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Investigation of the Magnetic Properties of Amorphous Multilayer Nanostructures $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ by the Transversal Kerr Effect

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Abstract

Magnetic properties in amorphous multilayer nanostructures $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with different content of the CoFeB magnetic alloy in metal-composite layers and inverse location of non-metallic phases C and SiO_2 in composite layers or in interlayers, were investigated by magneto-optical methods in the transversal Kerr effect (TKE) geometry.

Using the spectral and field dependences of the transversal Kerr effect TKE, it has been established that in the samples of both magnetic multilayer nanostructures (MLNS) the magneto-optical response and magnetic order are determined by the phase composition of the composite layers.

In samples of MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ with a post-percolation content of metal clusters in metal-composite layers, the maximum of absolute TKE values decrease by about 2.5 times compared with the initial amorphous $Co_{40}Fe_{40}B_{20}$ alloy, while the field dependences of TKE in samples of this MLNS has features that are characteristic of soft ferromagnets.

In samples of MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with a pre-percolation content of metal clusters in the oxide SiO_{2-x} matrix of metal-composite layers, the TKE spectral dependences fundamentally differed from the TKE of the initial amorphous $Co_{40}Fe_{40}B_{20}$ alloy both in shape and sign. The field dependences of the TKE in the samples of this MLN were linear, characteristic of superparamagnets.

Keywords: amorphous magnetic multilayer nanostructures, metal-composite layers, nonmagnetic interlayers, magnetic clusters of CoFeB, transversal Kerr effect TKE, spectral dependences of TKE, field dependences of TKE, soft ferromagnets, superparamagnets.

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1. Introduction

Currently, one of the most dynamically developing areas of modern solid-state physics is the study of fundamental properties and practical application of artificially created nanomedia with a heterogeneity scale of 1–10 nm.

From a fundamental point of view, nanogranular media turned out to be interesting objects with tunnelling electron transport, expressed size effects and complex magnetic properties due to the single-domain nature of ferromagnetic nanogranules isolated from each other by a dielectric [1-5]. It is possible to obtain magnetic structures with excellent prospects for use in the field of high frequency and microwave frequencies of electromagnetic radiation when transition metals (Fe, Co, Ni) or their alloys are used as one of the phases.

The object of intensive experimental and theoretical studies is the question of the effect of the composition and microstructure of nanomaterials formed in the process of self-organization of atoms during their production on the magnetic, magneto-optical, and magnetotransport properties of nanoheterostructures. Despite the large number of studies in this area, there is still no clarity in understanding the processes accompanying the structural rearrangement of matter, since it is difficult to predict the properties of films, in which the interaction of nanoparticles with each other, with the matrix, and with the substrate plays a significant role, with a huge influence of the size and surface effects inherent in nanoparticles/clusters, their boundaries and surfaces.

In our previous studies for the investigation of the atomic structure of amorphous multilayer nanostructures (MLNS) $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with a different number of bilayers, a different content of metal, dielectric SiO_2 and the carbon component and the inverce location of the last two in metal-composite layers or in interlayers, we used non-destructive methods for analysing interfaces, short-range order, and electronic structure [6–9]. Among them are X-ray diffractometry (XRD), X-ray reflectivity (XRR), ultrasoft X-ray emission spectroscopy (USXES), X-ray absorption near edges structure (XANES) and extending X-ray absorption fine structure (EXAFS) [6–9].

According to the results of these studies in the MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ with SiO_2 interlayers a better preservation of the planarity of the interface boundaries between metal-composite layers and SiO_2 interlayers, than in the MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with carbon interlayers, was found. Also, a better preservation of the integrity of the magnetic granules of the original CoFeB alloy with the Me-C and B-C bonds at the interphase boundaries inside the metal-composite layers was revealed. As a result, in these MLNS a noticeable mixing of metal-composite layers and SiO_2 interlayers did not occur, and the interfaces remain planar [6–8].

In the another MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with silicon dioxide in metal-composite layers and carbon interlayers a different situation was observed. The significant smearing of interface boundaries of metal-composite layer/carbon interlayer detected in it was due to the significant interaction of all components of the complex nanostructure with the formation of Me-O (first of all, Fe-O) and B-O bonds with oxygen of SiO_2 matrix at the interphase boundaries inside the metal-composite layers. As a result, the size of the granules of the initial alloy significantly decreased to clusters with lower coordination numbers, and the whole MLNS was more homogeneous [6–9].

