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Journal of Alloys and Compounds 408–412 (2006) 37–42

Journal of ALLOYS AND COMPOUNDS

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High-field magnetization process in novel TbFeCo/YFeCo magnetostrictive spring magnet type multilayers☆

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Available online 22 June 2005

Abstract

Magnetization process of conventional and discontinuous magnetostrictive spring magnet type multilayers (CMSMM and DMSMM, respectively) is investigated by means of magnetization, magnetostriction and magnetic force microscopy for sputtered $Tb_{0.4}(Fe_{0.55}Co_{0.45})_{0.6}/Y_xFe_{1-x}$ and $Tb_{0.4}(Fe_{0.55}Co_{0.45})_{0.6}/Y_x(Fe_{0.7}Co_{0.3})_{1-x}$ (denoted as Terfecohan/Y_xFe_{1-x} and Terfecohan/Y_x(Fe,Co)$_{1-x}$, respectively) multilayers with a variable Y-content $0 \leq x \leq 0.2$. Various magnetic behaviour such as in-plane magnetic anisotropy, out-of-plane magnetic anisotropy, field-induced transition and exchange bias phenomenon are observed. Optimization of large magnetostriction and large magnetostrictive susceptibility are discussed in terms of the magnetization reversal, exchange coupling between sandwiched amorphous TbFeCo and nanostructured YFeCo-layers.
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Keywords: Magnetic films and multilayers; Magnetization process; Elasticity; Magnetic force microscopy

1. Introduction

The combination of rare earth–transition metal alloys and transition metals in spring magnet type multilayers, e.g. TbFeCo/FeCo, opened a new approach for developing low-field giant magnetostriction [1]. For conventional magnetostrictive spring magnet type multilayers (CMSMM), in which the soft FeCo-layer is structurally homogeneous in either crystalline (c) or amorphous (a) state, the magnetostriction $\lambda^{\gamma,2}$ ($= \lambda_{//} - \lambda_{\perp}$) of the order of 10^{-3} and magnetostrictive susceptibility ($\chi_\lambda = d\lambda^{\gamma,2}/d\mu_O H$) of about 10^{-1} T^{-1} were reported [1–3]. Recently, we have prepared novel magnetostrictive multilayers names as discontinuous magnetostrictive spring magnet type multilayers (DMSMM), in which the soft FeCo-layer is structurally heterogeneous in nanostructure (n) state. Such a novel exchange-spring configuration was realized for sputtered $Tb_{0.4}(Fe_{0.55}Co_{0.45})_{0.6}/Y_xFe_{1-x}$ and $Tb_{0.4}(Fe_{0.55}Co_{0.45})_{0.6}/Y_x(Fe_{0.7}Co_{0.3})_{1-x}$ (denoted as Terfecohan/Y_xFe_{1-x} and Terfecohan/Y_x(Fe,Co)$_{1-x}$, respectively) multilayers with a variable Y-content $0 \leq x \leq 0.2$ and individual layer thickness $t_{TbFeCo} = 12$ nm and $t_{YFeCo} = 10$ nm [4,5]. As regards the R (and Y)-concentration dependence of the microstructure, in these studies, a rather high Tb-content of 40 at% was fixed in order to maintain the amorphous structure in the individual magnetostrictive Terfecohan-layers. The soft magnetic YFeCo-layers, however, can be formed in either homogeneous crystalline, amorphous or heterogeneous nanostructure state depending on the Y-content and/or additional heat treatments. This approach opened an alternative route towards not only high-performance magnetostrictive materials, but also a new generation of excellent soft magnetic nanocomposite materials. The structural, magnetic and magnetostrictive investigations have been performed for various CMSMM and DMSMM configurations. The relevant results are summarized in Table 1. This paper presents magnetization process investigations performed by means of magnetization, magnetostriction as well as magnetic force microscopy. The results are discussed in terms of the TbFeCo–YFeCo exchange coupling, the soft layer microstructure and the interfacial structure.

☆ Invited Paper presented at the RE'04 in Nara.

