Advanced Functional Amorphous Magnetic Microwires for Technological Applications

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***Abstract*—Several routes allowing the development of low cost magnetic microwires coated by insulating, flexible and biocompatible glass-coating with tunable magnetic properties are overviewed. Amorphous microwires can present excellent magnetic softness and Giant MagnetoImpedance (GMI) effect. A high GMI effect, obtained even in as-prepared Co-rich microwires, can be further improved by appropriate heat treatment (including Joule heating, conventional annealing and/or stress-annealing). The observed versatile magnetic properties of amorphous microwires are suitable for various applications, such as magnetic sensors, electronic surveillance, wireless communication or biomedical applications.**

Keywords- magnetic microwires; magnetic softness; giant magnetoimpedance effect.

I. INTRODUCTION

Amorphous magnetic materials can present an unusual combination of excellent magnetic properties (e.g., high magnetic permeability, Giant magnetoimpedance (GMI) effect, magnetic bistability, Matteucci and Widemann effects) and superior mechanical properties (plasticity, flexibility) making them suitable for numerous industrial applications [1]-[7]. Furthermore, the preparation method involving rapid melt quenching is quite fast and inexpensive and the above mentioned magnetic softness can be realized without any complex post-processing treatments [3]-[5].

The development of novel applications of amorphous materials requires new functionalities, i.e., reduced dimensions, enhanced corrosion resistance or biocompatibility [8]. Therefore, great attention has been paid to development of alternative fabrication methods allowing preparation of amorphous materials at micro-nano scale involving melt quenching [6]-[8].

The main technological interest in GMI effect is related to one of the largest sensitivity to magnetic field (up to 10 %/A/m) among non-cryogenic effects [4]-[8]. Such features of the GMI effect make it quite attractive for development of high performance sensors allowing detection of low magnetic fields and mechanical stresses [9]-[14]. The most common quantity for the characterization of the GMI effect is the GMI ratio, $\Delta Z/Z$, defined as:

$$\Delta Z/Z = [Z(H) - Z(H_{max})] / Z(Hmax), \quad (1)$$

where H is the applied axial *DC*-field with a maximum value, H_{max}, up to a few kA/m.

The value of GMI ratio and its magnetic field dependence are determined by the type of magnetic anisotropy: to achieve a high GMI ratio, a high circumferential magnetic permeability is essential [7][8]. Magnetic wires with circumferential easy axis exhibit double-peak magnetic field dependence of the real component of wire impedance (and consequently of the GMI ratio). However, magnetic wires with longitudinal easy axis present monotonic decay of the GMI ratio with increasing axial magnetic field with GMI ratio maximum at zero magnetic fields [7][8]. The highest GMI ratio up to 650% is reported for amorphous microwires [15]-[17]. However, the theoretically predicted maximum GMI ratio is about 3000% (i.e., a few times larger than the GMI ratio values reported experimentally) [18]. Additionally, theoretical minimum of the skin depth is about 0.3 μm [17][18].

The main features of the GMI effect have been successfully explained in terms of classical electrodynamics considering the influence of a magnetic field on the penetration depth of an electrical current flowing through the magnetically soft conductor [1][2]. High circumferential permeability typically observed in Co-rich amorphous wires with nearly-zero magnetostriction coefficient is essentially relevant for observation of high GMI ratio [1][2][4]-[6]. However, similarly to the magnetic permeability, the GMI effect has a tensor character [4]-[6][19]-[22]. The off-diagonal component of GMI can present anti-symmetrical magnetic field dependence with a linear region quite suitable for magnetic sensors applications [19][23].

One of the tendencies in modern GMI sensors is the size reduction. It must be underlined that the diameter reduction must be associated with the increasing of the optimal GMI frequency range: a tradeoff between dimension and frequency is required in order to obtain a maximum GMI

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effect [4]-[6][23]. Additionally, the GMI effect at microwave frequencies has been described considering the analogy between the GMI and the ferromagnetic resonance [4]. Consequently, the development of thin soft magnetic materials required for miniaturization of the sensors and devices requires an extension of the frequency range for the impedance toward the higher frequencies (GHz range).

Recently developed magnetic sensors using the GMI effect allow achieving nT and pT magnetic field sensitivity with low noise [10]-[14][24].

Presently, major attention is focused on high frequencies (GHz range) GMI applications owing to the development of thin magnetically soft materials and the recent tendency in miniaturization of magnetic field sensors [4]-[6][10]-[14][24].

