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MAGNETISM AND FERROELECTRICITY

Ferromagnetic Resonance and Magnetic Microstructure in Nanocomposite Films of $Co_x(SiO_2)_{1-x}$ and $(CoFeB)_x(SiO_2)_{1-x}$

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Abstract—This paper reports on the results of the investigation of the relation between the magnetic microstructure and ferromagnetic resonance (FMR) in ferromagnetic metal–insulator composites by using granular alloys $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ and $Co_x(SiO_2)_{1-x}$ as an example. A comparative analysis of the properties of FMR spectra and parameters of random magnetic anisotropy leads to correlations between these quantities. It has been found that the main mechanism that determines the FMR line width in the films under investigation is the exchange narrowing mechanism.

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1. INTRODUCTION

Granular structures containing ferromagnetic nanoparticles in a nonmagnetic matrix have a number of properties that make them promising materials for novel microelectronic devices. A number of interesting physical phenomena have been discovered in those materials: giant magnetoresistance, giant anomalous Hall effect, enhancement of magneto-optical phenomena, and so on. The effects are mainly due to features of microscopic magnetic structure of the materials.

The microstructure of such nanocomposites is determined by the easiness of creation of nanograins of 3*d*-metals in an insulating matrix of SiO_2. Since the surface energy of 3*d* metals is several times higher than that of SiO_2, the shape of metal grains inside the oxide is close to spherical. In particular, grains are spherical when the content of the metal phase is small. Numerical simulations predict three morphological types of the two-phase mixture depending on volume fraction of metal grains. When the fraction of metal is less than 0.3 metal particles are mainly isolated and randomly distributed over the insulating matrix. For the volume fraction from 0.3 to 0.7, both phases (metal and insulator) form interconnected networks. For the volume fraction of metal above 0.7, the phase microstructure is inverted: metal grains form a conducting matrix and the insulator is represented by isolated particles.

If exchange interaction between nanoparticles of the composite is strong, relations between macroscopic magnetic parameters (such as susceptibility, coercivity, width of the ferromagnetic resonance (FMR) line, and so on) and microscopic parameters of the spin system, such as size of nanoparticles (grains), intergrain (and intragrain) exchange interactions, local anisotropy, are described by the model known as the random anisotropy model [1–4], which has been suggested for amorphous ferromagnets. The random anisotropy destroys long-range ferromagnetic order in the spin system, but it is preserved on the scale of magnetic orientation coherence R_L due to exchange interaction. Studies have shown that microscopic magnetic properties of nanomagnets are governed by the size ($2R_L$) and anisotropy of stochastic domains that are formed spontaneously by large number of particles (of size $2R_c$).

In this paper, we present results of the investigation of the relation between the magnetic microstructure and main parameters of FMR in ferromagnetic metal–insulator nanocomposites by using $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ and $Co_x(SiO_2)_{1-x}$ nanocomposite films as an example.