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doi:10.1016/j.jallcom.2005.04.170

The paper is organized as follows. After the introduction, Section 2 deals with the a-Terfecohan/c-FeCo CMSMM. Section 3 presents magnetic and magnetostrictive properties of a-Terfecohan/n-YFeCo DMSMM at the critical Y-concentration for a naturally formed nanostructure in YFeCo-layers. Conventional approach to nanostructure is presented in Section 4 for DMSMM with high Y-content. Finally, concluding remarks are presented in Section 5.

2. a-Terfecohan/c-FeCo CMSMM

The sputtered Terfecohan/FeCo multilayers are formed with amorphous Terfecohan and crystalline bcc-FeCo-layers. According to the X-ray diffraction results, this microstructure almost remains unchanged with the annealing at $T_A \leq 450\,^\circ$C. The magnetic softness, however, is strongly improved: the magnetic coercivity of about 5.0 mT observed in the as-deposited samples is reduced to 1.9 and 1.7 mT at annealing temperature $T_A = 350\,^\circ$C, then to 1.1 and 0.6 mT at $T_A = 450\,^\circ$C for Terfecohan/Fe and Terfecohan/FeCo multilayers, respectively (see Table 1). The magnetostriction, however, exhibits an optimum at $T_A = 350\,^\circ$C. As illustrated in Fig. 1a and b, the annealing enhances significantly both magnetostriction and magnetostrictive susceptibility. The magnetic softness modification is usually attributed to the stress release. This is consistent with the above-mentioned coercivity decrease. The enhancement of the magnetostriction, however, is mainly governed by the rare earth magnetization. In this case, it is possible that beside the stress release the annealing affects also a fine structure of interfaces, which cannot be detected by X-ray and TEM-electron diffractions. This favours the interfacial exchange coupling and leads the sperimagnetic Tb-cone angle to be closed and the magnetostriction to be enhanced. The larger magnetostriction in the 350 °C-annealed Terfecohan/FeCo multilayer $\lambda^{\gamma,2} = 562 \times 10^{-6}$ in comparison with that of 498×10^{-6} in the corresponding Terfecohan/Fe multilayer (see also in Table 1), on one hand, is attributed to the larger magnetostriction contribution of FeCo-layers than that of Fe-layers. On the other hand, it can be related to the larger TbFeCo/FeCo interfacial exchange energy. The optimal magnetostrictive susceptibility (of $13.4 \times 10^{-2}\,T^{-1}$) of the TbFeCo/Fe multilayer, however, is larger than that (of $11.5 \times 10^{-2}\,T^{-1}$) of the TbFeCo/FeCo one. The larger anisotropy of FeCo with respect to that of Fe must be the origin. As a small remark for this section, one can be concluded that in the CMSMM, the TbFeCo/FeCo is favourable for the large magnetostriction, whereas the TbFeCo/Fe exhibits a tendency to enhance the magnetostrictive susceptibility. It is interesting to verify this point in the other exchange-spring configurations.

In higher fields, however, the magnetostriction exhibits a negative slope. In multilayered systems, properties such as magnetization or anisotropy differ from one layer to the next, so the magnetization reversal occurs at different coercive fields for each layer. When the reversal takes place in a given

Table 1
The values of coercivity $\mu_0 H_C$ (mT), magnetostriction $\lambda^{\gamma,2}$ (10^{-6}) and magnetostrictive susceptibility χ_λ ($10^{-2}\,T^{-1}$) of conventional (C) and discontinuous (D) MSMM

Samples	As-deposited				$T_A = 350\,^\circ$C				$T_A = 450\,^\circ$C			
	Type	$\mu_0 H_C$	$\lambda^{\gamma,2}$	χ_λ	Type	$\mu_0 H_C$	$\lambda^{\gamma,2}$	χ_λ	Type	$\mu_0 H_C$	$\lambda^{\gamma,2}$	χ_λ
Terfecohan/Fe	C	5	313	5.4	C	1.9	498	13.4	C	1.1	142	5.6
Terfecohan/FeCo	C	5	326	2.7	C	1.7	562	11.5	C	1.0	466	12.4
Terfecohan/$Y_{0.1}Fe_{0.9}$	D	3	416	19.4	D	1.0	720	30.7	D	0.4	145	8.0
Terfecohan/$Y_{0.1}(FeCo)_{0.9}$	D	3	538	7.0	D	1.6	658	17.4	D	1.5	311	16.8
Terfecohan/$Y_{0.2}Fe_{0.8}$	C	6.5	130	0.1	C	1.7	167	4.7	D	0.4	170	21
Terfecohan/$Y_{0.2}(FeCo)_{0.8}$	C	5	435	2.4	C	1.6	310	4.1	D	0.6	345	16.1

