

Mössbauer Studies on Tb_xFe_{1-x} Alloy Films with Perpendicular Magnetic Anisotropy

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Abstract—Amorphous ferrimagnetic Tb_xFe_{1-x} films with perpendicular magnetic anisotropy and $Tb_xFe_{1-x}/NiFe$ exchange-coupled structures characterized by unidirectional anisotropy are obtained. The magnetic and chemical inhomogeneity of alloys of Tb_xFe_{1-x} compensation composition is established on the basis of Mössbauer studies of these systems.

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INTRODUCTION

Unidirectional anisotropy has been observed in $Tb_xFe_{1-x}/NiFe$ and $Dy_xCo_{1-x}/NiFe$ film structures [1] in which the layers of Tb_xFe_{1-x} and Dy_xCo_{1-x} amorphous ferrimagnetic alloys are formulated in the area of compensation structures and characterized by integrated perpendicular magnetic anisotropy, while anisotropy of the easy-axis type of the NiFe layer is observed in the film plane. Studies of the magnetic properties of the DyCo monolayer amorphous films and also of the DyCo/NiFe exchange-coupled structures allow us to hypothesize as to the nanoheterogeneity of amorphous ferromagnetic alloys of compensation composition. The concept of two magnetic phases, i.e., P_1 (for which $M_{Co} > M_{Dy}$) and P_2 (for which $M_{Co} < M_{Dy}$) nanophases, was introduced for these systems based on analysis of the results from numerous experiments [2, 3]. Depending on the composition of a Dy_xCo_{1-x} alloy film, one of the nanophases of the alloy spin system serves as the main form (matrix), and the other as an impurity. In the compensation area, the direction of main phase magnetization is oriented orthogonally to the film plane because of perpendicular anisotropy that appears there. Due to the exchange interaction with the magnetization of the 3d metal sublattice of the main phase, the magnetization of the 3d metal sublattice in the impurity phase has a planar component that is involved in exchange interaction with the magnetization of the NiFe alloy layer [4]. Even though the compensation composition (in which the saturation magnetization at room temperature is zero) is a composition with $x \approx 22$ at % for both Tb_xFe_{1-x} and Dy_xCo_{1-x} ferrimagnetic alloy films, amorphous Tb_xFe_{1-x} alloys differ from their amorphous Dy_xCo_{1-x} counterparts by a number of features. The Curie temperature and the average hyperfine field on the Fe nucleus of amor-

phous ferromagnetic Tb_xFe_{1-x} alloys diminish with an increase in the Fe content over a wide range of concentrations, while it is well known that an increase in the 3d metal concentration of amorphous Dy_xCo_{1-x} alloys raises the Curie temperature [5].

This work is devoted to the synthesizing and investigating the magnetic properties of monolayer amorphous Tb_xFe_{1-x} alloys and bilayered exchange-coupled $Tb_xFe_{1-x}/NiFe$ structures by means of Mössbauer spectroscopy. Nuclear gamma resonance is practical for studying amorphous Tb_xFe_{1-x} heterophase films due to the sensitivity of the nuclear gamma resonance absorption spectra to the local orientation of the magnetization vector within the film with respect to the direction of the gamma quanta beam [6].

EXPERIMENTAL

Our investigated Tb_xFe_{1-x} and $Tb_xFe_{1-x}/NiFe$ films (10 < x < 35 at %) were obtained by thermal evaporation [1–3]. The magneto optical Kerr effect in fields of up to 15 kOe and hysteresimetry in fields of up to 250 Oe, applied in the plane of the film at a frequency of 50 Hz, were used as our experimental methods. Thermal treatment at $T = 250^\circ\text{C}$ for 10 minutes was conducted in a vacuum chamber in which the residual gas pressure did not exceed $p = 5 \times 10^{-6}$ mm Hg. Mössbauer spectra of the investigated Tb_xFe_{1-x} films were obtained at room temperature. During deposition, the alloy films were 95% enriched with Fe^{57} isotope.