It is well known that magneto-optical (MO) methods provide unique information about magnetic and electronic structure of new complex nanomaterials, and that is why they are effective and informative [10]. The MO properties of amorphous metal-dielectric nanocomposites depend on the phase composition and structure of the metal and dielectric phases, on the volumetric content of the magnetic component and on the shape and size of magnetic granules [11–14].

In amorphous metal-dielectric nanocomposites with oxide matrices, an increase in the MO response was observed for concentrations in the percolation region [13, 14].

For pre-percolation systems ($x < x_{per}$) a superparamagnetic state is characteristic, due to the presence of nanoscale magnetic inclusions located in a nonmagnetic medium [15–19]. In this case, a prerequisite for the existence of superparamagnetism is the absence of interaction between ferromagnetic nanogranules. With an increase in the relative volume of the

ferromagnetic phase, the distances between the granules decrease up to their contact, and as a result of the exchange interaction between the magnetic moments of the atoms of different granules a correlation of the magnetic moments of the structure as a whole arises. This determines the ferromagnetic nature of the heterogeneous object.

The form of the spectral TKE dependences significantly changed upon transition to layer-by-layer deposited nanocomposites [20]. The change in the field dependences of the TKE of the samples deposited layer-by-layer had the same tendency as in the bulk granular nanocomposite – the evolution of the magnetization curves from superparamagnetic to ferromagnetic occurred with an increase in the concentration of *x*. The percolation threshold in layer-by-layer deposited composites shifts to the region of lower concentrations of the ferromagnetic phase.

The MO properties of nanostructures were significantly affected not only by the concentration of the metal phase, but also by the thickness of the composite layers. The thickness of the films is set by the rotation rate of the substrate around the sputtered targets [20, 21]. Studies of the effect of nanocomposite manufacturing technology on their MO properties showed that the system with the thinnest layers should have a percolation threshold at the lowest value of *x* of all the systems studied, which was observed experimentally [20].

A comparison of the MO properties of nanocomposites in oxide matrices and a carbon matrix showed that the form of the spectral and field TKE dependences depends on the type of matrix and strongly differs for SiO_2 and C matrices. This was due to the peculiarities of the formation of nanocomposites with a carbon matrix [22].

The aim of this study was to obtain information about the magnetic properties in amorphous MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with a different content of metal, dielectric SiO_2 and/or C components and the inverse location of the last two in metal-composite layers or in interlayers by studying the transversal Kerr effect TKE, as well as the influence of the structural features of the MLNS on these properties.

2. Experimental

MLNS with different composition of metal-composite layers and different interlayers $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ were obtained by the ion-beam sputtering method (with a layer/interlayer thickness gradient) onto a sitall substrate from two different targets, one of which consisted of a plate of an amorphous metal alloy $Co_{40}Fe_{40}B_{20}$ with carbon or quartz inserts, located at different distances from each other. For the production of the thickness gradient of the metal-composite layers and non-metallic interlayers between the target and the substrates, a *V*-shaped screen was placed. The deposition was carried out in an Ar atmosphere at a pressure of $5 \cdot 10^{-4}$ Torr [17–20].

The transversal Kerr effect which consists in changing the intensity of linearly p-polarized light reflected by a sample magnetized perpendicular to the plane of light incidence was used for the investigation of the magneto-optical properties of the MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ [22–24].

The ratio (δ) of the difference in the intensities of the light reflected by the sample in the presence (I) and absence of a magnetic field (I_0), respectively, measured in the experiment, to the intensity of light I_0 determines the value and sign of the TKE:

$$\delta = (I - I_0) / I_0 = \Delta I / I_0. \quad (1)$$

The spectral and field dependences of the transversal Kerr effect were measured using an automated MO spectrometer in the energy range of incident light quanta E from 0.5 to 4 eV. The amplitude of the applied alternating magnetic field reached 3 kOe. For the registration of the signal, a dynamic method was applied, which allows measuring the relative change in light intensity up to 10^{-5}. In this case, the measurement error did not exceed 5%. The measurements were carried out at room temperature.