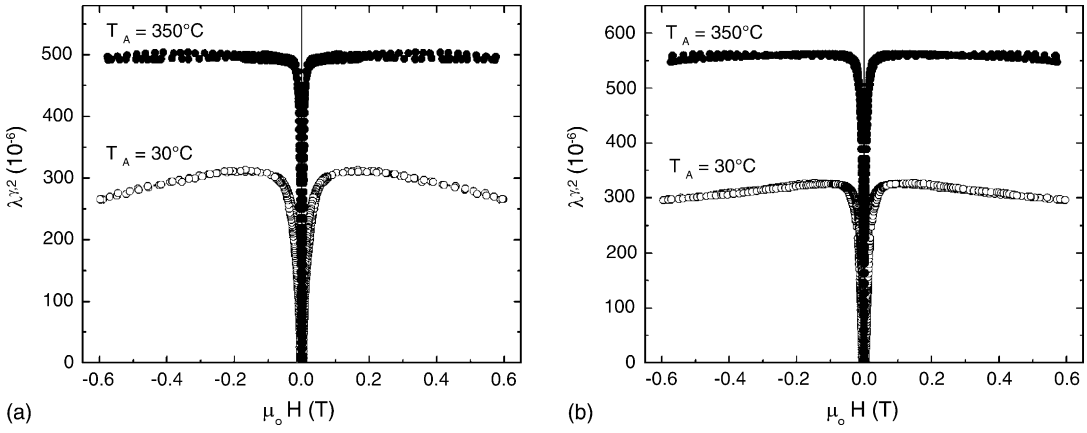

Fig. 1. Magnetostriction data of Terfecohan/Fe (a) and Terfecohan/FeCo (b) CMSMM.

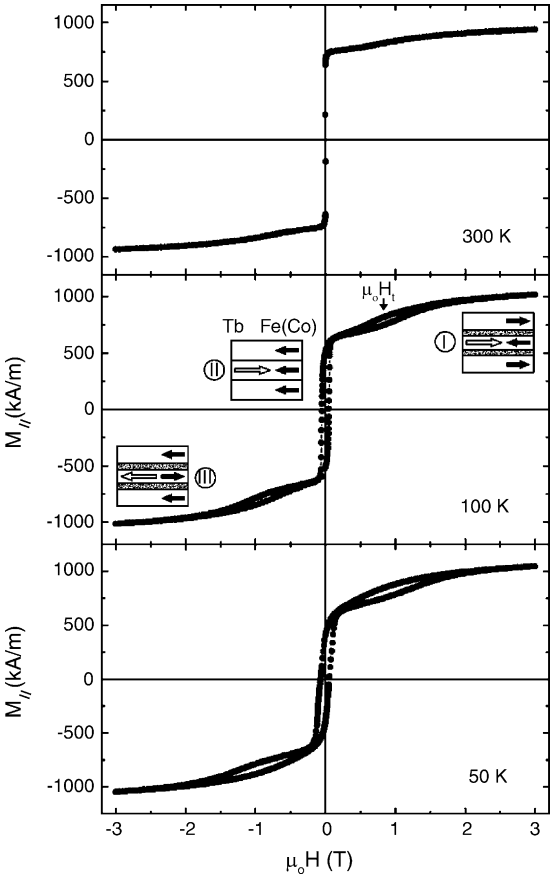

Fig. 2. In-plane magnetic hysteresis loops of as-deposited Terfecohan/Fe CMSMM. The corresponding (low-field) ferrimagnetic (II) and (high-field) ferromagnetic (I and III) configurations are illustrated in the insert, in which dark areas at interfaces indicate EDW.

layer but not in the adjacent one, a so-called extended domain wall (EDW) will be formed at the interfaces and results in a negative contribution to the parallel magnetostriction [6]. As will be indicated below, this is not the case for the films under investigation.

