle size observed in the films, not annealed.

The knowledge of the film thickness and the effective radiuses of metal magnetic areas can define their distribution function of field demagnetization value.

The films were cut along the concentration gradient of the metal phase into samples sized $2.5 \times 5 \text{ mm}^2$. Magnetic resonance characteristics of the films at room temperature were determined by EPR-microwave spectrometer at a frequency of 9.36 GHz of rf field.

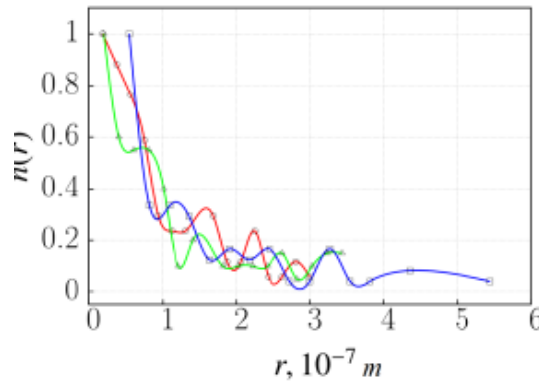


Figure 2. Particle size distribution of Al film for concentrations of magnetic phase $x = 0.45$ (red line), 0.51 (green line), 0.57 (blue line)

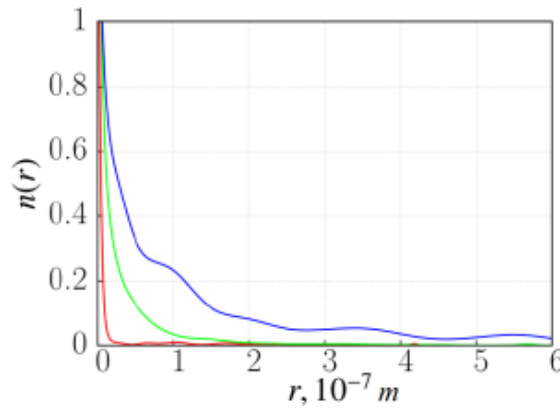


Figure 3. Particle size distribution in Al film without annealing (red line) and after annealing by $T=700$ K (green line), $T=843$ (blue line)

Dependence of the resonance absorption line width on dc magnetic field is shown in Figure 4 (circles). Fig.4 shows that increasing the concentration of the metal magnetic phase $x = 0.3/0.34$ results in first increasing of the resonance line width and then decreasing after reaching the maximum. The increasing of the line width with x increasing is associated with a random distribution of granules in volume of the film and granules size difference, leading to the presence of various small local demagnetizing fields and weak overlap of neighboring granules (the presence of superparamagnetic state). In small x area one of the contributions can be estimated according to the equation:

$$\Delta H_{por} = 1.5(4\pi M_s) \left(\frac{\lambda}{1+\lambda} \right), \quad (1)$$

where M_s - the average magnetization of the film saturation, λ - the material porosity. This contribution is shown in Figure 4 by the curve at low concentrations of magnetic phase. You can see that this dependence is qualitatively consistent with the experimental results.

Reduction in the resonance line width at concentrations of the metal phase exceeding the percolation threshold can be explained by "exchange narrowing". Kurtosis coefficient was chosen as a measure of the resonance line width. For a polycrystalline sample of the spherical shape the degree of resonance peak sharpness is proportional to the square of the magnetization:

$$E_x \propto 0.12 \left(\frac{4\pi M_s}{H_{an}} \right)^2 - 3, \quad (2)$$

where M_s - the magnetization of the sample, H_{an} - induced anisotropy field.

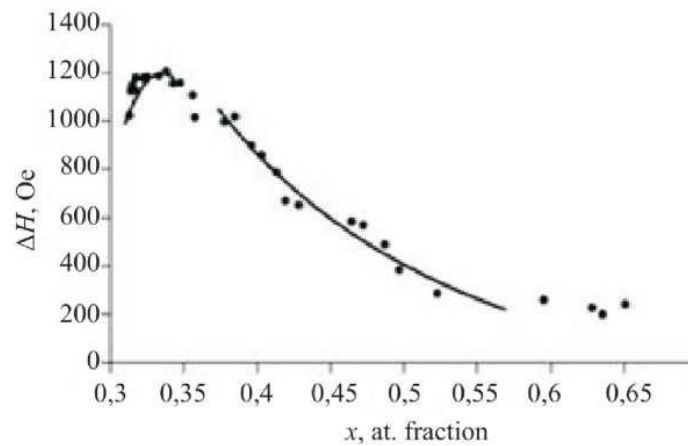


Figure 4. Resonance absorption line width of A1 films

You can see that under the condition of magnetic phase high concentrations the line narrows. To take into account the exchange interaction we need add to the right side of equation (2) the component proportional to

$$\frac{1}{r_0} \left(\frac{\omega_{ex}}{\gamma H_{an}} \right)^2, \quad (3)$$

where r_0 - the average grain size, $\omega_{ex} = 2E_{ex}/\hbar$ - the exchange frequency, E_{ex} - the exchange interaction energy.

