Magnetic properties of nanostructured systems based on $TbFe_2$

R. Ranchal*, V. Gutiérrez-Díez, V. González-Martín

Dpto. Física de Materiales, Fac. CC. Físicas, Universidad Complutense de Madrid. Ciudad Universitaria s/n, Madrid 28040, Spain

A B S T R A C T

The aim of this work is to study the magnetic properties of annealed $[Fe_3Ga/TbFe_2]_n$ heterostructures grown by sputtering at room temperature. The interest of investigating multilayers comprised of $TbFe_2$ and Fe_3Ga is their complementary properties in terms of coercivity and magnetostriction. We have studied the thickness combination which optimizes the magnetic and magnetostrictive properties of the annealed multilayers. The crystallization of the Laves phase upon the thermal treatment in heterostructures with thick $TbFe_2$ layers promotes the increase of the coercivity. This crystallization seems to be prevented by the low mechanical stiffness of the Fe_3Ga. $[Fe_3Ga/TbFe_2]_n$ heterostructures show promising characteristics, λ of 340 ppm and a H_C of 220 Oe, for the development of new magnetostrictive devices.

Keywords:
Sputtering
$TbFe_2$
Galfenol
Thin films
Laves phase
Coercivity
Magnetostriction

1. Introduction

Magnetostrictive materials are widely employed in sensors and actuators devices due to their capability to be used in wireless technology. Materials systems with a high magnetostriction constant (λ) and a low coercive field (H_C) are required for the development of reliable devices. The crystalline $TbFe_2$ Laves phase exhibits the highest magnetostriction constant at room temperature [1]. However, its high coercivity of ~4 kOe, is a drawback for industrial applications [2]. Heterostructures comprised of $TbFe_2$ and soft magnetic layers such as Ni, Fe, or FeB have already been studied [3–6]. In these systems, the combination of $TbFe_2$ and low coercivity materials was expected to provide multilayers with a low H_C and a noticeably magnetostriction constant. Although the coercivity is low, the magnetostriction in some cases is also highly reduced. $TbFe_2$/FeCo multilayers show excellent properties with a saturation magnetoelastic coupling coefficient (λ *Y*, being *Y* the Young's Modulus) of 27.5 MPa and a hysteresis of less than 25 Oe [7].

Galfenol ($Fe_{1-x}Ga_x$) exhibits a pretty high λ, 400 ppm for quenched bulk samples with a 19% and 28% of Ga [8]. Furthermore, its low coercivity and high ductility make of Galfenol a very attractive material for magnetic sensors and actuators. Several studies have been devoted to the study of the structural and magnetostrictive properties of bulk [9–17] and more recently to the investigation of thin Fe–Ga films [18–23]. A review about the magnetostrictive properties of the Fe–Ga alloys has recently appeared [24]. Heterostructures comprised of FeGa and transition magnetic metals alloys such as NiFe and FeCoB have also been reported [25,26]. In this work we present the coercivity and magnetostrictive characteristics of annealed $[Fe_3Ga/TbFe_2]_n$ heterostructures. The interest of these heterostructures is the possibility of achieving new materials with a pretty high magnetostriction but low coercivity due to the complementary properties of $TbFe_2$ and Fe_3Ga in terms of coercivity and magnetostriction. The experimental results presented in this work show the possibility of engineering these two characteristics by means of the layer thickness.

2. Experimental techniques

Samples were grown by the sputtering technique at room temperature on glass substrates. The Ar pressure during growth was 2×10^{-3} mbar and the power was 120 W for the deposition of the $TbFe_2$ and 100 W for the Fe_3Ga layers, respectively. Mo buffer and capping layers (20 nm) were used to protect the heterostructures against oxidation. They were deposited with a dc power of 90 W and at an Ar pressure of 2×10^{-3} mbar. We have grown three different set of samples: (a) a set in which the $TbFe_2$ thickness was kept constant at 12.5 nm whereas the Fe_3Ga layer ranged between 12.5 and 100 nm, (b) multilayers where the Fe_3Ga thickness was fixed at 12.5 nm and the $TbFe_2$ thickness ranged between 12.5 and 100 nm, and (c) heterostructures where the thickness of the $TbFe_2$ and Fe_3Ga is the same and ranged between 12.5 and 100 nm. Hereafter, we will denote these set of samples as (a), (b) and (c). The three series of samples have one in common, the heterostructure with the thinnest studied layers (12.5 nm), i.e. $[Fe_3Ga(12.5\,nm)/TbFe_2(12.5\,nm)]_{16}$. Hysteresis loops at room temperature were carried out in a vibrating sample magnetometer (VSM). The optical cantilever method has been used to infer the magnetostrictive properties of the heterostructures [27]. Thermal treatments were carried out in Ar atmosphere during 1 h at 400 °C. After the annealing, samples were cooled-down at a rate of 20 °C/min because fast cooling-downs enhance the magnetostrictive properties of the Galfenol layers [8].