Shown in Fig. 2 is the magnetization data of the as-deposited Terfecohan/Fe CMSMM. A field-induced magnetic transition is observed at $\mu_O H_t$. The phenomenon becomes more pronounced as the temperature decreases. Similar behaviour is obtained for Terfecohan/FeCo. For these multilayers, it is reasonable to assume that the magnetization in the Terfecohan-layers is dominated by Tb [7]. Thus, the corresponding (low-field) ferrimagnetic (II) and (high-field) ferromagnetic (I and III) configurations of the magnetization process are illustrated in the inset of Fig. 2. The EDW formation is well established above $\mu_O H_t$, which is much higher than the fields, where the negative contribution of magnetostriction starts to occur. Moreover, the room temperature field-induced transition is similar for both systems, but their high-field magnetostrictive susceptibility is quite different. For the corresponding annealed samples, both negative magnetostriction slope and field-induced transition almost disappear. In addition, the magnetization is enhanced in the (low-field) ferrimagnetic state. The findings may connect to the atomic configuration in the Terfecohan/Fe interfaces, which is strongly modified by annealing. In this context, the negative magnetostriction component could be attributed to the amorphous phase formed in interfaces. The question, however, still opens for studies in more details.

3. Naturally formed a-Terfecohan/n-YFeCo DMSMM

Naturally formed YFeCo nanograins are observed in multilayers with Y-concentration of $x = 0.1$. This nanostructure formation is associated to the reduction of the thermodynamic driving force for the crystallization caused by the Fe substitution in the Y_xFeCo_{1-x}-layers. In this DMSMM, the observed coercivity value of 3 mT (see Table 1) is still high. However, it is about a half of that obtained in the corresponding Terfecohan/FeCo CMSMM. This may be attributed to the specific nanostructure, in which each FeCo nanocrystal is largely decoupled from the other ones via the non-magnetic matrix. After releasing the stress introduced during the deposition, the coercivity as small as 0.4 mT is reached in the 450 °C-annealed Terfecohan/Fe-film. A higher coercivity is always observed in the corresponding a-Terfecohan/n-YFeCo samples.

Shown in Fig. 3 are magnetostriction data. Clearly, the magnetostriction develops rapidly at the magnetic fields of a few militestla. Optimization of the large magnetostriction ($\lambda^{\gamma,2} = 720 \times 10^{-6}$) as well as large magnetostrictive susceptibility ($\chi_\lambda = 30.7 \times 10^{-2}\,T^{-1}$) was obtained for the 350 °C-annealed film. The obtained χ_λ value is almost 30 times higher than that obtained in the well-known Terfenol-D alloy and comparable with that of the Metglas 2605SC. For Terfecohan/$Y_{0.1}(FeCo)_{0.9}$-films, the largest magnetostriction and parallel magnetostrictive susceptibility are equal to $\lambda^{\gamma,2} = 658 \times 10^{-6}$ and $\chi_\lambda = 17.4 \times 10^{-2}\,T^{-1}$. At present, one may have a remark that in the novel DMSMM, the a-Terfecohan/n-$Y_{0.1}Fe_{0.9}$ DMSMM is the best composition for combining both large magnetostriction and large magnetostrictive susceptibility.

Magnetization data are presented in Fig. 4 for the as-deposited and annealed a-Terfecohan/n-$Y_{0.1}Fe_{0.9}$ DMSMM, respectively. Beside the field-induced magnetic transition at $\mu_O H_t$, one observes also a phenomenon of exchange biasing at low temperatures. The exchange-biasing phenomenon is a property of antiferromagnetic (AF)/ferromagnetic (F) bilayer systems. Similar behaviour is found in exchange-spring magnets, where the hard layer replaces the AF layer as biasing layer [8]. At present, the observed phenomenon may relate to the enhancement of the hysteresis of the field-induced transitions below 100 K. In this case, the magnetization curves under investigation can be considered as minor loops only. The recoil curves show the exchange-spring behaviour, which resembles the exchange-bias loops of other systems (see Fig. 5). At $T = 10$ K, the exchange field $\mu_O H_{ex}$ equals