3. Results and discussion

Table 1 shows the thicknesses (in nanometres) of bilayers equal to the sum of the thicknesses of metal-composite layers and interlayers, nominal (calculated based on the geometry and deposition rate) and experimental (according to reflectometry data [7, 8]) for MLNS samples, obtained by ion-beam sputtering, in which the magneto-optical

Table 1. Nominal and experimental bilayer thicknesses (metal-composite layer + interlayer) for MLNS samples of two types (nm)

	$[(CoFeB)_{60}C_{40}/SiO_2]_{200}$			$[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$		
Sample numbers	1	2	3	23	33	43
Metal-composite layer	3.3	4.0	4.6	5.9	6.4	6.5
Interlayer	1.7	2.0	2.4	1.2	1.4	1.6
Nominal bilayer thickness	5.0	6.0	7.0	7.1	7.8	8.1
Bilayer thickness according to reflectometry data [7, 8]	5.44	6.44	6.57	7.35	8.15	8.43

properties were studied. The studied samples were numbered in accordance with the increase in bilayer thicknesses and overall thicknesses: *1, 2, 3* for MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and *23, 33, 43* for MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$.

Figure 1 shows the obtained dependences of transversal the Kerr effect TKE on the energy of light quanta for samples of two MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with similar average values of bilayer thicknesses of about 6–8 nm, but with different numbers of bilayers 200 and 46 and, therefore, different total MLNS thicknesses.

Spectral dependences of two MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ shown in Fig.1, demonstrated that the TKE in them is fundamentally different by sign. The maximum absolute values of TKE in the samples of both MLNS were approximately 2.5 times lower than in the film of the initial amorphous $Co_{40}Fe_{40}B_{20}$ alloy, to which corresponds the upper curve in Fig. 1. It should be noted that the general form and signs of the TKE spectral dependences in both MLNS corresponded to the TKE spectral curves from film composites of the same compositions $(CoFeB)_x(SiO_2)_{1-x}$ and $(CoFeB)_xC_{1-x}$ [23], obtained using the same unit as the studied MLNS.

As we stated above, using non-destructive X-ray analysis methods such as XRR, USXES, XANES and EXAFS, it was found [6–9] that in MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ with a high content of magnetic alloy (after the percolation threshold) and carbon in the metal-composite layers, but with oxide interlayers, not only the interfaces, but also the clusters of the initial amorphous CoFeB alloy (with the Me-C and B-C boundary bonds) were preserved, which correlates with the data of spectral TKE dependences and explains the similarity of the shapes and positive values of MLNS spectra and the initial alloy. However,

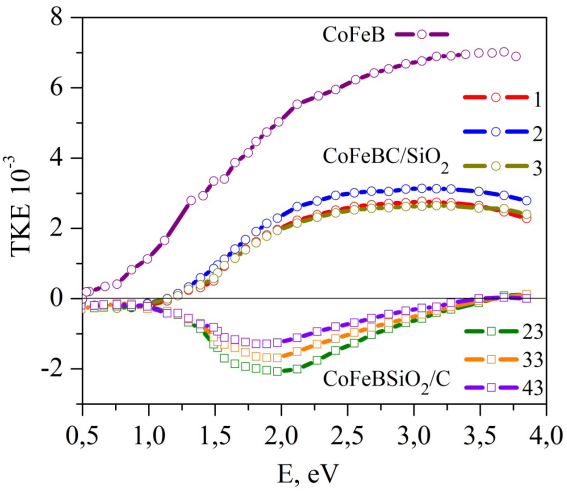


Fig. 1. Spectral dependences of TKE in amorphous samples with different thicknesses for MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ – *1*, *2* and *3*; for MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ – *23*, *33* and *43* and in the film of the amorphous alloy $Co_{40}Fe_{40}B_{20}$ (upper curve)

the influence of dielectric SiO_{2-x} interlayers and the carbon matrix surrounding metal clusters led to a decrease in the relative content of the metal phase in the total volume of the MLNS structure and, accordingly, to a decrease in the TKE modulus by almost 2.5 times (Fig. 1).

Using the same diagnostic methods in the MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with significantly lower magnetic alloy content x = 34 at. % (up to the percolation threshold) and oxide matrix in the composite layers, a significant smearing of the planarity of the interfaces with the formation of oxides of 3*d*-metals, mostly iron oxides was revealed [7–9]. All this leads to a higher mixing of atoms of the metal-composite layers and interlayers, a decrease in the size of metal clusters, and a higher homogeneity of the entire MLNS as compared to the previous one. These circumstances can explain the negative spectral dependences of TKE in MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ in Fig. 1.

