Influence of Stress Relaxation on the Magnetization Process of Hitperm-Type Glass-Coated Microwires

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Abstract: The remagnetization process of most amorphous and nanocrystalline glass-coated microwires with positive magnetostriction coefficient occurs through the single and large Barkhausen jump. This article encompasses a study on the magnetization process of thin Hitperm-type glass coated microwires. The complex stress distribution inside these microwires enables us to investigate the influence of; the measuring frequency, applied tensile stresses, as well as current annealing, and conventional annealing at wide range of temperatures. A systematic elucidations have been discussed in the framework of the microwire’s geometries and the shape anisotropy that arises during its fabrication process, with the aim to provide an assessment of the criteria for selecting the necessary conditions to be designed in high-performance sensors.

Keywords: Glass-coated microwires, Nanocrystalline microstructure, Switching field.

INTRODUCTION

The magnetic domain wall (DW) motion is an important topic of research not only because of its potential technological application in several magnetic devices \[1, 2\], but also due to the interest in understanding of the fundamental physics associated with the dynamical processes. In this context, amorphous and nanocrystalline glass-coated microwires are interesting subjects for studying a single domain wall dynamics and the magnetic behavior of systems with reduced dimensions. During the production of these microwires the frozen internal stresses, associated to fast solidification of metallic alloy inside the glass coating give rise to their interesting properties \[3, 4\]. One of the most unusual magnetic features of microwires with positive magnetostriction is the magnetic bistability associated with the fast magnetization switching by a single and large Barkhausen jump as a result of the displacement of single magnetic DW along the wire \[4-8\]. It is also worth mentioning that such property is mostly governed by a peculiar domain structure, as a consequence, the magnetization of these microwires can reach just two values of remanent magnetization \(+/-\) Mr.

Aforementioned domain structure is a consequence of the internal stresses arising during the fabrication process.

One of the internal stresses sources is the simultaneous rapid solidification of the metallic nucleus inside the glass coating \[9, 10\]. Moreover the solidification form the surface produce so-called thermal stresses \[9\]. The strength of internal stresses of glass-coated microwires is mostly determined by the ratio, \(\rho\), between the metallic nucleus diameter, \(d\), and the total composite wire diameter, \(D\), \((\rho=d/D)\) \[3, 9\].

As a result of magnetoelastic interaction between the magnetic moments and stresses introduced during the microwires production, the domain structure of amorphous microwire with positive magnetostriction consists of single axial domain, which is surrounded by a radial domain structure as given schematically in Figure 1 \[3, 4, 6\].

Moreover, small closure domain appears at the end of the microwire in order to decrease the stray fields \[10\]. Existence of these small closure domains appearing at the end of the wire has been experimentally confirmed from the magnetization profile observation \[4\].

Consequently, it is widely assumed, that the switching field is determined by the magnetoelastic
anisotropy that depends on the strength of internal stresses and on the magnetostriction coefficient value. Besides, generally amorphous materials and particularly as-prepared microwires present quite large relaxation effects that can be anticipated in the resulting switching field time instability [11]. In a broader sense, due to this relaxation phenomena an additional magnetic field is required in order to facilitate the domain wall motion out of its position. Such additional pining field is found to be significantly contributed in the total switching field [11]. Finally, the total switching field can be simply summed as:

\[ H_{sw}^P = \frac{1}{M} \left( \varepsilon_p^2 \rho_p \left( 1 - e^{-t/\tau} \right) \right) \]  

(3)

where \( \varepsilon_p^2 \) corresponds to the interaction energy of the mobile defects with spontaneous magnetization, \( \rho_p \) is the density of the mobile defects, \( t \) is the measuring time, and \( \tau \) is the relaxation time which can be expressed by the Arrhenius equation:

\[ \tau = \tau_0 e^{Q/KT} \]  

(4)

where \( \tau_0 \) is the pre-exponential factor and \( Q \) corresponds to the activation energy of the mobile defects.

The coercivity of magnetically bistable materials is, therefore, determined by the switching field value [12, 13]. Coercivity is associated with the pinning wall of the domain wall. Aforementioned relaxation processes as well as thermal activation result in considerable frequency and magnetic field amplitude dependence of the coercivity observed in amorphous wires or ribbons [13, 14]. Thus it was shown, that the frequency dependence of coercivity in various amorphous materials at low enough frequency \( f \) can be phenomenologically expressed as:

\[ H = H_0 + B(fH_0)^{1/n}, \]  

(5)

where \( H_0 \) is the static coercivity, \( H_0 \) is the applied field amplitude and \( n \) is the coefficient ranging from 1 to 4 depending on sample geometry and the type of the hysteresis loop of the studied materials and \( B \) is a coefficient depending on intrinsic parameters of materials.