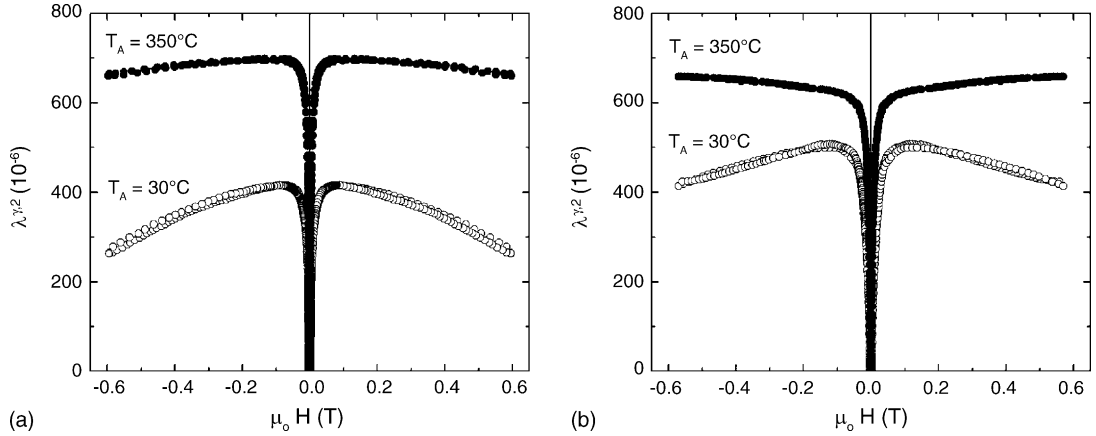

Fig. 3. Magnetostriction data of Terfecohan/$Y_{0.1}Fe_{0.9}$ (a) and Terfecohan/$Y_{0.1}(FeCo)_{0.9}$ (b) DMSMM.