The aim of this report is to provide recent results on the optimization of soft magnetic properties and the GMI effect in magnetic microwires.

The rest of the paper is structured as follows. In Section 2, we present the description of the experimental techniques, while in Section 3, we describe the results on the effect of post-processing on the GMI ratio of the studied microwires. We conclude this work in Section 4.

II. EXPERIMENTAL DETAILS

As already mentioned in the introduction, the GMI effect usually observed in soft magnetic materials phenomenologically consists of the change of the AC impedance, $Z = R + iX$ (where R is the real part, or resistance, and X is the imaginary part, or reactance), when submitted to an external magnetic field, H_0.

The electrical impedance, Z, of a magnetic conductor is given by [1][2]:

$$Z = R_{dc}\, krJ_0(kr)/2J_1(kr) \qquad (2)$$

with $k = (1 + j)/\delta$, where J_0 and J_1 are the Bessel functions, r is the wire's radius and δ the penetration depth given by:

$$\delta = \sqrt{\pi\sigma\mu_\phi f} \qquad (3)$$

where σ is the electrical conductivity, f the frequency of the current along the sample, and μ_ϕ the circular magnetic permeability assumed to be scalar. The DC applied magnetic field introduces significant changes in the circular permeability, μ_ϕ. Therefore, the penetration depth also changes through and finally results in a change of Z [1],[2].

The GMI ratio, defined as $\Delta Z/Z$, has been evaluated considering (1).

The use of a specially designed micro-strip sample holder placed inside a sufficiently long solenoid allows measuring of the magnetic field dependence of sample impedance, Z, using a vector network analyzer , as described in [24]. The described technique allows measuring of the GMI effect in extended frequency, f, range up to GHz frequencies.

Hysteresis loops have been measured using the fluxmetric method previously described in [25]. We represent the normalized magnetization, M/M_0 versus the magnetic field, H, where M is the magnetic moment at a given magnetic field and M_0 is the magnetic moment of the sample at the maximum magnetic field amplitude, H_m.

We studied Fe- and Co- rich microwires with metallic nucleus diameters, d, ranging from 10 up to 25 μm prepared using the Taylor-Ulitovsky method described in [5][8]. The Taylor-Ulitovsky method allows the preparation of the thinnest metallic wires (with typical diameters of the order of 1 to 30 μm) covered by an insulating glass coating [5][8].

The great advantage of these microwires is that the obtained diameter could be significantly reduced in comparison with the case of amorphous wires produced by the other rapidly quenching methods. However, in the case of glass-coated microwires the magnetoelastic anisotropy contribution is even more relevant since the preparation process involves not only the rapid quenching itself, but also simultaneous solidification of the metallic nucleus surrounded by the glass-coating with rather different thermal expansion coefficients [5][8][27][28].

In amorphous materials, the magnetocrystalline anisotropy is absent. Therefore, the magnetoelastic anisotropy is the main factor affecting the magnetic properties [5][6].

The magnetoelastic anisotropy, K_{me}, is given as:

$$K_{me} = 3/2\lambda_S\sigma_i \qquad (4)$$

where λ_S is the magnetostriction coefficient and σ_i is the internal stresses value [8].

The magnetostriction coefficient, λ_s, value in amorphous alloys can be tailored by the chemical composition [29]-[31]. Generally, Fe-rich compositions present positive λ_s - values (typically $\lambda_s \approx 20 - 40 \times 10^{-6}$), while for the Co-rich alloys, λ_s values are negative, typically $\lambda_s \approx -5$ to $- 3 \times 10^{-6}$. Vanishing λ_s values can be achieved in the Co_xFe_{1-x} ($0 \leq x \leq 1$) or Co_xMn_{1-x} ($0 \leq x \leq 1$) systems at x about 0.03 – 0.08 [29]-[32].

However, internal stresses, σ_i, arise during simultaneous rapid quenching of metallic nucleus surrounding by the glass coating due to the different thermal expansion coefficients. Consequently, the strength of internal stresses can be controlled by the glass-coating thickness: the strength of internal stresses increases with the increase of the glass-coating thickness [27][28].