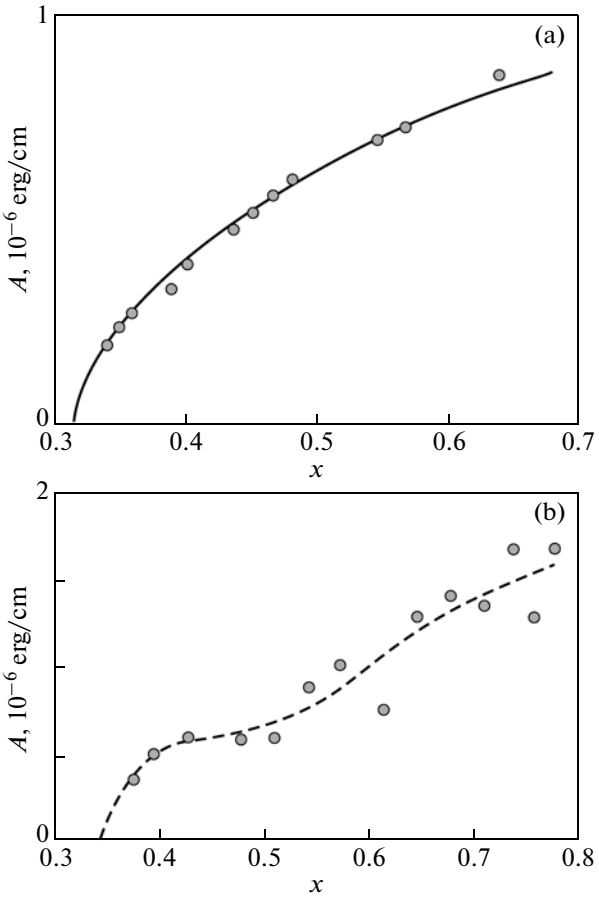

Fig. 1. Exchange interaction constant for (a) $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ and (b) $Co_x(SiO_2)_{1-x}$ composite films.

2. SAMPLES AND METHODS OF INVESTIGATION

The nanocomposites under study are complex systems that consist of nanograins of amorphous $Co_{41}Fe_{39}B_{20}$ alloy and crystalline Co randomly located in the insulating matrix with volume fraction of the metal component 0.3–0.7. Nanocomposites $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ and $Co_x(SiO_2)_{1-x}$ have been produced by ion-beam sputtering of compound targets in argon environment in a setup designed in the Voronezh State Technical University. Film thickness was 3–4 μm. Structure of the nanocomposites was studied by transmission electron microscopy, composition was checked by electron-probe X-ray microanalysis [5]. Low-temperature and field dependences of magnetization $M(T, H)$ were measured using vibration magnetometer in field range up to 14 kOe and temperature range 90–300 K. Resonance parameters were measured using an ÉPA-2M spectrometer (frequency, 9.2 GHz).

We employ technique that allows us to derive size of the element of micro-magnetic structure of the nanomagnet (size of stochastic domain), magnitude of effective anisotropy in the region and size of elements of the nanostructure (size of nanoparticles) and its local anisotropy, and spatial dimensionality of the system of exchange-coupled ferromagnetic nanoparticles from the curves of magnetization approaching saturation [6]. A comparative analysis of the properties of the FMR spectra and parameters of random magnetic anisotropy leads to correlations between those quantities.

3. RESULTS AND DISCUSSION

The investigation of low-temperature behavior of saturation magnetization of the films has shown that $M(T)$ of all nanocomposites in the range of concentrations under consideration ($0.3 < x \le 0.7$) fit well the Bloch law $\Delta M(T)/M_s = BT^{3/2}$ and does not contain features that are specific for superparamagnetic particles. Concentration dependences of the Bloch constant B are qualitatively similar for both series of samples, $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ and $Co_x(SiO_2)_{1-x}$. Value of B decreases from 23×10^{-6} to 6×10^{-6} $K^{-3/2}$ with increase in content of metal phase. Exchange interaction constant A is calculated from B using standard expressions [7]. The obtained values of A for nanocomposites under study are shown in Fig. 1.

Magnetization curves were measured in fields up to 12 kOe and plotted in log–log coordinates $((M_s - M)/M_s, H)$ that reveal power dependences $\Delta M \sim H^{-\alpha}$ and simplify finding their slopes and exponents. Magnetization curves on approach to saturation in both series of nanocomposite films fit the Akulov dependence $M(H) \sim (H)^{-2}$ for all values of x in the fields above 3–6 kOe, which makes it possible to calculate root mean square fluctuation of the local magnetic anisotropy field aH_a ($H_a = 2K/M_s$ is the local magnetic anisotropy field and a is the symmetry factor, which is equal to $a = 1/15^{1/2}$ for uniaxial anisotropy). The quantity H_a decreases with increase in the metal phase content. In the range from 1 to 3–6 kOe magnetization approaches its saturation value as $M \sim H^{-\alpha}$. Exponent α is related to the effective dimensionality of magnetic microstructure in this field range. The magnitude of the field $H_R = 2A/MR_c^2$, where the crossover between power laws takes place, leads to estimate of the correlation radius of random anisotropy R_c. When field decreases in the region of $H < H_R = 2A/MR_c^2$, functional form of $M(H)$ curve changes. The change is due to rise and proliferation of exchange-correlated deviations of magnetization in the spin system (those deviations are called "magnetization ripple" in metal films). It is shown in [8] that in exchange-correlated systems with random anisotropy, length of the ripple $R_H = (2A/MH)^{1/2}$ is limited both from above and from below: $R_c \le R_H \le R_L$. Therefore, functional form of