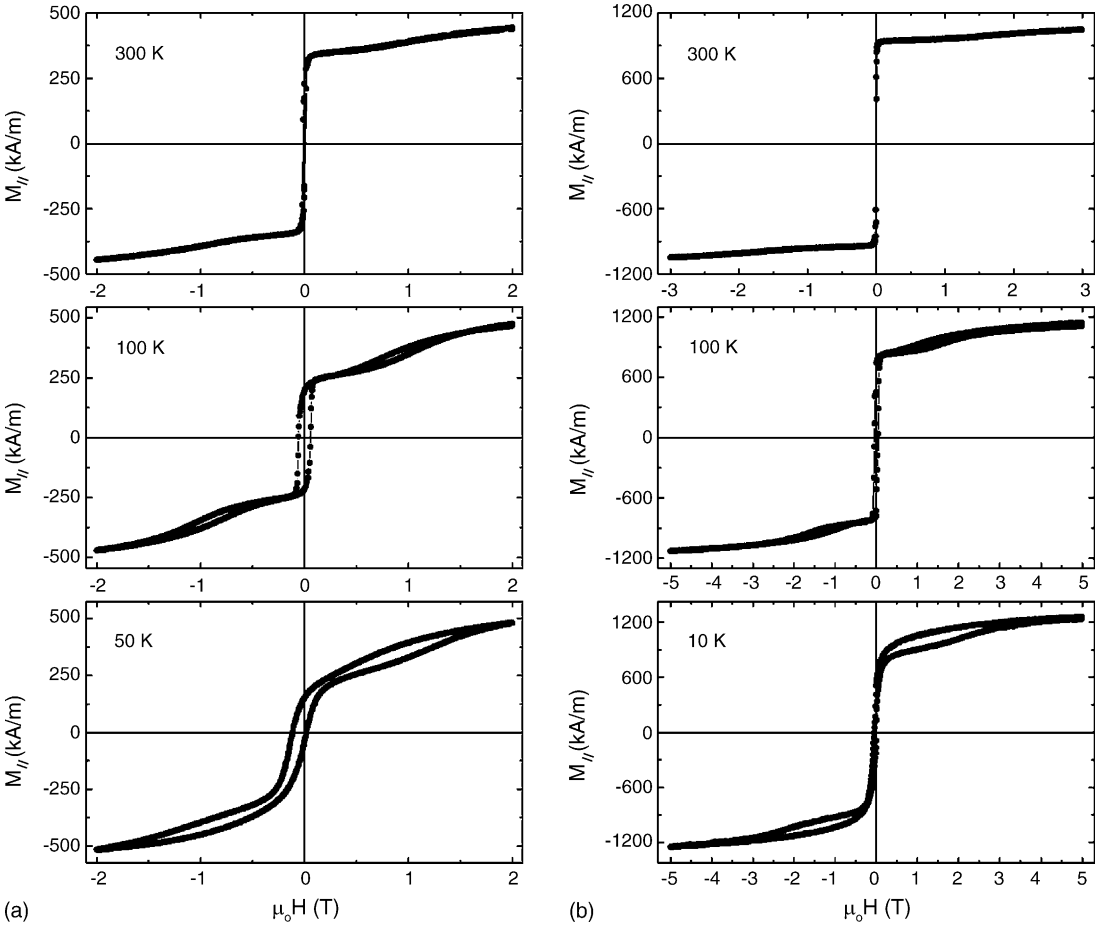

Fig. 4. In-plane magnetic hysteresis loops of as-deposited (a) and 350 °C-annealed (b) Terfecohan/$Y_{0.1}Fe_{0.9}$ DMSMM.

to 0.17 and 0.09 T for the as-deposited and annealed films, respectively. Scaling the magnetization of the soft layer M_{YFe} to the formula of $\mu_O H_{ex} = \gamma/M_{YFe}t_{YFe}$, it turns out that the energy of a domain wall γ is the same order of magnitude in the two samples, i.e. the γ value remains unchanged by annealing.

4. Bottom–up approach to a-Terfecohan/n-YFeCo DMSMM

DMSMM can also be reached from a-Terfecohan/a-YFeCo CMSMM with additional annealing treatments. In our investigations, the amorphous state of the soft magnetic

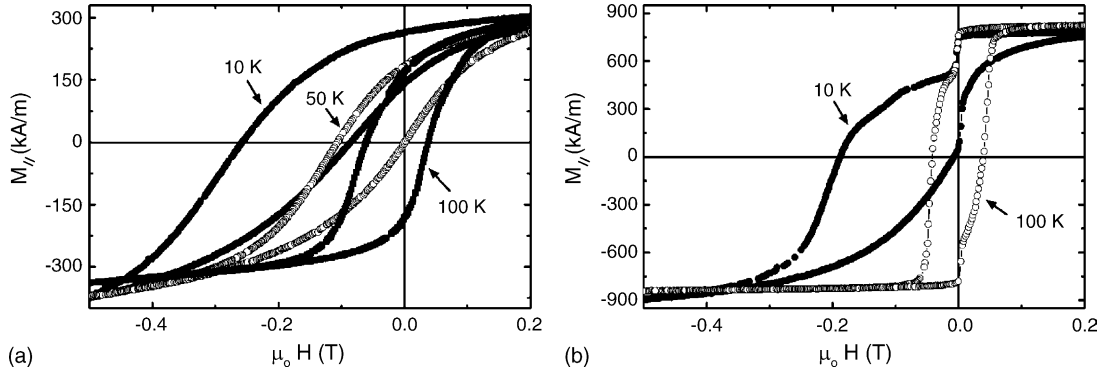

Fig. 5. Exchange-biasing observed in as-deposited (a) and 350 °C-annealed (b) Terfecohan/$Y_{0.1}Fe_{0.9}$ DMSMM.