III. EXPERIMENTAL RESULTS AND DISCUSSION

As mentioned above, the magnitude and the magnetic field dependence of the GMI effect (including off-diagonal components) is intrinsically linked to the magnetic anisotropy [4]-[8]. Consequently, both hysteresis loops, $\Delta Z/Z(H)$ dependence and maximum value of the GMI ratio, $\Delta Z/Z_m$, are affected by λ_s sign and value and by the magnitude of internal stresses, σ_i . The magnetostriction

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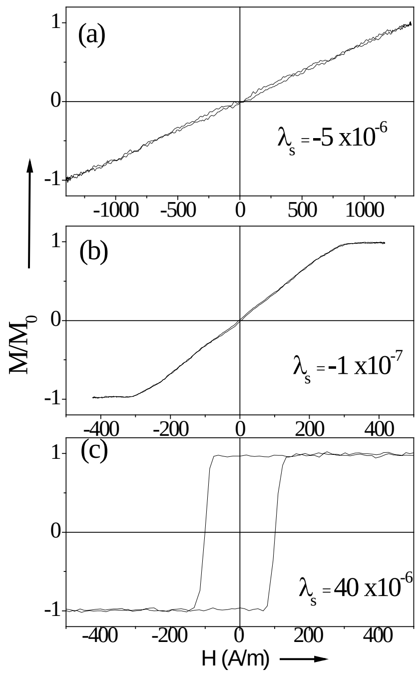

Figure 1. Hysteresis loops of as-prepared $Co_{77.5}Si_{15}B_{7.5}$ (a), $Co_{67.1}Fe_{3.8}Ni_{1.4}Si_{14.5}B_{11.5}Mo_{1.7}$ (b) and $Fe_{75}B_9Si_{12}C_4$ (c) microwires.

coefficient drastically affects the character of the hysteresis loops of magnetic microwires: i) Co-rich microwires (see Figure 1a for $Co_{77.5}Si_{15}B_{7.5}$) with negative magnetostriction constant ($\lambda_s \approx -5 \times 10^{-6}$) have almost unhysteretic loops with extremely low coercivity, H_c. However, the magnetic permeability of $Co_{77.5}Si_{15}B_{7.5}$ microwires is not high enough since they also present high enough magnetic anisotropy field, H_k. ii) Co-Fe-based microwires with vanishing magnetostriction constant ($Co_{67.1}Fe_{3.8}Ni_{1.4}Si_{14.5}B_{11.5}Mo_{1.7}$, $\lambda_s \approx -10^{-7}$) generally present lower H_k values and hence higher magnetic permeability (see Figure 1b). iii) Finally, Fe-rich microwires ($Fe_{75}B_9Si_{12}C_4$) with positive magnetostriction constant ($\lambda_s \approx 40 \times 10^{-6}$) present rectangular hysteresis loops and consequently low magnetic permeability (see Figure 1c).

As can be appreciated from Figure 2b, $Co_{67.1}Fe_{3.8}Ni_{1.4}Si_{14.5}B_{11.5}Mo_{1.7}$ microwire presents the highest maximum GMI ratio, $\Delta Z/Z_m$ (about 240% at 500 MHz). Quite low $\Delta Z/Z_m$ values are observed for $Fe_{75}B_9Si_{12}C_4$ microwire ($\Delta Z/Z_m \approx 15\%$, see Figure 2c). Moderate $\Delta Z/Z_m$ values ($\Delta Z/Z_m \approx 120\%$) are observed for $Co_{77.5}Si_{15}B_{7.5}$ microwire (see Figure 2a).

The other difference in $\Delta Z/Z(H)$ dependencies for microwires with different magnetostriction coefficients is the character of $\Delta Z/Z(H)$ dependencies: for microwires with $\lambda_s>0$, a single maximum $\Delta Z/Z(H)$ dependence with $\Delta Z/Z$ maximum at $H=0$ is observed (Figure 2c). However, for $\lambda_s<0$ double- maximum $\Delta Z/Z(H)$ dependencies with $\Delta Z/Z$ maximum at $H=H_m$ are observed (Figures 2b,c).

