Stripe domains in electrodeposited Ni$_{90}$Fe$_{10}$ thin films

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1. Introduction

The interplay between magnetism and mechanical properties gives rise to magnetostrictive effects [1,2]. One way to characterize magnetoelastic materials is by the magnetostriction constant ($\lambda$), that expresses the change of dimension when a magnetic material is magnetized:

$$\lambda = \frac{l_l - l_0}{l_0} \quad (1)$$

being $l$ the length in the magnetic saturated state and $l_0$ the initial length, respectively. For the fabrication of miniaturized spintronics devices, there is nowadays a great interest in developing the same performance in nanometric magnetoelastic systems as in bulk materials. In addition, there is an increasing attention in magnetoelasticity since it can be exploited in spin waves generation [3–6]. Spin waves are collective excitations of the electron spin system, and they have been proposed for applications such as information transfer with low energy dissipation, high-speed technology or analog computing [3,7–10]. In that sense, spin textures play a fundamental role for the stabilization and manipulation of spin waves, and therefore, a huge effort is performing for the development of tunable magnetic structures.

Stripe domains are among the spin textures capable to promote reproducible spin waves generation [11–14]. In materials with moderate perpendicular magnetic anisotropy (PMA), stripe domains are archetypical magnetic configurations that appear due to competing interactions because of the alternating up and down out-of-plane (OOP) orientation of the magnetization [15]. The quality factor ($Q$) gives an idea whether stripe domains can be expected or not in a material system [16]:

$$Q = K_u/2\pi M_{sat}^2 \quad (2)$$

where $K_u$ is the perpendicular magnetic anisotropy constant, and $M_{sat}$ is the saturation magnetization. For materials with moderate or low PMA, i.e. $Q < 1$, stripe domains appear above a theoretical critical thickness ($t_c$):

$$t_c = 2\pi A_{ex}/K_u \quad (3)$$

being $A_{ex}$ the exchange energy per unit length. If $Q > 0.1$, stripe domains are wider than the layer thickness, whereas for $Q < 0.1$ they...
exhibit a periodicity equals to the layer thickness.

The family of Ni-Fe alloys is one of the most extensively studied ferromagnetic metallic systems due to the possibility of reaching small coercivity, large $M_{sat}$, and high permeability by means of composition and growth conditions. In Ni$_{85}$Fe$_{15}$ films, stripes have been observed because of columnar growth in sputtered layers [17] and have been promoted when coupled with other magnetic systems with large PMA as NdCo [18]. However, the null magnetostriiction of Ni$_{85}$Fe$_{15}$ makes it not possible to use it in magnetoelastic applications. Ni$_{85}$Fe$_{15}$ has been previously studied because of its magnetoelastic properties [19–20] since this composition exhibits a λ that can reach ~22 ppm. In addition, evaporated Ni$_{85}$Fe$_{15}$ thin layers on top of Cu can show a large OOP magnetic anisotropy contribution due to mechanical strain [19].

The main goal of this work is to achieve high quality Ni$_{85}$Fe$_{15}$ magnetoelastic thin films with tunable stripe domains. We have studied the influence of growth conditions in terms of both magnetic stirring and an applied magnetic field on the magnetic properties of Ni$_{85}$Fe$_{15}$ electrodeposited layers. Also, we have inferred a λ of ~22 ppm in these layers that reflects their high quality from the magnetoelastic point of view.

2. Experimental section

Ni$_{85}$Fe$_{15}$ layers were grown by electrodeposition on both glass (rigid) and kapton (flexible) substrates covered with a gold layer to increase the electrical conductivity of these two electrically insulating substrates. We have used a three-electrode cell with a platinum mesh as counter electrode and a Ag/AgCl (3 M NaCl) reference electrode. A PalmSens EmStat3 Blue potentiostat was used to perform the depositions that were performed with (500 ppm) and without magnetic stirring in non-rotating substrates at room temperature. In some cases, during growth it has also been applied an external magnetic field of 100 Oe perpendicular to the sample plane. The growth time of NiFe was adjusted to reach the expected thicknesses by means of the Faraday’s law:

\[ C = \frac{nFd}{M_{atom}S} \]  

where $C$ is the electric charge measured in the cathode, $n$ is the number of electrons involved in the reduction reaction, $F$ is the Faraday’s constant (96485,34C/mol), $d$ is the density of the electrodeposited material, $S$ is the area of the sample, $t$ is the expected thickness, and $M_{atom}$ is the molecular mass. In this work, thickness has ranged between 50 nm and 1.2 μm for NiFe layers. For simplicity, we will denote as thick those samples with a thickness equal or larger than 800 nm, and thin those with a thickness below this value.

