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Effect of Annealing on the Magnetic Properties of $(Co_{40}Fe_{40}B_{20})_x(SiO_2)_{1-x}$ Granular Nanocomposites

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Abstract—The effect of annealing on the magnetic properties of $(Co_{40}Fe_{40}B_{20})_x(SiO_2)_{1-x}$ granular composites fabricated via ion-beam sputtering is investigated. It is established that annealing changes the concentration dependences of the ferromagnetic resonance field and linewidth and shifts the regions of metal phase concentration corresponding to the spin-wave resonance.

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INTRODUCTION

The development of radio electronics, computer engineering, and wireless communication, along with the enhancing of data transfer speed, require new magnetic materials. Materials for high-frequency microelectronic devices must have strong magnetic anisotropy fields, high saturation magnetization, and narrow ferromagnetic resonance (FMR) linewidths [1]. One interesting class of materials that meets these requirements is granular magnetic films [1–4].

Granular magnetic materials are composites consisting of ferromagnetic grains distributed in dia- or paramagnetic matrices. Granular films with nanoparticles of Fe, Co, and alloys based on them are characterized by high saturation magnetization and are therefore of practical interest [2–4]. Improving the hysteresis properties of granular films with magnetically hard Fe and Co grains can allow their application as magnetically hard materials [2–7]. Films with magnetically soft metal grains in nonmagnetic dielectric matrices are of interest as materials for high-frequency electronics, due to their combination of high resistivity and high permeability [8, 9].

The properties of granular media depend on the volume fraction of magnetic grains and are explained in percolation theory. When volume fraction x of the grains lies below so-called percolation threshold x_p, they are isolated from one another. The properties of a granular medium below the percolation threshold depend on the magnetic anisotropy constant of the grains. If the latter is higher than the volume density of the heat fluctuation energy, the medium exhibits high coercivity. In light of this, some authors consider the maximum in the dependence of coercivity on the volume fraction of magnetic grains to be an indicator of the percolation threshold in a material [2–7, 10–12]. If the magnetic anisotropy constant of the grains is in contrast lower than the volume density of the heat fluctuation energy, the granular medium transitions to a superparamagnetic state characterized by the absence of a magnetic hysteresis [13–16].

It has been shown that the $(Co_{40}Fe_{40}B_{20})_x(SiO_2)_{1-x}$ films investigated in this work exhibit a number of intriguing physical effects, including giant magnetoresistance, giant anomalous Hall effect, enhanced magnetooptical effects, and fractal dimensionality of the magnetic structure [17–22]. Heat treatment is a highly efficient tool for controlling the properties of such materials, allowing to alter characteristics by changing the microstructure (size, shape, and number of grains and their distribution over the matrix volume) and grain structure under the action of annealing. To establish the possibility of forming the required magnetic characteristics of composite films via heat treatment, we studied the effect annealing has on some of the magnetic properties of $(Co_{40}Fe_{40}B_{20})_x(SiO_2)_{1-x}$ composite films.

EXPERIMENTAL

$(Co_{40}Fe_{40}B_{20})_x(SiO_2)_{1-x}$ nanocomposites were formed via ion-beam sputtering of composite targets in an argon atmosphere [17]. The material was deposited onto uncooled sitall substrates in modes that ensured the purity of the sputtered films. The substrate temperature during deposition was no higher than 393 K, and the resulting films were 3–4 μm thick. The microstructures of the obtained nanocomposites were studied via transmission electron microscopy (TEM) and their composition was controlled using electron probe X-ray microanalysis [17]. It was established that $Co_{40}Fe_{40}B_{20}$ amorphous nanograins are randomly dis-

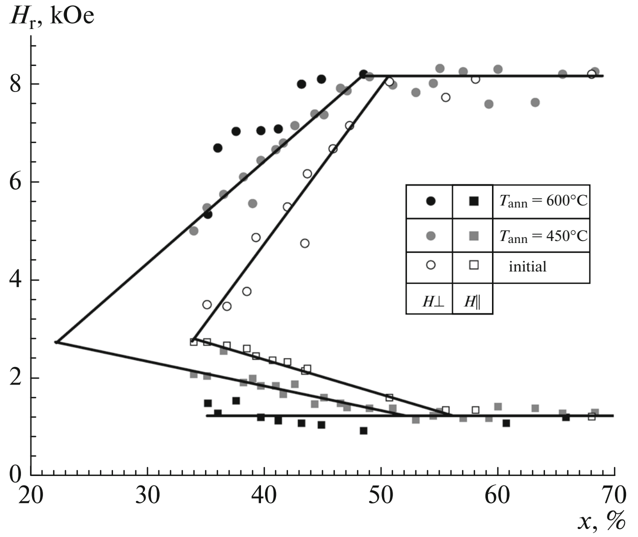

Fig. 1. Ferromagnetic resonance field vs the ferromagnetic component concentration in our $(Co_{40}Fe_{40}B_{20})_x(SiO_2)_{1-x}$ granular films.

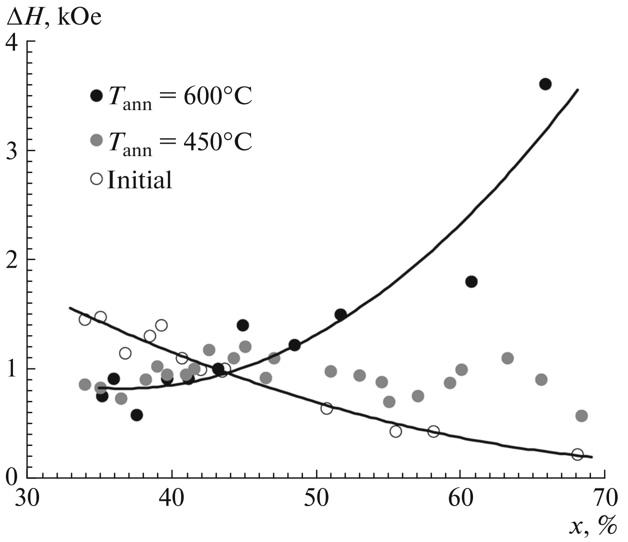

Fig. 2. Ferromagnetic resonance linewidth vs ferromagnetic component concentration in our $(Co_{40}Fe_{40}B_{20})_x(SiO_2)_{1-x}$ granular films.

tributed in a dielectric matrix at a metal component content of 35–70%. The metal grain size in the investigated samples varied between 2–8 nm and grew along with the metal phase content. Annealing at temperatures of 450 and 600°C lasted for 30 min.