3. Experimental results and discussion

In a previous work, we reported on the structural properties of $[TbFe_2/Fe_3Ga]_n$ multilayers. We observed signatures of the bcc structure in the Fe_3Ga layers and of the cubic Laves phase in the $TbFe_2$ layers [28]. In this work, we study the thickness combination which optimizes the magnetic and magnetostrictive properties of $[Fe_3Ga/TbFe_2]_n$ multilayers. We have only investigated annealed samples because we have previously observed the low magnetostrictive properties of as-grown $TbFe_2$ thin films grown by sputtering at room temperature [29]. The thermal treatments are necessary to enhance the magnetoelastic properties because the $TbFe_2$ phase with the highest magnetostriction constant is the crystalline Laves phase. Following a previous work of Clark [2], the H_C of $TbFe_2$ is related to its structural crystallinity and hence, in this work we use the coercivity of the heterostructures to track the crystallization of the $TbFe_2$ layers upon the thermal treatment.

We have inferred the coercivity of the annealed $[Fe_3Ga/TbFe_2]_n$ heterostructures from the hysteresis loops recorded at room temperature (Fig. 1(a)). In Fig. 1(b) we show the dependence of the H_C on the layer thickness for the three series of samples. The coercivity of the heterostructure of set (a) with 100 nm Fe_3Ga layers, H_C = 57 Oe, is similar to the value obtained in single Fe_3Ga films, 65 Oe, deposited under the same growth conditions and also post-annealed at 400 °C. Moreover, in this set (a) of samples it is achieved the minimum coercivity among all the studied heterostructures, H_C = 27 Oe, when the Fe_3Ga thickness is 37.5 nm. This H_C is even lower than that of Fe_3Ga single layers. This low H_C value can be due to the antiferromagnetic coupling between the heavy rare earths (RE) and the magnetic transition metals (TM) [30]. In the $TbFe_2$ layers there exists this antiferromagnetic coupling between the Tb and Fe atoms and it is also expected to exist in the $Fe_3Ga/TbFe_2$ interfaces between the Tb and the Fe atoms of adjacent layers. This antiferromagnetic coupling decreases the coercivity as already observed in other systems consisted of RE and TM such as permalloy($Ni_{80}Fe_{20}$)/gadolinium multilayers [31]. The presence of TbFeGa alloys at the interfaces can also account for this low H_C. This possibility will be discussed below.

