MAGNETO-OPTICAL AND MAGNETIC RESONANCE PROPERTIES OF NANO-SCALED GRANULAR FILMS (CoFeB)_x(SiO_2)_{100-x} and (CoFeB)_xC_{100-x}

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Abstract. The results of experimental studies of magneto-optical (MO) and magnetic resonance (MR) properties of nanogranular film structures (CoFeB)_x(SiO_2)_{100-x} and (CoFeB)_xC_{100-x} obtained by the ion-beam sputtering method are presented. Magnetic spectra were registered by the method of ferromagnetic resonance (FMR), and magneto-optical spectra were registered by observation of Transversal Kerr Effect (TKE). A comparative analysis of the experimental MO spectra and FMR spectra for systems (CoFeB)_x(SiO_2)_{100-x} and (CoFeB)_xC_{100-x} indicates that a multi-phase structure realizes in the (CoFeB)_xC_{100-x} system. The existence of a multiphase structure is associated with possible plasma-chemical processes that can occur during the synthesis of nanosystems with carbon.

1 Introduction

Nanoheterogeneous structures are promising functional materials for various practical applications (for example, in spintronics, one-electron electronics, etc.). One of the main provisions that technologists usually follow when synthesizing such structures is the impossibility of forming chemical compounds between the phases of a heterogeneous system. The relevance of this restriction can be confirmed by the following example. In the synthesis of film nanostructures of the CoFeB-Si type, silicon enters into chemical reactions with metals to form magnetic and nonmagnetic complexes. As a result, the introduction of even a small amount of silicon led to a significant change in the magnetic and magneto-optical properties up to disappear [1-3]. Unfortunately, the number of combinations of compounds that by means of self-organization processes can form a multiphase nanoscale structure and, at the same time, are mutually inert, is limited. In synthesis of ordered nanostructures the requirement of mutual inertness for the starting materials may in some cases be redundant. It is shown experimentally [4] that the process of self-organization with formation of a heterogeneous structure is possible also when using starting materials which themselves and / or their individual elements can engage each other in a chemical reactions to form a new (previously absent) compounds. For this reason, it seems very promising to study the cases for which the process of self-organization with formation of a heterogeneous structure is possible against the background of plasma-chemical reactions in the synthesis by ion-plasma sputtering. An example of such actually realized systems can be thin nanogranular film structures of the type a ferromagnet- carbon matrix. How will the plasma-chemical synthesis processes affect physical properties of the new heterogeneous materials? The answer to this question is currently lacking.

The aim of the present investigation is to compare the magneto-optical and magnetic resonance characteristics of nanogranular magnetic films synthesized by the ion-plasma sputtering from substances for which plasma-chemical processes are possible, with the similar characteristics of the systems synthesized from mutually chemically neutral materials.

2 Object of research

We have studied nanogranular film structures with magnetic granules of amorphous CoFeB embedded in the non-magnetic SiO_2 matrix (system 1) and in the carbon matrix (system 2). Nano-systems were obtained by ion-beam sputtering. Thin films with thickness h and magnetic phase concentration x both varying along the length of the samples have been prepared using alternate deposition without interlayers. For the system 1 parameter h varied from 210 to 420 nm and x from 10 to 40 at% (thickness of one deposited layer varied from 4.6 to 8 nm). For the system 2 the intervals were 50 nm < h < 250 nm and 17 at% < x < 56 at% (thickness of one layer varied from 1.0 to 4.9 nm). In the direction transverse to the length of the structure, the thickness of...
the sputtered layer and the magnetic phase concentration did not change. The direction in the plane of the sample, coinciding with the longitudinal direction of the original nanostructure, is further denoted by the vector \( \mathbf{i} \), and the direction in the plane of the film transverse to it is vector \( \mathbf{j} \).

It is known that carbon can interact both with transition metals and alloys on their basis, and with boron [4]. In our case, along with nano-sized CoFeB granules, it is possible to form nanoparticles from metastable (at room temperature) compounds of the CoC, Co3C, Fe3C, and B4C type. Since nanoparticles of carbides with FM metals exhibit magnetic properties [5,6], their presence in the studied system (CoFeB)C \(_{100-x} \) should manifest itself both in magnetic resonance and transversal Kerr effect (TKE).

