MOSSBAUER STUDY OF LASER-ANNEALED Tb-Fe FILMS FOR MAGNETO-OPTIC RECORDING

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Abstract-Mossbauer spectroscopy and magneto-optical (MO) studies of thin amorphous Tb_xFe_{100-x} films ($11 < x < 34$ at.%), subjected to the pulsed laser annealing, were carried out. The TbFe films (160 nm thick) were RF-sputtered onto glass substrates; both sides of the films were passivated with SiN layers (85 nm). Both Mossbauer and MO measurements confirmed that in the successively annealed films spontaneous magnetization is well aligned ^{57}Fe along the film surface. The evolutions of hyperfine field distribution at ^{57}Fe and MO data reveal that the laser annealing results in the atomic ordering of TbFe alloys. The crystalline phase of α -Fe appears in the Fe-rich films subjected to laser annealing.

KEYWORDS: MAGNETO-OPTIC RECORDING, MOSSBAUER SPECTROSCOPY, AMORPHOUS FILMS, LASER ANNEALING

INTRODUCTION

Amorphous rare-earth-transition-metal (RE-TM) films are successively used for reversible magneto-optic recording. In these applications the film must present the magnetic anisotropy directed along the film normal.

A significant difficulty, however, lies in the instability of these materials under the influence of pulsed laser irradiation. It is known that magnetic, magneto-optic and structural properties of RE-TM are rather sensitive to the laser annealing [1,2]. The laser annealing to 690°C [1] has introduced a low-coercive magnetic component in addition to the initial ferrimagnetic behavior and doubled the magnetization of the film. Annealing at transient temperatures near 3000°C resulted in loss of the perpendicular anisotropy. The laser annealed passivated TbFeCo films have been probed by use of X-ray and neutron reflectivity measurements [2]. The samples were found to be highly oxidized with a top layer containing terbium carbonate and cobalt oxide.

In this work we studied the relaxation of TbFe films under the pulsed laser annealing and utilized Mossbauer spectroscopy to obtain information about the magnetic microstructure of the film components.

EXPERIMENTAL

Samples of Tb_xFe_{100-x} films were deposited onto glass substrates by RF-sputtering in an argon atmosphere. The nominal thicknesses of TbFe films were 160 nm. Composite target was constituted from pure iron disk and Tb sheets. RE concentration could be varied in the limits of $x = 11-34$ at.%. To increase the efficiency of Mossbauer spectroscopy measurements, the target was doped with ^{57}Fe isotope plates, fixed on the iron disk surface. To prevent oxidation, the TbFe films were sandwiched with RF-sputtered SiN films of 85 nm thickness.

Magnetic (coercive force H_c) and MO parameters (Faraday - θ_F and polar Kerr rotation angles) were measured with MO hysteresis loop tracer ($\lambda = 633$ nm, $H < 10$ kOe).

Mossbauer spectra were measured at room temperature in a transition geometry with the film placed normal to the γ -ray beam. Spectra were recorded with a constant acceleration-type spectrometer in combination with a ^{57}Co -Cr source. Experiments [3] demonstrated that Mossbauer spectra for as-deposited films with $x > 15$ consisted of widened sextuplets with small amplitudes of 2-nd and 5-th lines (Fig. 1a, curve A). Outer lines (1-st and 6-th) were much wider than well resolved inner lines (3-rd and 4-th). All spectra of as-deposited films demonstrated the absence of the peaks corresponding to Fe-containing phases with regular structure (α -Fe, Fe-oxides, Fe-Tb

Fig.4. Mossbauer spectra (a) and hyperfine field distributions (b) for $Tb_{26}Fe_{74}$ film:

A - before annealing,
 D - after the final laser annealing.

compounds). Spectra were characterized by distribution of their parameters and this indicated that all as-deposited samples were in amorphous state. The same conclusion was obtained using X-ray diffractometry.

The computer program DISTRI [4] for Mossbauer spectra recognition was based on the regularization method. Hyperfine field H_n (Fig. 1b), Mossbauer isomer shift δ , quadruple splitting ϵ along with their averaged values and standard deviations could be obtained with the help of DISTRI.

The annealing of the films was carried out with Nd:YAG Q-switched laser pulses ($\lambda = 1,06 \mu\text{m}$, $\tau = 5 \text{ ns}$). The samples of $\sim 1 \text{ cm}^2$ square were successively scanned with laser pulsed beam. Spot size on the sample was from $2,4$ to 8 mm^2 , the energy density of $\sim 0,1 \text{ J/cm}^2$ produced transient temperatures $>1000^\circ\text{C}$.

Mossbauer and MO measurements were carried out after each annealing cycle. Laser treatment cycles were repeated until the MO measurements (Fig. 2) revealed the absence of remnant Faraday rotation (in-plane magnetization of the films).

Fig. 2. Faraday hysteresis loops for $\text{Tb}_{18}\text{Fe}_{82}$ film: A - before, B - after the 1-st, C - after the 2-nd, D - after the final laser annealing.

Fig. 3. Averaged hyperfine field H_n , Faraday rotation angles ϕ_F and Curie temperature as functions of Tb content in TbFe films.

RESULTS AND DISCUSSION

As-deposited films

It is known that the hyperfine field $\vec{H}_n$ of the ^{57}Fe nuclei is tightly connected with the magnetic moment $\vec{\mu}_{\text{Fe}}$ of the iron atom. In this case $\vec{H}_n = -k \cdot \vec{\mu}_{\text{Fe}}$, where for amorphous RE-Fe alloys $k \approx 158 \text{ kOe}/\mu_B$ [5,6]. This means that the H_n value and its vector direction correspond to the value and direction of the iron magnetic moment μ_{Fe} .