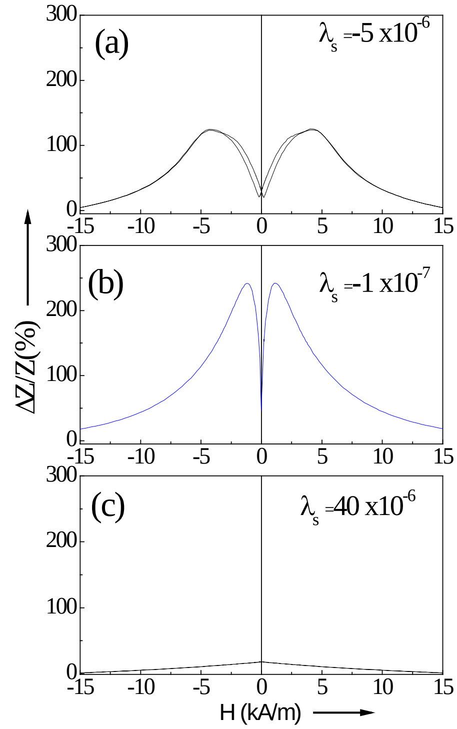

Figure 2. ΔZ/Z(H) dependencies of as-prepared $Co_{77.5}Si_{15}B_{7.5}$ (a), $Co_{67.1}Fe_{3.8}Ni_{1.4}Si_{14.5}B_{11.5}Mo_{1.7}$ (b) and $Fe_{75}B_9Si_{12}C_4$ (c) microwires measured at 500 MHz.

It is commonly assumed that the H_m value corresponding to the peaks (maximum $\Delta Z/Z$ value) is linked to the average value of the anisotropy field, H_K, at high frequency values, and to the effective anisotropy distribution in the sample. In this regard, the observed $\Delta Z/Z(H)$ dependencies correlate with the hysteresis loops: the highest H_m value is observed for $Co_{77.5}Si_{15}B_{7.5}$ microwire with the highest H_k value (see Figure 1a). A single maximum $\Delta Z/Z(H)$ dependence with $\Delta Z/Z$ maximum at $H=0$ corresponds to the $Fe_{75}B_9Si_{12}C_4$ microwire with axial magnetic anisotropy (Figure 1c).

Such different magnetic anisotropy of microwires with positive and negative magnetostriction is related to the internal stresses distribution intrinsically related to the fabrication of microwires [4]-[8]. The radial distribution of internal stresses calculated considering quenching stresses related to rapid quenching of the metallic alloy from the melt as well as complex tensor stresses related to the difference in the thermal expansion coefficients of metal and glass the axial stresses are the largest ones up to ~ 0.85 R (where R is the metallic nucleus radius) [8]. Thus, the main volume of the microwire nucleus is under the tensile stresses near the axis of the metallic nucleus. However, closer to the surface, the compressive stresses are dominant. Additionally, the strength of internal stresses is determined by the thickness of the non-magnetic glass-coating: the strength of internal stresses increases with the increasing of the glass-coating thickness.

Therefore, as reported earlier [5][8], hysteresis loops and GMI effect are affected by the ratio $\rho=d/D$, where d is the

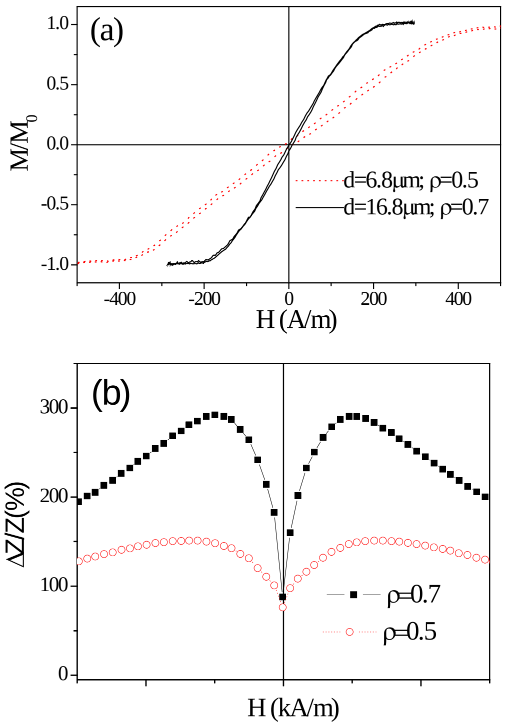

Figure 3. Hysteresis loops (a) and *ΔZ/Z(H)* dependencies measured at 500 MHz (b) of as-prepared $Co_{67}Fe_{3.85}Ni_{1.45}B_{11.5}Si_{14.5}Mo_{1.7}$ microwires with different ρ-ratios.

diameter of metallic nucleus and *D*-total microwire diameter. Some examples are shown in Figure 3, where the hysteresis loops and *ΔZ/Z(H)* dependencies of as-prepared $Co_{67}Fe_{3.85}Ni_{1.45}B_{11.5}Si_{14.5}Mo_{1.7}$ microwires with different *ρ* ratios are shown.