RESULTS AND DISCUSSION

The form of the magneto optical hysteresis loop measured from the polar Kerr effect allows us to iden-

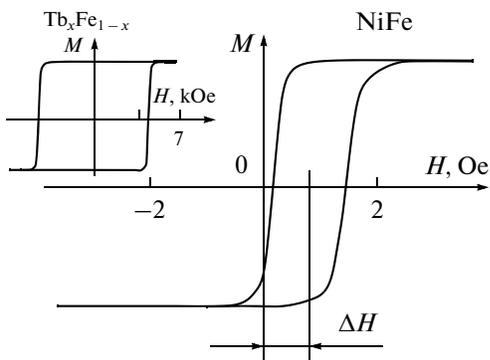


Fig. 1. Hysteresis loop of the NiFe layer in the exchange-coupled $\text{Tb}_x\text{Fe}_{1-x}/\text{NiFe}$ structure; the magneto-optical hysteresis loop of the $\text{Tb}_x\text{Fe}_{1-x}$ layer is shown in the insert ($x = 20$ at %).

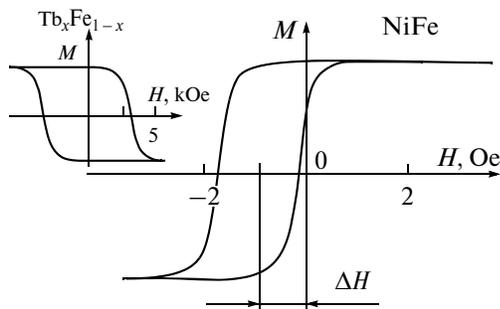


Fig. 2. Hysteresis loop of the NiFe layer in the exchange-coupled $\text{Tb}_x\text{Fe}_{1-x}/\text{NiFe}$ structure; the magneto-optical hysteresis loop of the $\text{Tb}_x\text{Fe}_{1-x}$ layer is shown in the insert ($x = 23$ at %).

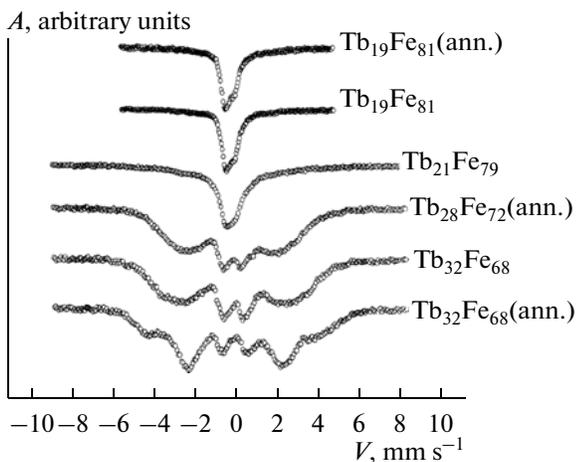


Fig. 3. Mössbauer spectra of our amorphous $\text{Tb}_x\text{Fe}_{1-x}$ alloy films recorded at room temperature.

tify the dominant sublattice in amorphous rare-earth-transition-metal (RE-TM) alloy films with perpendicular magnetic anisotropy. If $M_{\text{Fe}} > M_{\text{Tb}}$, the hysteresis loop is right-handed. The insert in Fig. 1 shows this type of hysteresis loop for an amorphous alloy of precompensation $\text{Tb}_{20}\text{Fe}_{80}$ composition. If $M_{\text{Fe}} < M_{\text{Tb}}$, the magneto optical hysteresis loop is left-handed. This type of loop is characteristic of an amorphous alloy film of post-compensation $\text{Tb}_{23}\text{Fe}_{77}$ composition (insert in Fig. 2). We next synthesized our bilayered exchange-coupled $\text{Tb}_x\text{Fe}_{1-x}/\text{NiFe}$ films. Hysteresis loops of the soft magnetic NiFe layer, measured in the exchange-coupled $\text{Tb}_x\text{Fe}_{1-x}/\text{NiFe}$ structures, are shown in Figs. 1 and 2. The form of these loops indicates unidirectional anisotropy in the planar structures.

Thermal annealing reduces the magnitudes (or leads to the complete disappearance) of both the perpendicular anisotropy of $\text{Tb}_x\text{Fe}_{1-x}$ alloy films (as demonstrated by a drop in the coercive field intensity and rectangularity of the magneto optical hysteresis loops of monolayer $\text{Tb}_x\text{Fe}_{1-x}$ films) and the unidirectional anisotropy of exchange-coupled $\text{Tb}_x\text{Fe}_{1-x}/\text{NiFe}$ films (as demonstrated by a drop in the value or the disappearance of the ΔH bias field).

Mössbauer spectra of monolayer $\text{Tb}_x\text{Fe}_{1-x}$ ferrimagnetic films characterized by perpendicular anisotropy are shown in Fig. 3. The spectral lines appear to be broad, which is typical of amorphous systems.