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Aforementioned frequency and magnetic field amplitude dependence of coercivity has been analyzed for the case of magnetically bistable samples when the magnetization process takes place as the depinning of existing domain wall and consequent domain wall propagation. Switching field increasing and change of the slope of the vertical regions of hysteresis loops have been observed in magnetically bistable materials [13-15]. Aforementioned dependences have been attributed to the counterbalance between the sweeping rate, \( dH/dt = 4fH_0 \) (i.e. increasing of \( H_0 \) results in faster change of magnetic field \( dH \) for the same time interval, \( dt \)) and the switching time related to the time needed for the domain wall propagation along the sample [16].

Nanocrystalline Fe-rich amorphous materials attracted considerable attention because of excellent
soft magnetic properties and obtaining an enhanced GMI effect [17, 18]. Excellent magnetic softness observed in FeSiB NbCu (Finemet) alloys are usually attributed to vanishing magnetocrystalline anisotropy as well as to vanishing magnetostriction value of the material consisting of nano-sized grains, with an average size about 10 nm, embedded in an amorphous matrix [17]. Alternative group of Cu-free nanocrystalline materials - HITPERM alloys with improved soft magnetic properties have been discovered at the end of 90-s [18, 19]. For the Taylor –Ulitovsky technique involving drawing of the metallic alloy surrounding by the glass from the melt at high temperatures Zr-containing alloys are not suitable for the glass-coated microwires fabrication. Therefore, first attempts for HITPERM-type microwires fabrication using Hf instead of Zr have been reported [20]. Recently, a quite high domain wall (DW) velocity up to 3 km/s in HITPERM – type (Fe_{40}Co_{38}Mo_{4}B_{18}) microwires and very stable almost independent on temperature DW dynamics have been reported [21].

Such wires Fe_{40}Co_{38}Mo_{4}B_{18} showed an increased grain size of nanocrystalline phase (19 nm) that deteriorates magnetic softness. It has been shown that Cu addition usually promotes the nucleation of the bcc grains [18, 21]. Hence addition of Cu is used here to obtain finer nanocrystalline structure.

Aforementioned internal stresses are particularly important in the case of high and positive magnetostriction materials like Fe_{40}Co_{38}Mo_{4}B_{18}.

Consequently, our study is focused on the study of influence of glass coating on magnetization switching in glass-coated Fe_{38.5}Co_{38.5}B_{18}Mo_{4}Cu_{1} HITPERM –type microwires.

**MATERIALS AND METHODS**

Studied glass-coated microwires were produced by modified Taylor-Ulitovsky method described elsewhere [3, 4]. Essentially, the laboratory process consists of the drawing of molten metal that fills the glass capillary and a microwire is thus formed where the metal core is completely coated by a glass shell. We concentrated on Hitperm-type glass-coated microwires with the nominal composition Fe_{38.5}Co_{38.5}B_{18}Mo_{4}Cu_{1} and geometries (D= 22.5 μm, d= 9.4 μm, ρ= 0.41) which exhibit magnetic properties typical for the most demanded Fe- microwires with positive magnetostriction constant (see hysteresis loop in Figure 1b) [4].

The switching field, $H_{sw}$, has been measured by induction method using triangular waveform to feed the primary coil in a wide range of frequencies: 10–1000 Hz at the room temperature. When the magnetic field $H$ in the primary coil reaches the value of switching field $H_{sw}$, closure domain propagates along the microwire in a single Barkhausen jump and sharp voltage maximum is induced in the pickup coil. Hence, the value of the switching field can be easily obtained from the position of maximum on the oscilloscope screen. The maximum amplitude of exciting magnetic field $H_0$ was kept constant (1037 A/m) for all measurements.

Joule annealing has been performed at room temperature. The heating process is released by electrical current $I$ passing into the microwire sample rather than by external heating coil as described elsewhere [22]. Sample annealing under or without stress has been performed in a controlled conventional furnace at different temperatures (300-500°C for 1 hour) as described elsewhere [23]. The final value of the applied tensile stresses within the metallic nucleus and the glass layer has been calculated in according to [23].

**RESULTS AND DISCUSSION**

Figure 2 displays the switching field dependence as a function of the measuring frequency of studied sample. The frequency dependence on the switching field as demonstrated in Figure 2 consists of three different regimes as it has been already observed before in similar composition of Hitperm-type glass-coated microwires [24].