To calculate the width of the line, you can use the experimental dependence of the FMR linewidth of the resistivity and magnetization of the film:

$$\Delta H \propto \frac{R_c}{M_s}, \quad (4)$$

where R_c — the resistivity of the film. Concentration dependence of the FMR linewidth calculated by the formula (4) is shown in Figure 4.

4. Theoretical calculations

For the theoretical calculation of the resonance fields we use the Dubowik model. 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 $\frac{N_x}{4\pi} = \frac{N_y}{4\pi} = N_{\parallel}$ and $\frac{N_z}{4\pi} = N_{\perp}$. The values of the demagnetizing factors are determined by the known formulas Osborn based on the data obtained from the AFM images of films.

We write the free energy of an ensemble of particles in the following form:

$$w = -M \cdot H + \frac{1}{2} f^2 M N_{\text{film}} M + \frac{1}{2} f(1-f) M N_{\text{part}} M \quad (5)$$

where f – the volume concentration of the metal phase in the composite, N_{film} – demagnetizing factor of film, N_{part} - demagnetizing factor of one particle. To find the ferromagnetic resonance equation for described geometry of the problem the polar axis OX was chosen. In this case, the free energy of an ensemble of particles can be written as:

$$w = -H_0 M_z f \sin \varphi \cdot \sin \theta + \frac{1}{2} M_s^2 f^2 \sin^2 \varphi \cdot \sin^2 \theta + \frac{1}{2} M_s^2 f(1-f)(N_p(\cos^2 \varphi \cdot \sin^2 \theta + \cos^2 \theta) + N_{\perp} \sin^2 \varphi \cdot \sin^2 \theta)$$

$Q=(f-(1-f)(N_{\parallel} - N_{\perp}))$, then the equilibrium conditions of the magnetization vector equations can be written:

$$\varphi_0 = \frac{\pi}{2}, \quad \sin \theta_0 = \frac{H_0}{4\pi M_s Q}$$

when $H_0 < 4\pi M_s Q$ and

$$\varphi_0 = \frac{\pi}{2}, \quad \theta_0 = 0$$

when $H_0 \geq 4\pi M_s Q$.

Using Smith-Beljers equation we can obtain:

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

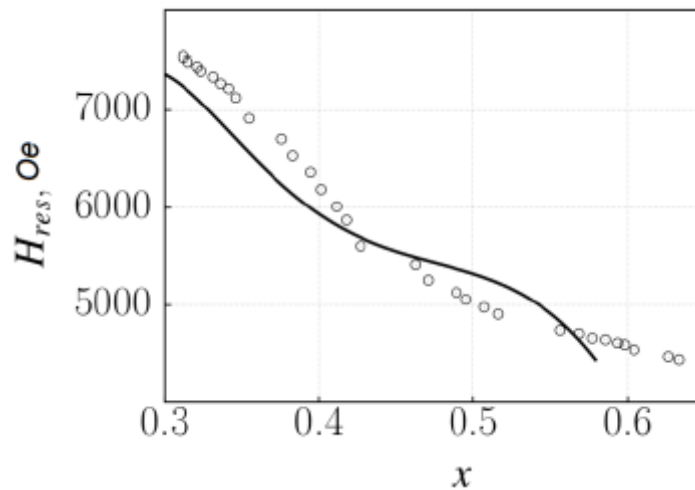


Figure 5. Resonance field of Al films

Let compare values of the resonance fields obtained from the formula (9) with the experimental data. From Fig. 5 can be seen that the chosen model gives a satisfactory result in the concentration range $f = 0.3 \wedge 0.6$. The discrepancy between theory and experiments in the area of low concentrations of magnetic metal due to the limitation of the scope of the selected model, which consists in the fact that the relation (5) adequately describes the ensemble of identical and regularly spaced particles. In real films at low concentrations of metals the particle size and shape widely changes, as well as a random location in the volume of the film. At high concentrations of metal $f > 0.6$, the formation of adhesions complex shape of the particles also remains outside the scope of applicability of the model.

5. Acknowledgements

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6. References

- [1] Kotov, L. 2008 *Advanced Materials Research* **47** 706
- [2] Kotov, L. et al 2012 *Journal of Nanoscience and Nanotechnology* **12** 1696