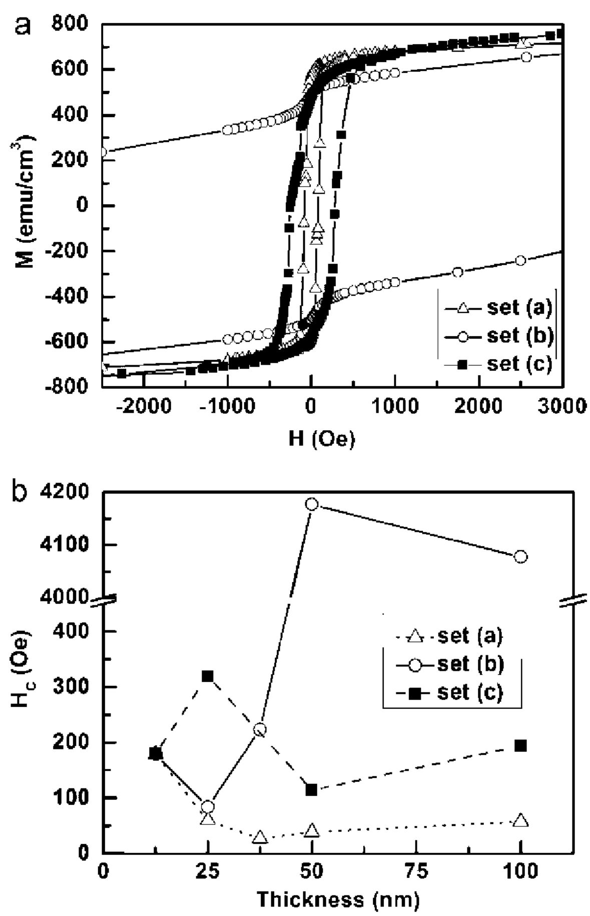

Fig. 1. (a) Hysteresis loops recorded at room temperature of three different heterostructures: set (a) $[Fe_3Ga(100\,nm)/TbFe_2(12.5\,nm)]_3$ (△), set (b) $[Fe_3Ga(12.5\,nm)/TbFe_2(100\,nm)]_3$ (○), and set (c) $[Fe_3Ga(100\,nm)/TbFe_2(100\,nm)]_2$ (■). (b) Coercivity as a function of the layer thickness of the three set of samples: set (a) (△), set (b) (○), and set c (■). All the samples were annealed at 400 °C.

In the set (b), there is a dramatic increase of the H_C up to 4 kOe when the $TbFe_2$ thickness is higher than 37.5 nm (Fig. 1(b)). This strong increase of the H_C indicates a complete crystallization of the $TbFe_2$ Laves phase. Samples of set (c) do not exhibit this strong increase of the coercivity pointing to a partially crystallization of the $TbFe_2$ layers in those samples. H_C is low in these heterostructures of set (c) even when the $TbFe_2$ thickness is 100 nm being remarkable the H_C of just 195 Oe obtained in the $[Fe_3Ga(100\,nm)/TbFe_2(100\,nm)]_2$ multilayer (Fig. 1). Then, the crystallization of the Laves phase takes place in heterostructures with 50 nm $TbFe_2$ layers deposited on 12.5 nm Fe_3Ga layers [set (b)] but it is prevented when the $TbFe_2$ is deposited on top of Fe_3Ga layers with the same thickness [set (c)].

It is also important to remark the shape of the hysteresis loops of the samples of set (b) and (c) presented in Fig. 1(b). This shape indicates the presence of two uncoupled magnetic phases with different coercivity. This behavior is similar to what is observed in exchange-spring magnets comprised of soft and hard magnetic phases in which the layer thickness has not been optimized and each magnetic system rotates at a different magnetic field [32]. Although we do not observe this behavior in all the samples, it is important to notice that the layer thickness should be optimized not only to provide a low H_C and a high λ but also to achieve structures in which the magnetic systems are rigidly coupled.

Previous studies on sputtered Tb–Fe thin films have reported about the influence of the mechanical stiffness of the substrate and/or the buffer layer on the crystallization process of this material system [29,33]. Materials with a high Y/α factor, being α the thermal expansion coefficient, promote the crystallization of the Laves phase. It is important to remark that the best results are related to a high Y/α factor in the substrate or buffer and not to a Y/α close to that of $TbFe_2$. Thus, although $TbFe_2$ and Fe_3Ga have similar Y/α values of around $Y/\alpha = 6 \times 10^6$ GPa K, this value is low in comparison to Mo (71×10^6 GPa K) or Si (43×10^6 GPa K) that are known to promote an optimum crystallization of the Laves phase [29]. Therefore, in set (c) the coercivity can be low because the $TbFe_2$ are deposited on top of Fe_3Ga layers that have a low Y/α factor.