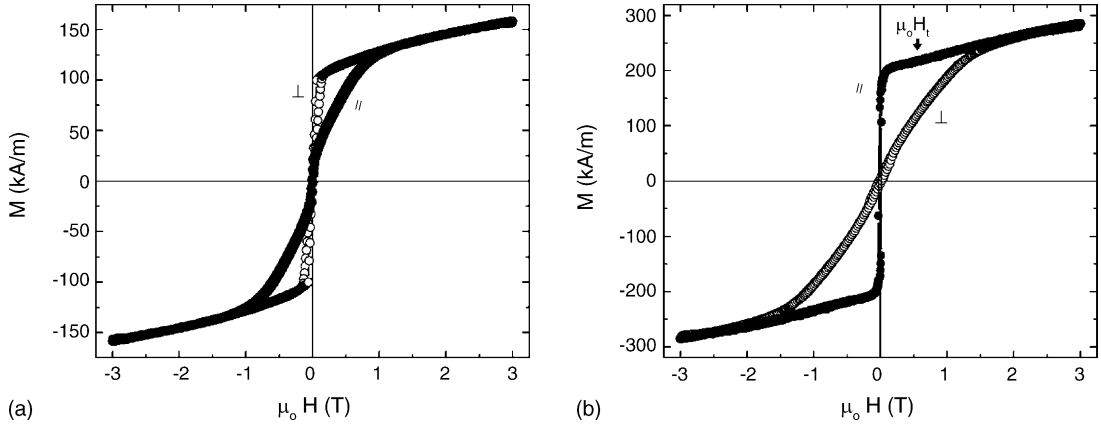

Fig. 6. Room temperature in-plane (//) and out-of-plane (⊥) magnetic hysteresis loops of Terfecohan/$Y_{0.2}Fe_{0.8}$ (a) and Terfecohan/$Y_{0.2}(FeCo)_{0.8}$ (b) CMSMM.

layers is obtained by increasing the Y-concentration. Here, the example is given for the a-Terfecohan/a-$Y_{0.2}Fe(Co)_{0.8}$ multilayers.

Room temperature magnetization data are presented in Fig. 6. The as-deposited multilayer with amorphous YFe-layers reveals an out-of-plane magnetization. In addition, the (low-field) magnetization is rather low (M_s = 100 kA/m in comparison with those of 560 and 500 kA/m of as-deposited Terfecohan/Fe and Terfecohan/$Y_{0.1}Fe_{0.9}$-films, respectively). This could be ascribed to a non-magnetic state of the amorphous $Y_{0.2}Fe_{0.8}$-layers at room temperature [9]. Thus, the coupling between successive TbFeCo-layers is weak and the magnetostrictive layers preserve their intrinsic perpendicular magnetic anisotropy as already found in single layer TbFeCo-films [10]. Indeed, the perpendicular anisotropy is confirmed in the magnetic force microscopy image (Fig. 7): the domains are oriented perpendicular to the film plane and form regularly spaced stripe domain. The Terfecohan/$Y_{0.2}(FeCo)_{0.8}$-film, however, has higher magnetization value and an in-plane anisotropy. This means that the partial substitution of Co for Fe does not modify the amorphous state, but makes the $Y_{0.2}(FeCo)_{0.8}$-layers becoming magnetic at room temperature. In this case, the sample exhibits also the EDW formation at $\mu_O H_t$ (Fig. 6). After annealing, in according to the formation of bcc-Fe nanocrystals, the soft $Y_{0.2}Fe_{0.8}$-layers become ferromagnetic giving a remarkable contribute to the overall magnetization. The TbFeCo/Fe interlayer exchange interactions are now established, and hence the sample exhibits an in-plane magnetic anisotropy as the demagnetising field of the Fe-layers is governed. In addition, the magnetic softness is strongly improved

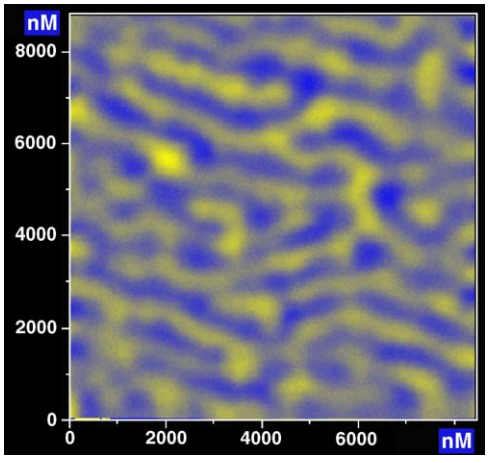

Fig. 7. Magnetic force microscopy image of Terfecohan/$Y_{0.2}Fe_{0.8}$ multilayer measured at $\mu_O H_t$ = 60 mT. The light and the dark areas present domains with magnetization pointing out-of and into the film plane.