The spectra of the $\text{Tb}_x\text{Fe}_{1-x}$ films with iron contents of $x > 25$ at % are Zeeman sextets with small amounts of a quadrupole doublet. Raising the Fe content of the samples increases the volume fraction of the paramagnetic doublet. The spectra of the $\text{Tb}_x\text{Fe}_{1-x}$ films ($x < 19$ at % of Tb) are paramagnetic doublets with small admixtures of a sextet, complicating the processing of the spectral data. The observed concentration dependences of the Mössbauer spectra of the investigated films are consistent with earlier studies on amorphous $\text{Tb}_x\text{Fe}_{1-x}$ alloys [6].

We used a two-stage analysis of the Mössbauer spectra. The $P(H)$ function was determined at the first stage. The number and parameters of the hyperfine structure (HFS) of nonequivalent positions and states of iron in the alloy were determined in the second stage from the positions of the maxima. A model spectrum fitted to the experimental spectrum by varying the entire set of hyperfine parameters was produced on the basis of this information. False parts of the spectrum vanish upon this adjustment.

The problem of determining the θ angles (the angles between the direction of the γ quanta beam and the local magnetization vector) for several phases of an inhomogeneous ferrimagnetic alloy upon adjusting all other HFS parameters generally does not have an unambiguous solution. It is necessary to impose limitations on the θ angles or to correct them. Using the magnetic heterophase approach we developed for amorphous ferrimagnetic alloys of the compensation composition, in this work we introduce the hypothesis

that there are two magnetic phases in an iron film; the magnetic moment of one of the phases is parallel ($\theta = 90^\circ$, \leftrightarrow) and the other is perpendicular ($\theta = 0^\circ$, \updownarrow) to the film plane. The sum of the two groups of sextets with $\theta = 0^\circ$ and $\theta = 90^\circ$ is used to determine the $P(H)$ functions. The determined $P(H)$ functions for the two compositions with $x = 28$ and 32 at % of Tb, and for the annealed $Tb_{32}Fe_{68}$ alloy, are shown in Fig. 4. The structure of the $P(H)$ dependences is clearly visible, indicating a finely structure chemical and topological short-range order in the observed magnetic phases. As can be seen from Fig. 4, the alloy regions with $\theta = 0^\circ$ (where the magnetic moment is perpendicular to the film plane) without regard to the composition of the investigated films are characterized by lower magnitudes of the hyperfine fields than the regions with $\theta = 90^\circ$. The alloy compositions that we analyzed were of the post-compensation type, so the regions with $\theta = 0^\circ$ were enriched with a rare earth element relative to the integral composition of the film. Regions of inhomogeneous alloy impurity, in which the magnetization vector is observed in the film plane ($\theta = 90^\circ$), are enriched with Fe, and are hence characterized by higher magnitudes of the hyperfine field.

The nonzero values of the $P(H)$ function for low fields correspond to the paramagnetic component of the spectrum. Thermal treatment leads to a considerable reduction in the volume fraction of the magnetic

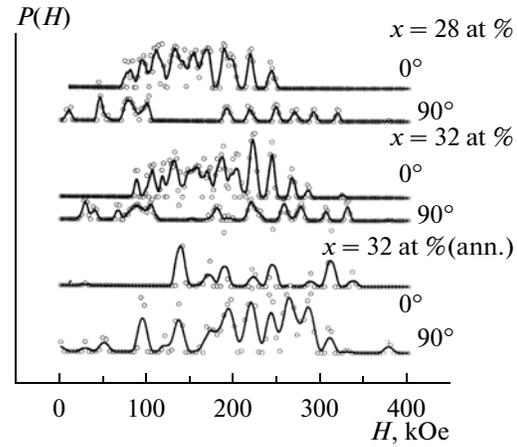


Fig. 4. Distribution of the $P(H)$ hyperfine fields for two orientations of the magnetization vectors: $\theta = 0^\circ$ (the magnetic moment is perpendicular to the film plane) and $\theta = 90^\circ$ (the magnetic moment is in the film plane).

phase with perpendicular orientation of the magnetization vector relative to the film plane, and to the disappearance of perpendicular anisotropy. This is consistent with the magneto-optical measurements that we performed on the annealed Tb_xFe_{1-x} films. The parameters of partial sextets were obtained in the second stage of spectrum analysis, and are given in the table.