Moreover, it should be taken into account, that the same negative spectral TKE dependences were observed in all thin-film samples of composites of variable composition $(CoFeB)_x(SiO_2)_{1-x}$ [23], including the composition of composite layers of studied by us MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$.

The fact that CoFeB composites with different nonmagnetic phases and MLNSs with the corresponding metal-composite layers have spectral dependences similar in general form and TKE of the same sign allows us to conclude that in both MLNSs the MO response and magnetic order are determined by the phase composition and electronic structure of the metal-composite layers.

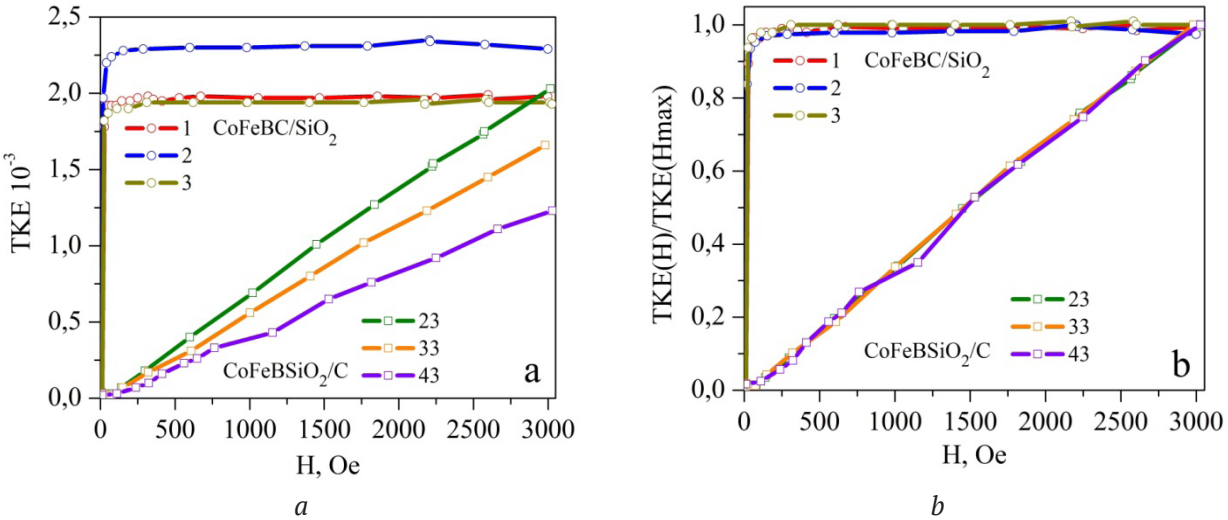
Since the value of the TKE is proportional to the magnetization of the sample, based on the analysis of the TKE dependence on the magnitude of the applied magnetic field, the magnetic order realized in the sample can be determined.

Fig. 2 shows the field dependences of the TKE on the strength of the applied magnetic field for samples of two types MLNS with different thicknesses of bilayers, metal-composite layers and interlayers presented in Table 1.

Comparison of the results for MLNS of two types in Fig. 2 shows that the form of the field dependences of the TKE in MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ has features characteristic of ferromagnets. A sharp increase in the magnetization of all studied samples (*1, 2, 3*) to saturation magnetization indicates that the samples of this MLNS are magnetically soft materials with a coercive force not exceeding several Oe. A slight difference in the magnitude of the effect for samples (*1*, *2*, *3*) was revealed. These differences correlated with the thickness of the samples in a regular way; the effect increased with an increase in the thickness of the composite layers, i.e., with an increase in the volume of the ferromagnetic phase.

As can be seen in Fig. 2, TKE dependence on the magnitude of the field is linear, characteristic for materials with a superparamagnetic magnetization in MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ samples with SiO_2 in metal-composite layers and impaired interfaces [7-9]. Such a magnetic order is characteristic of nanostructures in the pre-percolation state with a low content of the ferromagnetic phase and small sizes of magnetic particles [11–13], which are the samples of this MLNS, containing along with metal clusters of small sizes, metal-oxide and oxy-boride clusters of transition metals. Decrease in the slope angle of the linear TKE(H) dependences (Fig. 2a) with increasing thickness of *23, 33, 43* MNS samples $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ was most probably associated with a change in the morphology (shape and size) and density of magnetic granules in the general structure of the MLNS.