As previously discussed a frequency dependence model of coercivity in rapidly quenched amorphous materials depends on few factors and $n$ - coefficient of eq. (5) can take different values [14, 25], i.e.

1. in case of small domain wall mass, $m_{dw}$, and small measuring frequencies.
2. in case of small stiffness coefficient, α, and large measuring frequencies.
3. in case of small domain wall mass, $m_{dw}$, and large measuring frequencies.

At low frequencies, a slight decrease in the switching field has been observed, while upon
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As increasing the frequency (above 50 Hz) the switching field found to be dependent on $f^{1/n}$ [13, 14, 25]. Such decrease at low frequencies can be explained based on the magnetic after effect [26]. As the measuring frequency decreases, the measuring time increases and a stabilization of the domain structure through the structural relaxation takes place [11, 27]. In contrast, at higher frequencies, the magnetoelastic contribution becomes more pronounced, while the relaxation's one has no influence in this range which will be confirmed by the fitting of the experimental data.

In the present sample (Figure 2) the fitting gives parameter $n$ is equal 1. In according to previously published analysis of the frequency dependence of coercivity [14, 25] this case corresponds to small domain wall mass, $m_{sw}$, and low measuring frequencies. Consequently frequency dependence of the switching field can be expressed as:

$$H_{sw}^a = H_{co} + \frac{4fH_0(L + 2M_sA)}{K}$$

(6)

or

$$H_{sw}^a = 3H_{co} + fH_08LM_sA / K$$

(7)

where $M_s$ is the saturation magnetization, and $L$ is the damping coefficient. The field from micro-eddy current can be assumed as proportional to $Ad_x/d_t$ ($A$ is a proportionality constant).

As a result, we can assume that as-prepared Hitperm-type nanocrystalline glass coated microwires has a strong switching field dependence on the measuring frequency according to equation 6-8 depending mostly on both contributions as in equation 1.

As a result of their fabrication process, there is a complex stress distribution of the stresses induced in as-cast microwires. Usually, annealing of these microwires leads to a partial relaxation of the stresses induced during the fabrication processes [4, 9].

Figure 3 shows the frequency dependence of the switching field of as-prepared and annealed samples at various temperatures of both studied microwires. As can be deduced from Figure 3, annealed samples exhibit weak frequency dependence of coercivity. On the other hand as-prepared samples demonstrate complex frequency dependence discussed above. In these findings, all annealed samples show only slight change of the switching field upon increasing of the exciting frequency, and being variable as a function of the annealing temperature. Such change in the switching field value can be understood in the terms of the stress relaxation due to a different thermal expansion coefficient of metallic nucleus, $a_m$, and glass-coating, $a_g$.

Additionally similarly to the case of Finemet-type microwires [15] the influence of the devitrification process and the nano-grains precipitation must be considered for the interpretation of the $H_{sw}(f)$ dependence, In the case of Hitperm-type microwires these precipitation are $\alpha$-Fe-Co nano-grains [28,29]. Annealing at 300°C results in a partial relaxation of the stresses induced during the fabrication, and so switching field decreases due to decreasing of the magnetoelastic contribution.
In according to previously published analysis of the frequency dependence of coercivity [14, 25] for the case if \( n=2 \) (that corresponds to low frequency limits, i.e. when the relaxation time of the domain wall is high) the magnetoelastic contribution can be expressed as

\[
H_{sw}^\sigma = H_{co} + 4A \left( \frac{x_{cr} M_s}{L} + 2M_s A \right) f^2 = H_{co} + 4AP
\]

(8)

where \( x_{cr} \) is the critical displacement of the domain wall to be depinned.

The fitting parameter, \( P \), according to equation 6 is given by:

\[
P = 4A \left( \frac{x_{cr} M_s}{L} + 2M_s A \right)
\]

Annealing temperature, \( T_{ann} \), dependence of the switching field is shown in Figure 4. We can assume that stress relaxation and the nano-grains precipitation affect complex \( H_{sw}(T_{ann}) \) dependence depicted in Figure 4. Nano-grains precipitation can play an important role in the pinning centers for the domain wall displacement depending roughly on their distributions, the free space between them, as well as the exchange length [26,27] As a consequence, a switching field variation must be considered depending on the annealing temperature as well as the grain size significance. Further microstructural XRD studies are still necessary to confirm the nanocrystallization process of the studied sample. Furthermore, similar weak frequency dependence has been observed in case of utilizing current annealing instead of conventional annealing as can be appreciated from Figure 5. It worth mentioning that in case of current annealing a circumferential magnetic anisotropy has been introduced during the annealing process which can induce additional magnetic anisotropy [11]. Consequently we observed switching field decrease with the current value, \( I \), as demonstrated in Figure 5.