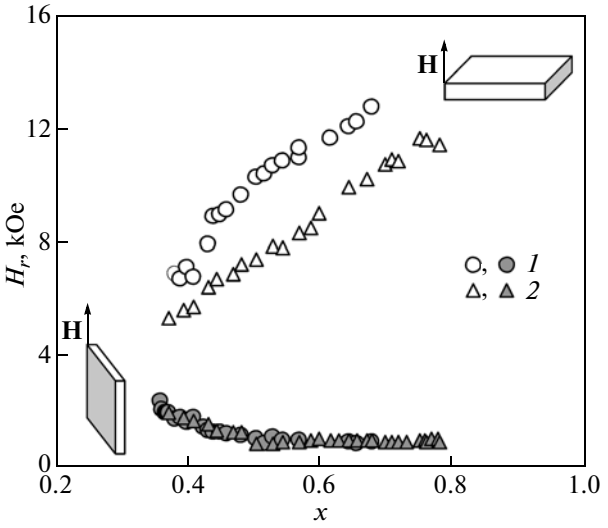

Fig. 2. Ferromagnetic resonance field for (*1*) $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ and (*2*) $Co_x(SiO_2)_{1-x}$ composite films for different field orientations (open and solid symbols).

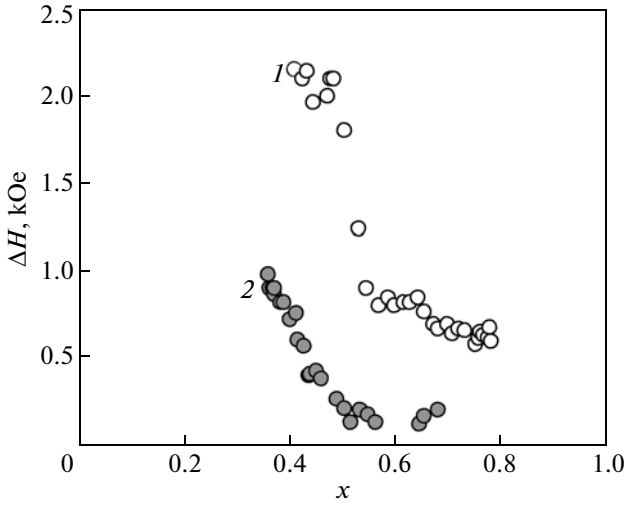

Fig. 3. Dependence of width of the ferromagnetic resonance line on metal phase content in (*1*) $Co_x(SiO_2)_{1-x}$ and (*2*) $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ nanogranular films.

$M(H)$ in the field range $H_L = 2A/MR_c^2 < H < H_R = 2A/MR_c^2$ can be determined from the expression

$$M = M_s\left(1-\left(\frac{aH_a}{H}\right)^2\right), \qquad (1)$$

where local anisotropy of the particle H_a is substituted by the effective anisotropy of the region covered by one ripple length R_H,

$$\frac{M_s - M}{M_s} = \frac{(aH_a)^2}{H_R^{d/2}}\frac{1}{H^{(4-d)/2}} = \frac{(a\langle H_a\rangle_L)^2}{H_L^{d/2}}\frac{1}{H^{(4-d)/2}}, \qquad (2)$$

$$H_L < H < H_R.$$

Here, d is the packing dimensionality of ferromagnetic grains, $\langle H_a\rangle_L = 2\langle K\rangle_L/M_s$ is the macroscopic anisotropy field in stochastic domain. It is established that magnitude of $a\langle H_a\rangle_L$ grows exponentially (along with H_a) with decrease in content of the metal phase.

Value of the parameter α varies in 0.5–1.5 range for both series of samples due to packing configuration of ferromagnetic grains in nanocomposites (this parameter for different contents of magnetic phase in $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ composite was discussed in details in [9]).