The electrolyte for Ni$_{85}$Fe$_{15}$ was water-based with a fixed pH of 2.2, and a composition of H$_2$BO$_3$ of 0.4 M, saccharine of 0.017 M, NiSO$_4$ of 0.7 M, and FeSO$_4$ 0.02 M, using a growth potential of −1.2 V in all cases.

We have used X-ray diffractometry (XRD) in the Bragg-Brentano configuration to study the structural properties. Measurements were performed in a D8 Brucker equipment using the Cu K$_\alpha$ wavelength (1.54056 Å). The composition of the layers has been obtained from energy dispersive X-ray spectroscopy (EDS) measurements performed in a JEOL JSM 6400 scanning electron microscope (SEM) working with an accelerator voltage of 20 kV. The experimental error for EDS measurements, ca. 1 at.%, has been inferred after averaging the composition measured in different regions of the samples. In-plane (IP) and OOP hysteresis loops were performed in a vibrating sample magnetometer (VSM) at room temperature. Magnetic Force Microscopy (MFM) measurements [21–22] were performed at ambient conditions using a custom modified system from Nanotec Electrónica S.L. The software WSxM [23] was used for data acquisition. This technique relies on the detection of the long-range magnetostatic force between the sample and a magnetic probe [24]. To detect the magnetic signal, it is necessary to perform two scans, one close to the surface to measure the topography and another one at typically 20–50 nm away from the surface to minimize van der Waals interactions to detect only long-range magnetic interaction. Amplitude modulation (AM) method was carried out enabling the phase-locked loop (PLL) to track the resonance frequency of the oscillating cantilever and the magnetic signal was therefore recorded in the frequency shift channel, in Hz. Assuming that the tip-sample influence is negligible, the positive MFM contrast corresponds to a repulsive interaction, while the negative signal is due to an attractive interaction. In our experiments, the tip oscillates ~20 nm and the second scan was performed at a typical distance of 40 nm. Commercial probes from Budget Sensors MagneticMulti75-G, with CoCr coating were used.

3. Results and discussion

First of all, we have characterized the samples deposited on rigid substrates. The composition of the samples was measured by means of EDS being obtained Ni$_{85}$Fe$_{15}$ in all cases. For the identification of the XRD diffraction peaks we have used the cards 04-024-7186 (Ni$_{85}$Fe$_{15}$) and 00-004-0784 (Au). Apart from the reflections related to the Au buffer layer, the XRD patterns show the (111) and (200) diffraction peaks of NiFe (Fig. 1). The experimental position of the NiFe diffraction peaks is not affected either by magnetic stirring or when a magnetic field perpendicular to the sample plane is applied during growth.

To determine the lattice parameter (a) we have used the Bragg’s law:

\[ n_d\lambda = 2d\sin\theta \]  

where $d_{hkℓ}$ is the distance between the family of planes with $h,k,\ell$ Miller indexes, $θ$ the diffraction angle, and $λ$ the radiation wavelength (Cu K$_\alpha$ in this case). In the NiFe cubic system, we can obtain $a$ from $d_{hkℓ}$ since:

\[ a = d_{hkℓ}\sqrt{h^2+k^2+\ell^2} \]  

In all cases it is obtained a lattice parameter of 3.5 Å pretty close to the expected value extracted from the used XRD file 04–024–7186. Therefore, all the samples exhibit a similar strain state and effects related to an hypothetical magnetoelastic magnetic anisotropy is expected to be the same in all of them.

We have measured IP hysteresis loops to check the possibility of stripe domains in thick Ni$_{85}$Fe$_{15}$ layers grown in different conditions (Fig. 2a). Clearly, we can observe the fingerprint of this type of domains, the characteristic shape denoted as ‘transcritical’ [15,25], when layers are deposited without magnetic stirring. For thick films a perpendicular

![Fig. 1. XRD diffraction patterns of 1.2 μm thick-Ni$_{85}$Fe$_{15}$ layers deposited under different growth conditions: stirring and $H = 0$, non-stirring and $H = 0$, non-stirring and $H = 100$ Oe. Curves have been shifted for clarity.](image-url)
The existence of some remanence in the OOP loops might be un
studied samples (Fig. 2b). Remanence is low although not completely
loop is almost zero due to the presence of stripe domains [15]. In the
transcritical shape is present in the IP loops, the remanence in the OOP
stirring have been confirmed by means of MFM (Fig. 3a). When the
of stripe domains in thick NiFe samples deposited without magnetic
anisotropies that promote an inhomogeneous magnetization.