The only exception is that there is some increase of the switching field upon annealing at 30 mA.

We have studied the effect of stress annealing (the stress applied during annealing was \( 3g_m=162.9 \text{ MPa} \) and \( 5g_m=270.9 \text{ MPa} \)) on the switching field of studied

![Figure 4: Dependence of the switching field as well as the proportionality constant on the annealing temperature of Fe_{38.5}Co_{38.5}B_{18}Mo_{4}Cu_{1} microwires, measured at 500 Hz.](image)

![Figure 5: Switching field dependence on different values of current annealed Fe_{38.5}Co_{38.5}B_{18}Mo_{4}Cu_{1} samples of both studied microwires, measured at 100 and 1000 Hz.](image)

![Figure 6: Switching field dependence as a function of the measuring frequency of microwires in as-prepared and stress annealed at 300°C Fe_{38.5}Co_{38.5}B_{18}Mo_{4}Cu_{1} samples and different applied stresses.](image)
microwire (see Figure 6). As can be appreciated, stress annealing affects frequency dependence of the switching field dependence. Moreover the value of the switching field has been increased upon applying (162.9 MPa) and then decreased upon increasing the value of the stresses applied during the annealing (Figure 7).

Figure 7: Dependence of the switching field as well as the proportionality coefficient on the applied tensile stresses of studied microwire, measured at 500 Hz.

The magnetoelastic contribution to the switching field is given by equation 2, herein, the mechanical stresses is given by the sum of the stresses induced during the fabrication together with the stresses applied during the measurements; \( \sigma_{\text{total}} = \sigma_i + \sigma_{\text{app}} \). Among these result presented in Figures 6, 7, an increase in the parameter, \( P \), has been observed. This increasing of \( P \)-coefficient reflects an increasing of the magnetoelastic anisotropy induced upon stress annealing under stresses.

Observed dependences can be explained considering the following features: usually thermal treatments affect the magnetoelastic anisotropy. Moreover after annealing the magnetoelastic anisotropy drastically decreases. On the other hand, after annealing (especially in the presence of applied stresses) induced magnetic anisotropy can play an important role in amorphous materials. Thus annealing at certain temperature but below the Curie temperature induces a macroscopic magnetic anisotropy with a preferred axis determined by the direction of the magnetization during the annealing [30]. In addition field induced magnetic anisotropy depends on the annealing temperature.

The microscopic origin of this field-induced anisotropy has been successfully explained considering the directional ordering of atomic pairs mechanism developed by Néel [30-33]. Consequently macroscopically isotropic amorphous alloys can exhibit macroscopic magnetic anisotropy in the case if they are subjected to appropriate annealing treatments at the presence of either a magnetic field (field annealing) or a mechanical stress (stress annealing).

In case of glass-coated microwires the situation is even more complex, because the presence of the glass-coating induces strong internal stresses. These internal stresses must be considered as the factor that can affect magnetic anisotropy after annealing of glass-coated microwires even without applied stresses. Particularly unusually strong effect of annealing on overall shape of hysteresis loops and hysteretic magnetic properties (coercivity, magnetic permeability, etc) has been explained considering effect of stress+field annealing [34, 35]. Previously we performed stress annealing of Fe-rich glass-coated microwires and we observed drastic changes of hysteresis loops, increasing of magnetic permeability, GMI effect and decreasing of coercivity [23]. These changes in amorphous microwires containing only one magnetic element (Fe) have been explained considering the so-called “back” stresses arising from glass-coating and compensating internal stresses induced during fabrication process with the axial component predominant in most of the metallic nucleus volume. Considering aforementioned we can predict that application of tensile stress during annealing must reinforce above reported tendency on increasing the switching field after annealing.

CONCLUSIONS

Recapping the headlines of the presented work, we can conclude that the switching field dependence of as-prepared samples shows complex frequency dependence in glass-coated microwires. This frequency dependence has been discussed in terms of two contributions to the switching field mechanism. Conventional annealing results in drastic change of the frequency dependence of switching field. This effect might be interpreted considering the precipitation of small grains and stress relaxation. Stress annealing induces a macroscopic magnetic anisotropy and hence affect frequency dependence of switching field and the switching field value. Understanding the strong and complicated stress distribution introduced throughout the microwire’s fabrication allowed us to tailor the
magnetoelastic anisotropy in order to obtain a different results that could deserve employing in many sensors application.

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