Magnetization $M(T, H)$ was measured on a vibrating sample magnetometer in an external field of up to 14 kOe in the temperature range of 90–300 K. The resonance characteristics were measured on a standard EPA-2M spectrometer at a frequency of 9.2 GHz.

RESULTS AND DISCUSSION

Ferromagnetic resonance fields $H_{r\perp}$ at a perpendicular orientation of the external magnetic field were higher than fields $H_{r\|}$ in an external field oriented parallel to the plane of the film (Fig. 1). This follows from the Kittel equation [23] and is related to the dipole-dipole interaction between grains leading to effective magnetic field anisotropy, which repeats the shape of the film's magnetic anisotropy [24]. With a reduction in the volume fraction of magnetic grains, the difference between $H_{r\perp}$ and $H_{r\|}$ remains almost invariable up to $x_p \approx 50\%$ and then starts to fall, indicating a drop in the anisotropy of the effective magnetic field. Grain concentration $x_p \approx 50\%$ is close to the percolation threshold estimated from the measured resistivity of these composites [25], i.e., to the concentration below which the role of a matrix is played by the nonmagnetic dielectric material SiO_2. Linear extrapolation of the $H_{r\perp}x)$ and $H_{r\|}(x)$ dependences when $x < 50\%$ results in the intersection of these lines near a field of 3 kOe that, according to the Kittel equation, is similar to the ferromagnetic resonance field of a homogeneous spherical ferromagnetic sample. This corresponds to the disappearance of the effective magnetic field anisotropy, and thus to negligible dipole–dipole interaction between grains. According to the calculations and empirical estimates made for a granular medium, such a situation should attain when $x \to 0$ [24, 26]. However, it can be seen in Fig. 1 that the concentration near which the sample becomes magnetically isotropic is 35 ± 2% for unannealed films and 22 ± 2% for films annealed at 450°C. Note that the considerable difference from zero for the grain concentration at which a granular film becomes magnetically isotropic has been reported in most experimental works [21, 26, 27]. Such a discrepancy between the theoretical prediction and experiment could originate in particular from unbound nanograins transitioning to the superparamagnetic state. In this case, the average dipole–dipole field on an individual particle could be arbitrary small. This assumption is consistent with previous observations of $(Co_{40}Fe_{40}B_{20})_x(SiO_2)_{1-x}$ films transitioning to the superparamagnetic state as x falls [17]. In addition, the growth of grain size upon annealing should stabilize the ferromagnetic state and thus lead to the increase in difference $H_{r\perp}(x) - H_{r\|}(x)$ seen in Fig. 1.

Heat treatment qualitatively changes the concentration dependence of FMR linewidth ΔH (Fig. 2). In the initial composites, the linewidth increases with a reduction in the metal phase fraction; after annealing, the ΔH value at high x grows sharply and the resonance line narrows with the decreasing fraction of metal grains (Fig. 2). Annealing of the nanocomposites thus reduces the FMR linewidth of the alloys below percolation threshold x_p and to an increase in it above the percolation threshold. Such behavior of ΔH can be understood using the literature data on the crystallization kinetics in amorphous $Co_{40}Fe_{40}B_{20}$ alloys [28, 29], since annealing for 30 min at temperatures of 450 and 600°C leads to crystallization of the $Co_{41}Fe_{39}B_{20}$ alloy. The bcc CoFe(B) crystal phase is nucleated at a temperature of 450°C, and the bcc CoFe(B) crystallites grow as the temperature rises to 600°C [28, 29]. As is well known, the growth of a crystallite upon the annealing of a nanostructured alloy increases the local anisotropy and FMR linewidth [30, 31]. Crystallite growth below the percolation threshold is complicated, since the grains are surrounded by the matrix's material and are almost not in contact with one another. The bcc crystal phase that forms upon annealing thus has a lower local magnetic anisotropy than the amorphous $Co_{40}Fe_{40}B_{20}$ phase. The features of crystallization and grain growth below and above the percolation threshold therefore explain the features observed in the concentration dependences of ΔH (Fig. 2).

In the initial CoFeB–SiO_2 nanocomposites, spin wave spectra were detected when $x > 45\%$. Annealing at 450°C shifted the range of concentrations ($x > 40\%$) of the observed spin wave resonance (SWR) slightly. After annealing at 600°C, the SWR spectrum was observed only when $37 < x < 45\%$. This inversion of the ranges of concentration of the SWR can be explained using the data presented in Fig. 2. The large (over 1 kOe) FMR linewidths lead to spin wave damping and make the excitation of standing waves impossible. The inversion in the ΔH values for the initial films and films annealed at 600°C naturally explains the inversion of the ranges of concentration of SWR observation.

CONCLUSIONS

Annealing changes the shape of the concentration dependences of ferromagnetic resonance fields and linewidths and shifts the ranges of metal phase concentration corresponding to the spin wave resonance observed in $(Co_{40}Fe_{40}B_{20})_x(SiO_2)_{1-x}$ granular composite films prepared by ion-beam sputtering.

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