Parameters of the positions observed in our Tb_xFe_{1-x} films

	Position	IS	H	QS	$W34$	$W16$	S
$Tb_{28}Fe_{72}$	$\updownarrow P_2$	0.12	84	0	0.58	2.64	0.28
	$\updownarrow P_2$	-0.02	152	-0.03	0.51	1.15	0.14
	\leftrightarrow	0	170	0.04	0.82	0.82	0.20
	$\updownarrow P_2$	0.01	194	0	0.37	0.58	0.06
	$\leftrightarrow P_1$	-0.02	218	0.05	0.76	0.81	0.13
	$\leftrightarrow P_1$	0.06	239	0.21	0.71	0.93	0.05
	$\leftrightarrow P_1$	0.06	257	0.21	0.77	1.01	0.05
			0.21	0	1.39	0.65	—
$Tb_{32}Fe_{68}$	$\updownarrow P_2$	0.15	95	0	0.39	1.8	0.16
	$\updownarrow P_2$	-0.01	174	-0.01	0.50	1.01	0.14
	\leftrightarrow	0	196	0.01	0.91	0.91	0.22
	$\updownarrow P_2$	0.04	221	-0.01	0.46	0.88	0.12
	$\leftrightarrow P_1$	-0.01	241	0.02	0.54	0.55	0.06
	$\leftrightarrow P_1$	0.08	260	0.08	0.81	1.73	0.10
	$\leftrightarrow P_1$	0.09	271	0.07	0.59	0.95	0.08
			0.25	0	1.27	0.86	—
$Tb_{32}Fe_{68}$ Annealing at 250°C for 10 min	\updownarrow	0.39	112	-0.12	0.77	0.88	0.08
	\leftrightarrow	-0.01	197	0.33	0.42	1.42	0.15
	\leftrightarrow	0.18	219	0.10	0.45	1.78	0.21
	\leftrightarrow	0.03	244	0.52	0.27	1.26	0.13
	\leftrightarrow	0.02	256	-0.36	0.61	0.65	0.09
	\leftrightarrow	0.18	267	0	0.79	0.79	0.10
	\leftrightarrow	0.17	301	0.02	0.72	1.05	0.15
			0.37	0	1.38	1.13	—

Note: IS is the isomer shift with reference to α -Fe, ± 0.02 mm/s; H is the hyperfine field on the nucleus, ± 3 kOe; $W34$ and $W16$ are the widths of the inner and outer lines of a sextet, respectively, ± 0.02 mm/s; and S is the fractional population of the position, ± 0.03 .

The data presented in the table show that the magnitudes of the hyperfine fields of the observed local nanoregions of $\text{Tb}_{28}\text{Fe}_{72}$ alloy have lower values relative to the H values of the $\text{Tb}_{32}\text{Fe}_{68}$ alloy nanoregions. These data are consistent with our current knowledge of the concentration dependences of the Curie temperature and the average magnitude of the hyperfine fields of amorphous ferrimagnetic $\text{Tb}_x\text{Fe}_{1-x}$ alloys. It should be noted that nanoregions with $IS = 0$ and $W_{34} = W_{16}$, which comprise $\sim 20\%$ of the material bulk, are registered in films of the investigated compositions that are characterized by perpendicular anisotropy. According to the established magnitudes of the hyperfine fields, $H = 170$ kOe ($\text{Tb}_{28}\text{Fe}_{72}$) and $H = 196$ kOe ($\text{Tb}_{32}\text{Fe}_{68}$). It is therefore logical to attribute them to the P_2 matrix magnetic phase, for which the $M_{\text{Fe}} < M_{\text{Tb}}$ inequality must hold. The magnetization vector of these areas should therefore be oriented perpendicular to the film plane, but the opposite was observed in our experiment. These areas are characterized by $\theta = 90^\circ$. The observed abnormality possibly occurs due to the specific magnetic ordering of these nanoregions and requires further research.

We can determine the volume fractions (the fractional occupancy of the S position) of the P_1 and P_2 magnetic phases from data given in the table. $S(P_1)$ is 23% and $S(P_2)$ is 48% for the films of $\text{Tb}_{28}\text{Fe}_{72}$ composition, while $S(P_1)$ is 24% and $S(P_2)$ is 42% for the films of $\text{Tb}_{32}\text{Fe}_{68}$ composition. The broad distribution of the hyperfine fields in the P_1 and P_2 phases is associated with the local chemical inhomogeneity in both the P_1 and P_2 phases. The population of paramagnetic

regions is $\sim 10\%$. Thermal treatment leads to a change in the phase composition of the alloy. In particular, the hyperfine field grows and perpendicular anisotropy disappears.

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