magnetic field applied during growth does not have any signif-
ificant effect on the transcritical shape of the IP hysteresis loop. On the
contrary, a layer with the same thickness and composition but deposited
in a magnetically stirred electrolyte, shows an IP hysteresis loop char-
acteristic of a soft magnetic alloy without stripe domains. The existence
of stripe domains in thick NiFe samples deposited without magnetic
stirring have been confirmed by means of MFM (Fig. 3a). When the
transcritical shape is present in the IP loops, the remanence in the OOP
loop is almost zero due to the presence of stripe domains [15]. In the
studied samples (Fig. 2b), remanence is low although not completely
null. The existence of some remanence in the OOP loops might be un-
understood considering that the studied samples do not completely fulfill
the requirement of the Murayama’s model [26], that it is generally used
to understand the formation of stripe domains. In that model, it is
needed that the magnetization is aligned in the direction parallel to the
stripes [26–27]. In the studied samples, some magnetization component
misaligned with respect to the stripes direction can promote the
observed remanence.

Considering our results and literature previously published in elec-
trodeposited and sputtered Ni-Fe alloys [17,27–29], we can correlate the
existence of stripe domains to the columnar growth. Moreover, our re-
sults indicate that in the considered as ‘thick’ thickness regime, equal or
above 800 nm, stripe domains are not affected by a perpendicular
magnetic field applied during growth. Therefore, the formation of stripe
domains in thick NiFe10 layers seems to be related to morphological
effects promoted when the electrolyte is not stirred. Then, magnetic
stirring is a fundamental growth parameter to tune stripe domains in this
magnetic alloy as their formation is avoided upon its use. It has not been
possible to check whether a perpendicular magnetic field can promote
stripe domains formation in stirring conditions because this field affects
magnetic stirring in our experimental set-up.

From the IP and OOP hysteresis loops of thick NiFe10 layers it can be
inferred the value of the perpendicular magnetic anisotropy and
calculate both, the theoretical critical thickness and the quality factor.
To obtain \( K_u \) we have followed the work of L. C. Garnier and coworkers
[15]. First, we have quantitatively calculated \( K_u \) by measuring the area
between the magnetization curves measured with the field in the IP and
OOP directions. For thick layers deposited in non-stirring conditions
either with and without a perpendicular applied magnetic field \( K_u \) of
around 500-10^3 erg cm^{-3} has been obtained. Considering the theoretical
value for the saturation magnetization \( M_{sat} = 590 \) emu cm^{-3} [30], it is
calculated a \( Q = 0.023 \) which means the layers have a moderate PMA.
This is in agreement with the fact that periodicity of the stripe domains
observed by MFM in these thick layers is similar to their thickness
(Fig. 3b).

Introducing the experimental value for \( K_u \) in equation (3), and taking
into account the reported value for NiFe10, \( A_{ex} = 1.4 \times 10^{-6} \) erg cm^{-3}
[30], it is calculated a \( t_r = 105 \) nm. This is an upper limit for \( t_r 
considering the method used to calculate \( K_u \). To check this value, we
deposited NiFe10 layers with a thickness ranging from 50 nm to
600 nm, i.e. thin layers, in non-stirring conditions and without an
external magnetic field. In these denoted as thin films, we have not
found evidence of the transcritical shape in the IP hysteresis loops up to
a thickness of at least 500 nm (Fig. 4). The inspection of the MFM images
(Fig. 5) for these thin NiFe10 samples reveals that even for a thickness
of 600 nm, well above the theoretical critical thickness, it is obtained a
contrast known as magnetic ripple [31–32] due to competing irregular
magnetic anisotropies that promote an inhomogeneous magnetization.
In fact, this magnetic ripple is also evident for a thickness of 600 nm
(Fig. 5c) although this sample exhibits a clear transcritical shape in the
IP hysteresis loops (Fig. 4b). Therefore, it seems that above the

Fig. 2. (a) IP hysteresis loops for 1.2 \( \mu \)m thick NiFe10 layers deposited in
different conditions: (–) stirring and \( H = 0 \), (●) non-stirring and \( H = 0 \) Oe, (■)
non-stirring and \( H = 100 \) Oe. (b) IP (●) and OOP (▲) hysteresis loops for 1.2 \( \mu \)m
thick NiFe10 layer deposited without an applied magnetic field and non-
stirring conditions.