For these MNLS, we observed that with an increase in the thickness of the metal-composite layers, the modulus of the TKE value decreases, and the highest negative effect of TKE was observed



a *b*

Fig. 2. Field dependences of TKE (H) on the magnetic field strength for amorphous MLNS with different sample thicknesses: $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ – *1, 2, 3* and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ – *23, 33, 43:* experimental (a) and normalized by the magnitude of the effect in the maximum field TKE(H)/TKE(H_{max}) (b)

for the MLNS with the smallest thickness of the metal-composite layers (curve *23* in Fig. 1). The thickness of the metal-composite layers affected the size and shape of the ferromagnetic granules, which, in turn, affected the MO properties.

As was shown in the study [20], the size of granules in layer-by-layer sprayed samples is smaller than the characteristic size in a bulk composite, and the probability of contact of granules with each other is higher at smaller sizes of the granules. For a system with thinner layers, the percolation threshold shifts to the region of lower concentrations of *x*, which should lead to an increase in the TKE in the region of a negative maximum, which was observed in our experiment for an MLNS with a SiO_2 matrix in composite layers.

TKE(H)/TKE(H_{max}) dependences normalized by the magnitude of the effect in the maximum field of the in Fig. 2b demonstrate complete coincidence of curves for samples with different thicknesses in each of the two MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$. This fact indicates that a small change in thickness of metal-composite layers, interlayers and general thicknesses of MLNS does not affect the magnetic order realized in structures, ferromagnetic in MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ samples and superparamagnetic in MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ samples.

4. **Conclusion**

Comparative studies of magneto-optical effects in two MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ and $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with different content of metal granules/clusters in metal-composite layers and inverse arrangement of non-metallic C and SiO_2 phases in metal-composite layers or interlayers were performed.

It was found that in both MLNS the magneto-optical response and magnetic order are determined by the phase composition of the metal-composite layers. The general form of the TKE spectral curves obtained from the MLNS corresponds to the dependences obtained for film composites deposited on a similar sitall substrate with a composition identical to the composite metal-containing layers of a multilayer structures.

The difference in the behaviour of the magneto-optical properties of two MLNS with different phase ratios: magnetic metal CoFeB, dielectric SiO_2 and carbon phases and inverse location of non-metallic phases in metal-composite layers or interlayers correlates with the data of studies of short-range order in amorphous MLNS [9].

Spectral dependences of the TKE of three studied MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ samples with different thicknesses of bilayers and post-percolation content of metal clusters x = 60 at. % were similar to the spectral dependence of the TKE of the initial amorphous $Co_{40}Fe_{40}B_{20}$ alloy both in form and in sign, at the energy of light quanta higher than 1.3 eV. This fact correlates with the results of structural studies of this MLNS, in which the preservation of interfaces and magnetic clusters of CoFeB was confirmed [6-9]. The relative fraction of the metallic phase in the total volume of this MLNS is lower than in the initial alloy, and therefore the maximum absolute TKE values decreased by about 2.5 times in comparison with the amorphous CoFeB alloy. In this case, the form of the field dependences of TKE of MLNS $[(CoFeB)_{60}C_{40}/SiO_2]_{200}$ had features characteristic of soft ferromagnets.

In another MLNS $[(CoFeB)_{34}(SiO_2)_{66}/C]_{46}$ with a pre-percolation content of metal clusters at x = 34 at. % in a SiO_{2-x} oxide matrix, the spectral dependences of TKE had a fundamentally different form, differing from the spectral dependence of TKE of the initial amorphous $Co_{40}Fe_{40}B_{20}$ both by form and sign.

This is due to the predominant interactions of metal clusters with the SiO_{2-x} dielectric matrix, which significantly reduced their size and coordination numbers, and, ultimately, determine the nature and sign of the spectral dependences of TKE. The TKE field dependences of this MLNS samples were linear, which characteristic of superparamagnets. This is primarily associated with a significantly lower relative content of CoFeB clusters with metal-boron-oxide shells immersed in a dielectric SiO_{2-x} layer of metal-composite layers. As a result, the exchange interaction between the atoms of metal clusters becomes impossible, and a complex heterophase system turns out to be in a superparamagnetic state.

Conflict of interests

The authors declare that they have no known competing financial interests or personal

relationships that could have influenced the work reported in this paper.

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