FMR spectra of $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$, $Co_x(SiO_2)_{1-x}$ nanocomposite films are studied for various orientations of the film plane in respect to the external field. Concentration dependences of the resonance fields measured in two orthogonal projections are shown in Fig. 2. Those results via the well-known Kittel's FMR equations produce information about the effective magnetic shape anisotropy as function of the magnetic phase content. Behavior of $H_r(x)$ indicates monotonous transformation of effective demagnetization factors from $N_x = N_y = N_z = 4\pi/3$ (isotropic granular medium) to $N_x = N_y = 0$, $N_z = 4\pi$ (solid metallic matrix with insulating inclusions). FMR spectrum in parallel orientation can be expanded into two or three Lorentzian curves with different amplitudes, but with the same H_r. In the orthogonal orientation (the external field is perpendicular to the film's plane) a single absorption line is observed for metal phase concentrations up to $x = 0.55$ in the case of $Co_x(SiO_2)_{1-x}$ and $x = 0.45$ in the case of $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$. When magnetic phase contents rises further, additional absorption lines appear in the microwave spectrum for both series of samples under study. Values of resonance fields of the additional modes indicate that spin-wave resonance spectrum is registered in those films.

The dependence of FMR line width on content of the metal phase $\Delta H(x)$ (Fig. 3) has negative gradient. The largest values of ΔH are typical for isotropic granular medium, the smallest values are observed for metal film with SiO_2 grains. Analysis of calculated exchange correlations in the nanocomposite films under study and FMR line width data revealed that $\Delta H(x)$, $\langle H_a\rangle_L$, and R_L are correlated.

One of the ideas that form foundation of modern attempts to explain FMR line width of nanomagnets is the notion of exchange narrowing of FMR line [10]. In brief it can be expressed by approximate equation $\Delta H(x) \approx k\langle H_a\rangle_L \approx k2A/MR_L^2$, i.e., FMR line width of nanomagnets is related to the mean anisotropy of sto-

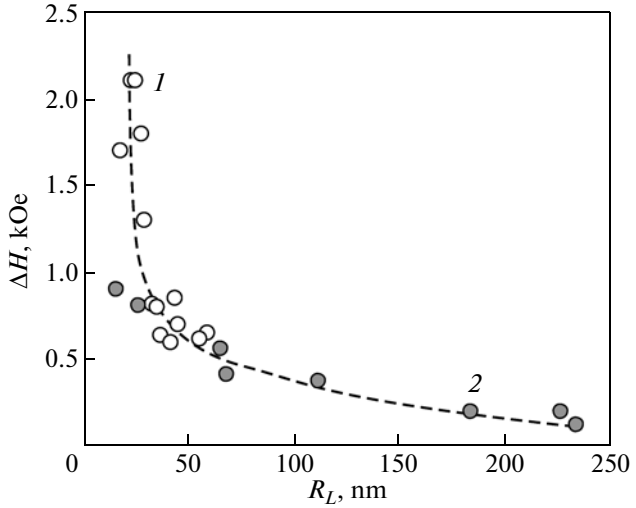

Fig. 4. Dependence of width of the ferromagnetic resonance line in (*1*) $Co_x(SiO_2)_{1-x}$ and (*2*) $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ nanogranular films on the magnetization correlation radius.

chastic domain instead of local anisotropy of grains, as in polycrystals. Using the exchange correlation data for the nanocomposite films under study and FMR line width data we can verify this proposition on a concrete example.

Figure 4 shows experimental dependence of the FMR line width on the magnetization correlation radius (size of stochastic domain) for $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ and $Co_x(SiO_2)_{1-x}$ films. Note that it is in agreement with the calculated dependence $\Delta H(x) \sim A(x)/R_L(x)^2$ (dash line). That indicates that the main mechanism that determines value of ΔH for the films under study is the exchange narrowing.

4. CONCLUSIONS

Investigation of FMR spectra in $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ and $Co_x(SiO_2)_{1-x}$ films measured in various orientations leads to determination of typical content $x \sim 0.36$ at which film can be described as an isotropic granular medium. At larger concentration conglomerates of metal grains are developed right to formation of continuous magnetic film.

Comparative analysis of measured magnetic properties (exchange interaction constant, local anisotropy field) and FMR spectra of $(Co_{41}Fe_{39}B_{20})_x(SiO_2)_{1-x}$ and $Co_x(SiO_2)_{1-x}$ nanocomposite films demonstrates that the main mechanism that determines FMR line width for the films in question is the exchange narrowing.

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