3 Experimental methods

A complex approach to the study of nanocomposites was used based on the study of magnetic and magneto-optical spectra. Magnetic resonance properties were studied by FMR, and magneto-optical properties were investigated by observing TKE spectra. The FMR spectra were measured with highly sensitive EPR spectrometer JEOL FA-300 at a frequency of 9.44 GHz at room temperature. The spectra for each sample were measured for different orientations of the magnetization field \( \mathbf{H} \) with respect to the film plane, from \( \alpha = 0^\circ \) (\( \mathbf{H} \parallel \mathbf{n} \)) to \( \alpha = 90^\circ \) (\( \mathbf{H} \perp \mathbf{n} \)), where \( \mathbf{n} \) is the normal to the film plane. Two series of experiments were performed. In one, the external magnetic field is \( \mathbf{H} \parallel \mathbf{i} \), in the other – \( \mathbf{H} \perp \mathbf{j} \) (Fig.1) The spectra were used to determine the resonance fields \( H_r \), the absorption line width \( H \) and the intensity of the first derivative of the absorption signal \( J \).

Magneto-optical properties were studied in the energy range of 0.5 - 4.2 eV in TKE geometry. An alternating magnetic field up to 3 kOe was aligned parallel to the sample surface and perpendicular to the light incidence plane.

4 Experimental results

4.1 Magneto-optical properties

Fig.2a shows TKE spectra for the system 1 samples with various concentrations of the ferromagnetic phase, as well as spectra of the reference specimen – alloy Co40Fe40B20. The TKE spectra of the amorphous nanocomposite (Co40Fe40B20)\(_x\)(SiO \(_2\)) \(_{100-x} \) essentially differ from the corresponding spectra of the amorphous alloy Co40Fe40B20. Particularly pronounced changes are seen in the energy range \( E < 1.5 \) eV. While in the alloy Co40Fe40B20 the decrease of the photon energy leads to diminishing of the TKE to zero (TKE \( \approx 0 \) at \( E \approx 0.5 \) eV), in the nanocomposites the TKE changes its sign and reaches extremal negative magnitude in the energy range 1–1.4 eV. In this interval the TKE magnitude of the nanocomposites by its absolute value is approximately 5 times as much as the one of the amorphous alloy Co40Fe40B20.

The general form of the TKE curves corresponds to the curves obtained in [7] for bulk composites of the identical composition deposited onto a fixed substrate. Fig.2b demonstrates the TKE dependence on the magnetizing field magnitude. This data was obtained at the fixed energy of the light quanta \( E = 1.97 \) eV. It is clearly seen that the samples with low content of the ferromagnetic phase (15-30\%) exhibit a linear dependence of the TKE on the magnetic field magnitude. Such behavior of the curves indicates a superparamagnetic character of magnetization process, which is specific for the samples in the state below the percolation threshold [7,8]. Then there is the transitional sample with magnetic phase content \( x = 35\% \), whose magnetic field dependence acquires the shape typical for ferromagnetics. With the further increase of concentration to 40\% the contribution from the ferromagnetic component still grows. In the percolation region, the ferromagnetic component of the sample is magnetized as a single big cluster, yet it seems there still remain separate small magnetic clusters, which do not interact with each other and produce a superparamagnetic contribution to the magneto-optic response of the nanostructure. For this system the percolation threshold is in the region 35-40.7\%, what is a little less than obtained in [7].

Fig. 2c shows TKE spectra of the (Co40Fe40B20)\(_x\)(C)\(_{100-x} \) composites. The TKE spectra of the system 2 substantially differ from the spectra of the system 1. However, by their shape they are identical to the corresponding dependence of the spectrum of the Co40Fe40B20 amorphous alloy. For the nanocomposites (Co40Fe40B20)\(_x\)(C)\(_{100-x} \) in the spectral interval with the maximal MO response the TKE absolute magnitude is roughly 2.5-3 times as less than in the alloy Co40Fe40B20. The major difference between these two systems is a virtually complete absence of the negative TKE in the metal-carbon system.

In fig.2d are given the results obtained for the TKE magnetic field dependence in the system with carbon content. Comparing this dependence with the similar one for the metal-dielectric system, we can say that already at low concentrations of the ferromagnetic phase the samples demonstrate non-linear magnetic field dependence, testifying the ferromagnetic behavior of the
Fig. 2. Spectral and field dependences of TKE for system 1 (a, b) and system 2 (c, d).

Fig. 3. The values of the resonance fields and the width (inset) of the FMR lines for system 1 (a) and system 2 (b).
system 2 in all investigated concentration range.