Fig. 3 shows data on the averaged hyperfine field H_n and Faraday rotation angles ϕ_F as a function of Tb content in the as-deposited films. For films with $x < 23$ H_n (and μ_{Fe}) values increase sharply with x growth. This may be associated with

Curie temperature (T_C) which increases and still remains close to room temperature in this range of Tb concentration. At the same time, for values of x greater than 23, H_n (and μ_{Fe}) decrease. The steep rise of T_C probably does not play a significant role in $H_n(x)$ (and $\mu_{\text{Fe}}(x)$) dependencies. In this case, decreases of H_n and μ_{Fe} are attributed to additional hybridization of terbium 5d and 6s electrons with iron 3d electrons [7] due to the increased Tb concentration. Experimental results for $x > 23$ give the value of $\partial\mu_{\text{Fe}}/\partial x \approx -19.8 \cdot 10^{-3} \mu_B/\text{at.}\%$ [3], which corresponds well with data of [5] for 500-nm thick films at $T = 4.2 \text{ K}$.

It is known that the MO rotation angles ϕ_F for TbFe films are proportional to the μ_{Fe} components which are perpendicular to film plane [8]. Saturated ϕ_F (normalized to Fe contents) and averaged hyperfine field dependencies (Fig. 3) demonstrate a linear correlation between MO activity and the Mossbauer parameter H_n . From this we conclude that the average angle between μ_{Fe} and the film normal does not vary strongly with Tb concentration for $11 < x < 34$.

Fig. 4. Mossbauer spectra (a) and hyperfine field distributions (b) for $\text{Tb}_{18}\text{Fe}_{82}$ film:

A - before annealing, D - after the final laser annealing.

The ratio of areas of the Zeeman sextuplet components p is another parameter which were obtained by DISTRI. For thin samples the p value is proportional to the ratio of probabilities of corresponding γ -transitions. The dependencies of these transitions $\{p_{ij}\}$ on the angle ϕ between H_n vector and γ -ray wave vector (film normal in our case) are different:

$$p_{1,6} : p_{2,5} : p_{3,4} = 3(1 + \cos^2 \phi) : 4(1 - \cos^2 \phi) : (1 + \cos^2 \phi) = 3:p:1 \quad (1)$$

The experimental results [3] reveal that the $\vec{H}_n$ and $\vec{\mu}_{\text{Fe}}$ vectors do not coincide with the film normal ($\phi=0$, $p=0$) or film plane ($\phi=90^\circ$, $p=4$). Several different models of non-zero mean angle between μ_{Fe} and film normal are discussed in [3].

We assumed that $\vec{\mu}_{Fe}$ vectors are uniformly distributed inside a cone. Calculated values of the mean angle φ between iron magnetic moments and the film normal are plotted in Fig. 6 (lower curve for as-deposited films).

Laser-annealed films

Experiments show that pulsed laser annealing modifies both MO hysteresis loops and Mossbauer spectra. For all the samples studied the annealing results in decrease of coercive force H_c and remnant MO rotation (see Fig. 2). Also, it can be seen clearly that perpendicular anisotropy decreases monotonically with laser treatment and vanishes after several annealing cycles.

Fig.5. Mossbauer spectra (a) and hyperfine field distributions (b) for $Tb_{14}Fe_{86}$ film:

A - before annealing,
 B - after the 1-st laser annealing,
 D - after the final laser annealing.

Fig.6. Mean angle between iron magnetic moments μ_{Fe} and the film normal:
 A - before,
 D - after laser annealing.

Fig.7. Concentration dependencies of the average hyperfine field H_n :
 A - before,
 B - after the 1-st,
 D - after the final laser annealing,
 E - hyperfine field values for crystalline Tb-Fe phases.

All samples show an increase in 2-th and 5-th lines of Zeeman sextuplets and decrease in 1-st, 3-rd, 4-th and 6-th lines (Fig. 1a, 4a and 5a). Modified shapes of the Mossbauer spectra are obviously results of declination of the iron magnetic moments from the film normal. Averaged angles between μ_{Fe} and the film normal (estimated by relationship (1) and plotted in Fig. 6) increase from $\varphi=20-30^\circ$ for as-deposited films to $\varphi \sim 80^\circ$ for annealed films.

Fig. 7 demonstrates the dependence of average hyperfine field H_n for both as-deposited and laser-annealed films with different Tb content. All films annealed exhibit a significant growth of the average hyperfine field H_n and, hence, a growth of the mean value of iron magnetic moment. This indicate that some kind of atomic ordering takes place during laser annealing. The values of H_n for successively annealed films approach the values of corresponding crystalline TbFe phases (upper line in Fig. 7).

Fig. 5 shows the evolutions of Mossbauer spectra and hyperfine field distribution function $p(H_n)$ for the film with low Tb content. It is seen that after several successive annealing cycles, sharp peak at $H_n \sim 330$ kOe appears on the $p(H_n)$ curve. This peak is a consequence of the crystalline α -Fe phase forming under the influence of laser annealing.

CONCLUSION

1. After successive laser annealing of amorphous TbFe films mean angle between iron magnetic moments and the film normal increases. Both Mossbauer spectra and MO hysteresis loop measurements confirm that spontaneous magnetization of the treated films appears to be aligned along the film surface.

2. For all films laser annealing leads to a significant increase of the average hyperfine field and, consequently, to the increase of the average value of Fe atoms magnetic moments. This indicate that some kind of atomic ordering takes place during the annealing.

3. The crystalline phase of α -Fe appears in the Fe-rich films subjected to laser annealing.

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