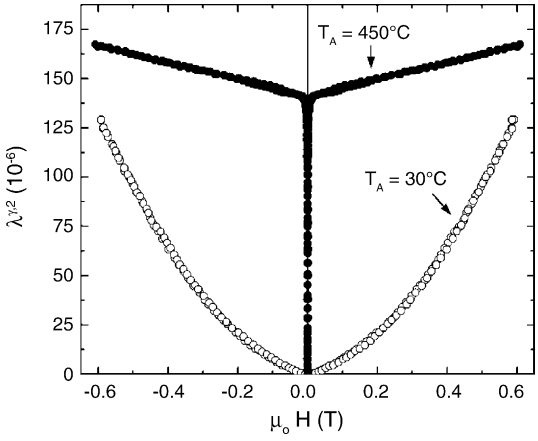

Fig. 8. Magnetostriction data of as-deposited (a) and 450 °C-annealed (b) Terfecohan/$Y_{0.2}Fe_{0.8}$ multilayers.

with the increasing annealing temperature: the coercivity decreases from 6.5 mT in the as-deposited state to 1.7 mT at $T_A = 350\,^{\circ}C$, and finally reaches the minimal value of 0.4 mT at $T_A = 450\,^{\circ}C$. This finding once again supports the above-mentioned tendency that the more the nanoscrystals are formed in the soft layers, the lower coercivity can be achieved in the multilayer. Generally, it is able to note that the large coercivity state is a common character of the CMSMM, in which the soft layers are structurally homogeneous in either the crystalline or amorphous state, whereas the low coercivity is the specific character of the DMSMM, in which the soft layers are formed in nanostructure state. For the 450 °C-annealed a-Terfecohan/n-$Y_{0.2}(FeCo)_{0.8}$, the magnetic coercivity as small as 0.6 mT is also obtained.

Magnetostriction of the as-deposited Terfecohan/$Y_{0.2}Fe_{0.8}$ CMSMM shows parabolic type field dependence, which is a characteristic of films having the perpendicular magnetic anisotropy (Fig. 8) [10]. After annealing, the magnetostriction curve exhibits a rather distinct shape, which is usually found for weak in-plane anisotropic materials. It is worth to note that although the magnetostriction reaches a modest value ($\lambda^{\gamma,2} = 170 \times 10^{-6}$), the magnetostrictive susceptibility as large as $21 \times 10^{-2}\,T^{-1}$ is still obtained in 450 °C-annealed a-Terfecohan/n-$Y_{0.2}Fe_{0.8}$ DMSMM (Table 1). For the a-Terfecohan/n-$Y_{0.2}(Fe,Co)_{0.8}$ DMSMM, however, the larger magnetostriction ($\lambda^{\gamma,2} = 345 \times 10^{-6}$) are achieved. The above remark for the different advantage of FeCo and Fe to λ and χ_λ is also valid here.

5. Concluding remarks

The magnetization process has been investigated for various configurations of the MSMM, in which the soft magnetic YFeCo-layers are formed in either homogeneous crystalline, amorphous or heterogeneous nanostructure state. The relevant results are summarized as follows: (i) the field-induced transition from the (low-field) ferrimagnetic to (high-field) ferromagnetic state combining with the EDW formation at the interfaces is usually observed in CMSMM and DMSMM, in which the magnetism is well established in the individual layers. (ii) The exchange-biasing phenomenon is found at low temperatures in naturally formed a-Terfecohan/n-$Y_{0.1}Fe_{0.9}$ DMSMM. These DMSMM are the best composition for combining both large magnetostriction and large magnetostrictive susceptibility. (iii) When the soft layers become non-magnetic, the coupling between successive Terfecohan-layers is weak and the magnetostrictive layers preserve their intrinsic perpendicular magnetic anisotropy. (iv) It is able to confirm that the magnetization reversal can be controlled by the number of the nucleation sites in the soft-layers. This leads the DMSMM to be the novel configuration for excellent magnetic and magnetostrictive softness.

Acknowledgement

This work was partly supported by the State Program for Nanoscience and Nanotechnology of Vietnam under the Project 811.204.

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