Another possibility for the low H_C achieved in some heterostructures is the presence of TbFeGa alloys at the interfaces. We observed these ternary alloys in the interfaces of as-grown $[TbFe_2/Fe_3Ga]_n$ multilayers [28]. The thermal treatments promote the diffusion of the Tb atoms from the interfaces to the Tb–Fe layers although this diffusion process seemed to be prevented in samples comprised of layers thinner than 50 nm. In those $[TbFe_2/Fe_3Ga]_n$ multilayers with thin layers, the Tb–Fe layers have a lower Tb content than the 33% related to the $TbFe_2$ composition. Although TbFeGa alloys can be present in heterostructures with thin layers studied in this work, the low H_C achieved in the set (c) cannot be explained considering the presence of these alloys. We have obtained low H_C values in the samples of set (c) even when the $TbFe_2$ and Fe_3Ga

layers have a thickness of 100 nm (Fig. 1). The presence of TbFeGa alloys cannot fully explain the experimental results presented in this work. Therefore, the crystallization process promoted by the mechanical stiffness of the buffer layer on top the $TbFe_2$ is deposited seems to play an important role on the magnetic properties of $[Fe_3Ga/TbFe_2]_n$ multilayers.

A representative magnetostriction measurement is shown in Fig. 2(a) being summarized the dependence of the magnetostriction constant on the thickness layer for the three set of samples in Fig. 2(b). Large λ values, $>1 \times 10^3$ ppm, are achieved in samples of set (b) with $TbFe_2$ thicknesses above 37.5 nm. In these samples the Laves phase is crystallized being possible to obtain large magnetostriction constants. Nevertheless, the high coercivity of these heterostructures (Fig. 1(b)) prevents their use in reliable applications.

In this work, we are investigating not only the route to achieve high magnetostrictive but also low coercivity systems. And thus, in Fig. 3 we present the magnetostriction constant as a function of the coercivity. We have just considered those samples with a H_C lower than 300 Oe. It is noteworthy that bulk quenched Galfenol samples show a λ of 400 ppm but in sputtered $Fe_{81.6}Ga_{18.4}$ thin films have been reported maximum values of 150 ppm [20]. Our annealed samples have λ values below 100 ppm when the coercivity is lower than 150 Oe. Nevertheless, λ increases up to 340 ppm when the coercivity is 220 Oe. These optimum values have been achieved in the $[Fe_3Ga(12.5\,nm)/TbFe_2(37.5)\,nm]_7$ heterostructure of set (b). Samples of that set (b) with thicker $TbFe_2$ layers exhibit a huge coercivity while with thinner layers the λ is too small. Then, with this

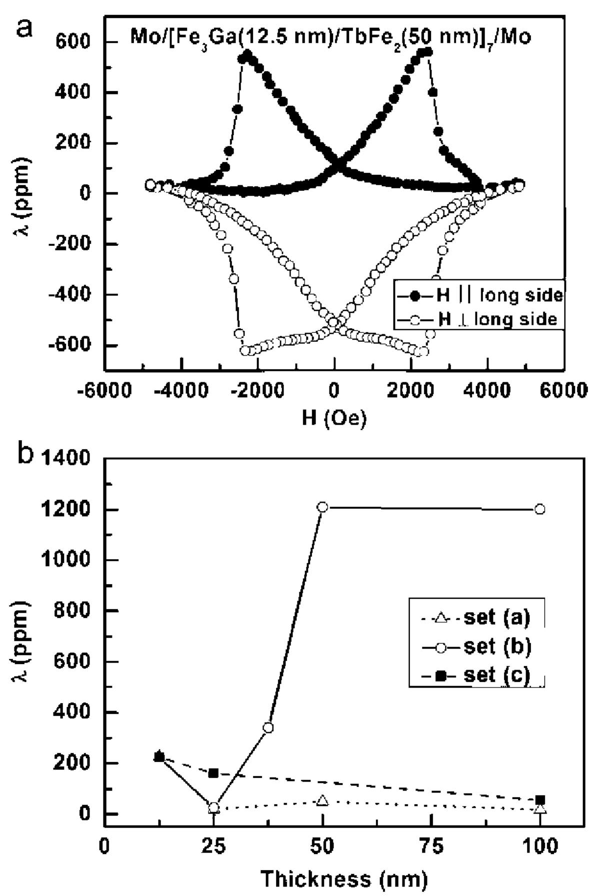

Fig. 2. (a) Magnetostriction as a function of the applied magnetic field of the $[Fe_3Ga(12.5\,nm)/TbFe_2(50\,nm)]_7$. The direction of the magnetic field is parallel (●) and perpendicular (○) to the long side of the cantilever. (b) Magnetostriction constant as a function of the thickness layer for the three set of samples: set (a) (△), set (b) (○), and set c (■).