Consequently, the control of internal stresses by tailoring of the *ρ*–ratio is an effective method for GMI ratio tuning.

As mentioned above, the other important parameter for GMI ratio optimization in magnetic microwires is the frequency. Indeed, the frequency must be high enough in order to have the skin depth lower than the sample radius (strong skin effect). *ΔZ/Z(H)* dependencies measured at different frequencies in as-prepared $Co_{67}Fe_{3.9}Ni_{1.4}B_{11.5}Si_{14.5}Mo_{1.6}$ (d=25.6 μm, D= 26.6 μm) microwires are shown in Figure 4a. This composition at the given geometry (d=25.6 μm, D= 26.6 μm, ρ=0.96) present high maximum GMI ratio, $\Delta Z/Z_m$: at optimal frequency of about 300 MHz $\Delta Z/Z_m$ ≈550% can be achieved (see Figure 4b). However, thinner (d=10.8 μm) microwire of the same chemical composition at this frequency exhibit $\Delta Z/Z_m$ ≈400% (see Figure 4b). From the $\Delta Z/Z_m(f)$ dependence for $Co_{67.7}Fe_{4.3}Ni_{1.6}Si_{11.2}B_{12.4}C_{1.5}Mo_{1.3}$ microwires with d=10.8μm and d=25.6 μm, we can appreciate that for $Co_{67.7}Fe_{4.3}Ni_{1.6}Si_{11.2}B_{12.4}C_{1.5}Mo_{1.3}$ microwires with d=10.8μm the optimal frequency is about 700 MHz at which $\Delta Z/Z_m$ ≈550% can be achieved.

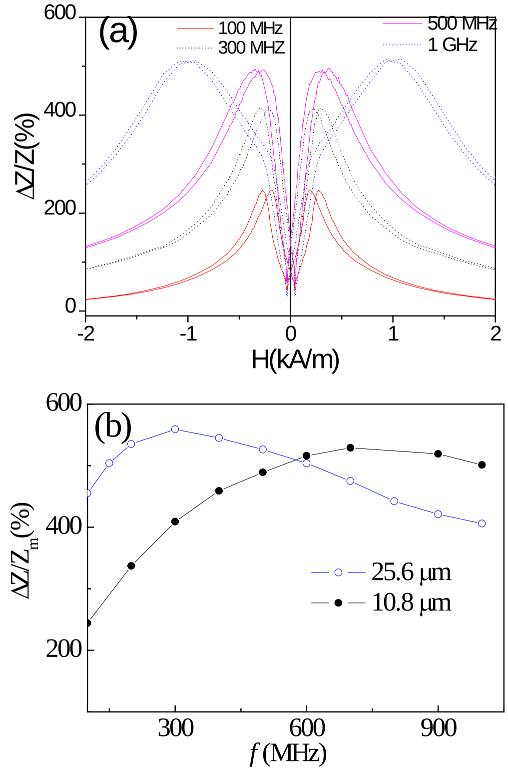

Figure 4. *ΔZ/Z(H)* dependencies measured in as-prepared $Co_{67}Fe_{3.9}Ni_{1.4}B_{11.5}Si_{14.5}Mo_{1.6}$ (d=25.6 μm, D= 26.6 μm) microwires (a) and $\Delta Z/Z_m(f)$ dependence for $Co_{67.7}Fe_{4.3}Ni_{1.6}Si_{11.2}B_{12.4}C_{1.5}Mo_{1.3}$ with d=10.8μm, D=13.8μm and d=25.6 μm, D= 26.6 μm microwires.

The aforementioned examples provide the routes for optimization of GMI effect in Co-rich microwires.

IV. CONCLUSIONS

We measured the GMI magnetic field, frequency dependencies and hysteresis loops in magnetic microwires produced by the Taylor-Ulitovsky technique.

We observed that the GMI effect and magnetic softness of microwires are intrinsically related and can be tailored either by controlling the magnetoelastic anisotropy of as-prepared microwires or by controlling their internal stresses and structure by heat treatment. Studies of the GMI effect of amorphous Co-Fe rich microwires reveal that microwires of appropriate chemical composition and geometry present the GMI effect at GHz frequencies. A high GMI effect has been achieved and discussed. The election of appropriate measuring conditions can be beneficial for the optimization of the GMI effect of magnetic microwires. Magnetic microwires with optimized magnetic properties are suitable for several applications, like magnetic sensors, electronic surveillance, wireless communication or biomedical applications.

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