Fig. 3. (a) MFM image taken at remanence for NiFe10 layers deposited in
non-stirring and \( H = 0 \) conditions with a thickness of 1.2 \( \mu \)m. Image size: 30 \( \mu \)m
× 30 \( \mu \)m. (b) MFM image of a smaller area and its corresponding profile (c)
where the stripe domain periodicity can be obtained.
theoretical $t_{cr}$ of 105 nm it is first developed a magnetic ripple because the PMA is not large enough to counterbalance other sources of magnetic anisotropy present in the layers. In fact, stripe domains start to be visible by MFM for a thickness of 800 nm that is pretty large in comparison to the theoretical $t_{cr}$ (Fig. 5d). Therefore, it is needed a much higher thickness than the theoretical critical value to observe by MFM stripe domains. This difference between experimental results and the theoretical prediction can be understood considering that the expression $t_{cr} = \frac{2\pi\sqrt{A_{ex}/K}}{\mu U}$ was obtained within the framework of Murayama’s model considering that the local magnetization does not change its direction with respect to the direction parallel to the stripes [26–27]. The magnetic ripple observed by MFM reflects the presence of random sources of anisotropy that can misalign the magnetization being therefore the Murayama’s model no longer valid.

Finally, to check the quality of the thick Ni$_{90}$Fe$_{10}$ layers from the magnetoelastic point of view, we have inferred the value of $\lambda$ following a routine reported in previous works [33–35]. Since the samples studied in this work are polycrystalline, this method can provide an average value for $\lambda$. To do that, a 1.2 μm-thick Ni$_{90}$Fe$_{10}$ layer was grown in non-stirring conditions and without a perpendicular magnetic field on top of an outward bent 150 μm-thick kapton substrate with a radius curvature of $\rho = 3.42$ cm that promoted a compression state in the released layers as sketched in Fig. 6a. To check whether the use of flexible substrates affects the magnetic properties, we have deposited as a sample of reference a 1.2 μm-thick Ni$_{90}$Fe$_{10}$ layer on top of a non-bent kapton substrate. In comparison with thick layers deposited on rigid glass substrates, this sample exhibits a small decrease of $K_{OOP}$ from 500·10$^3$ erg/cm$^3$ to 455·10$^3$ erg/cm$^3$ but with a transcritical shape still visible in the IP hysteresis loop (Fig. 6b).

The induced mechanical deformation ($\epsilon$) after the release of the sample deposited in the outward bent substrate is:

$$\epsilon = \frac{h}{2\rho}$$  

Then, in our particular geometry (Fig. 6a) and for a $h$ that considers
the thickness of both substrate (150 μm) and NiFe layer (1.2 μm) it is
promoted a compression deformation of $\varepsilon = -0.002$ with a compressive stress ($\sigma$):

$$\sigma = \frac{E \varepsilon}{1 - \nu^2}$$

(8)

where $E$ and $\nu$ are the Young’s modulus and Poisson ratio of the NiFe
layer, respectively. In this case, the values are $E = 180$ GPa and $\nu = 0.3$
[36]. Due to this IP compressive stress, it is induced an IP magnetoelastic
anisotropy, that decreases the perpendicular anisotropy to $263 \times 10^3$ erg/cm$^3$. From a linear fit to the expression [1–2]:

$$K = \frac{3}{4} \mu_0 \sigma$$

(9)

in which $K$ is the magnetic anisotropy of compressed and non-
compressed layers grown on flexible substrates, it is inferred a $\lambda$ of
$22$ ppm, pretty close to the reported value in the bulk alloy.

Therefore, this investigation demonstrates the possibility of using the
electrodeposition technique to fabricate magnetoelastic Ni$_{90}$Fe$_{10}$ films
in which the PMA can be tuned thanks to the mechanical deformation.

4. Conclusions

In summary, we have successfully demonstrated the capability of using the electrodiposition technique to grow high-quality magnetoelastic Ni$_{90}$Fe$_{10}$ films. Magnetic stirring appears as a fundamental growth parameter since it avoids the formation of stripe domains. In NiFe samples that exhibit stripes, we have obtained an experimental value of $K_U$ of around $500 \times 10^3$ erg/cm$^2$ which promotes a moderate PMA with a $Q = 0.023$ and a critical thickness of 105 nm. However, above this theoretical critical thickness it is first obtained a magnetic ripple that the PMA can counterbalance other sources of random magnetic anisotropy. Finally, layers with stripe domains also present a high quality from the magnetoelastic point of view as reflected by the $\lambda$ of around $-22$ ppm, pretty close to the reported value in the bulk alloy.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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