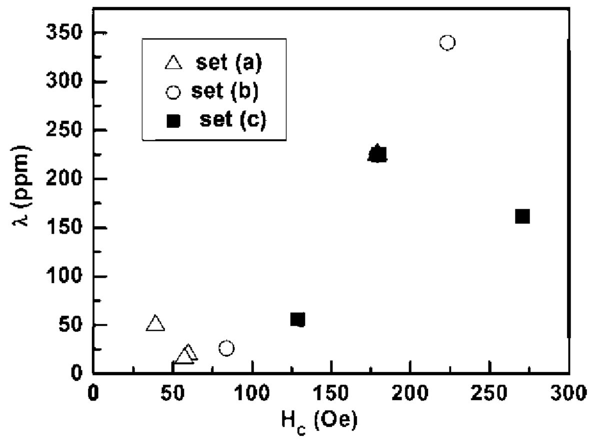

Fig. 3. Magnetostriction constant as a function of the coercivity for the three set of samples: set (a) (△), set (b) (○), and set c (■). We have not included in the graph those samples with a coercivity higher than 275 Oe.

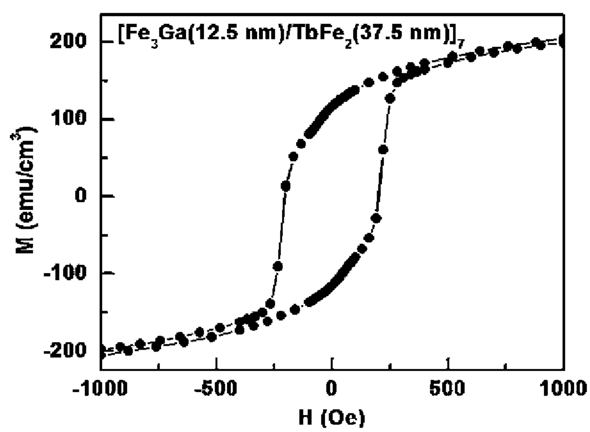

Fig. 4. Hysteresis loop at room temperature of the $[Fe_3Ga(12.5\,nm)/TbFe_2(37.5\,nm)]_7$ heterostructure.

combination of thicknesses, $[Fe_3Ga(12.5\,nm)/TbFe_2(37.5)\,nm]_7$, the $TbFe_2$ is partially crystallized as indicated by its moderate H_C being possible to enhance the magnetostrictive properties without a strong increase of the H_C. In the rest of the samples, the crystallization of the $TbFe_2$ is so negligible that the magnetostriction constant is rather low. Furthermore, the hysteresis loop of the optimized sample, $[Fe_3Ga(12.5\,nm)/TbFe_2(37.5)\,nm]_7$, shows that the hard and soft magnetic layers are strongly coupled since there is just one magnetization jump in the loop [Fig. 4]. Although not so good as those values reported by Quandt and Ludwig in $TbFe_2$/FeCo multilayers [7], $\lambda \sim 400$ ppm and $H_C \sim 25$ Oe, our results show the promising characteristics of the $[Fe_3Ga/TbFe_2]_n$ heterostructures. Therefore, the optimization of the layer thickness appears as a route to engineer the magnetic and magnetostrictive properties of the $[Fe_3Ga/TbFe_2]_n$ multilayers.

4. Conclusions

In conclusion, our experimental results indicate that the combination of hard and soft magnetostrictive materials such as $TbFe_2$ and Fe_3Ga is a promising route to produce new magnetostrictive systems with a low coercivity and a high magnetostriction constant. Multilayers comprised of thick $TbFe_2$ layers show a magnetostriction constant higher than 1×10^3 ppm. Nevertheless, their high H_C of around 4 kOe prevents their use in applications. The layer thickness is an important factor to engineer the magnetic properties of these heterostructures due to the influence of the buffer mechanical stiffness (Y/α) on the $TbFe_2$ crystallization process. $[Fe_3Ga/TbFe_2]_n$ heterostructures show promising characteristics with a λ of 340 ppm and a H_C of 220 Oe.

Acknowledgments

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