4.2 FMR properties

In the samples of the system 1 for β ∈ [0, 90°] only one mode was observed in FMR spectra. In the spectra of the system 2 samples as a rule, several absorption lines were seen at all orientations of \( H \). With \( x \) decreasing from 56 to 17 at.% the number of modes in the spectra also decreased, namely, from 8 to 3 for \( \alpha = 0 \), and from 3 to 1 for \( \alpha = 90° \). The modes observed at normal magnetization (\( \alpha = 0 \)) in the spectra of all samples of system 2 with concentrations \( 17 \leq x \leq 56 \) at.% will be further designated as A, B and C (in decreasing order by \( H \)). For tangential magnetization (\( \alpha = 90° \)) the mode A was observed in samples with \( x \geq 23 \) at.%, the mode B with \( x \geq 17 \) at.%, and the C mode - with \( x \geq 40 \) at.%. In approximation of weakly coupled oscillation circuits these modes can be interpreted as homogeneous oscillation modes of magnetization vectors of different magnetic sub-systems, each formed by nanoparticles of one particular composition (carbides of Co and Fe and/or CoFeB).

In the film plane an anisotropy of \( H_r \) was observed. At tangential magnetization the difference between \( H_r \) magnitudes for the cases \( H \perp i \) and \( H \perp j \) may reach magnitudes near 30, 100 and 200 Oe for the modes A, B and C correspondingly. The observed variations of \( H_r \) can be explained (in the framework of the theory of FMR in granular nanostructures [9]) taking into account the following assumptions:

- the FMR properties are mainly influenced by shapes of magnetic granules;
- magnetic granules are ellipsoids with one of the main axes oriented normally to the film plane.

With \( \alpha \) varying from 0 to 90° \( H_r \) monotonically decreases. The curves \( H(\alpha) \) are almost symmetrical to the axis \( \alpha = 0 \).

The concentration dependence of resonance fields \( H_r \) for the normal and in-plane magnetization for the systems 1 and 2 are shown in fig.3a and fig.3b, correspondingly (the curves \( H(\alpha) \) are shown only for the case \( H \perp i \)).

In the samples of the system 1 with concentrations less than \( x \approx 33 \) at.% the resonance fields at normal magnetization are smaller, and at the in-plane magnetization, on the contrary, bigger, than the magnitudes \( H_r \) of the resonance modes in the samples of the system 2. For concentrations \( x > 39 \) at.% the situation changes. In the samples of the system 2 the resonance fields at normal magnetization are bigger, and at the in-plane magnetization smaller, than the magnitudes \( H_r \) of the resonance modes in the samples of the system 1 (comparison is made for the samples with equal \( x \)).

Fig.3a,b also demonstrate the concentration dependence of absorption line width \( H \) for the systems 1 and 2, correspondingly (the curves \( H(\alpha) \) are shown only for the case \( H \perp i \)).

Absorption lines of the A, B and C modes in the samples of the system 2, as a rule, have the widths around 50 – 200 Oe both for in-plane and normal magnetizations.

The width of a single resonance mode in the samples of the system 1 with \( c \leq x < 32 \) at.% is close to 1000 Oe. It is substantially larger (up to 10 times), than the widths of A, B and C absorption lines in the samples of the system 2 with the same \( x \).

The observed abrupt decrease of \( H \) in the samples of the system 1 with increase of \( x \) from 32 to 41 at.% is explained by approaching to the percolation threshold. For the composite \((\text{CoFeB})_x(\text{SiO}_2)_{100-x}\), the percolation threshold - i.e. region of the structural transformation from superparamagnetic to ferromagnetic state, was observed at the concentration of magnetic granules \( x \approx 43 \) at.% [7].

5 Conclusion

A comparative analysis of experimental MO and FMR spectra of the systems \((\text{CoFeB})_x(\text{SiO}_2)_{100-x}\) and \((\text{CoFeB})_xC_{100-x}\) testifies that in the synthesis of the system 2 some plasma-chemical processes probably take place – chemical reactions of carbon with metals and boron. As a result there appear new (previously absent) compounds (presumably, \( \text{Co}_2\text{C}, \text{Co}_3\text{C}, \text{Fe}_2\text{C}, \text{B}_4\text{C} \)). Some formed nanoparticles exhibit magnetic properties. The multimode type of the FMR spectra of the system 2 can be explained by different compositions of the particles. The shape of the forming particles depends on their composition. With increase of the metallic phase content \( x \) the shape of the granules (independently of composition) come nearer to the spherical one.

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