Development of magnetically soft microwires with GMI effect

V. Zhukova, M. Ipatov and A. Zhukov
Dpto. de Fís. Mater., UPV/EHU San Sebastián 20009, Spain

E-mail: arkadi.joukov@ehu.es

Abstract. Thin amorphous magnetically soft microwires attract recently great attention because of their excellent soft magnetic properties and giant magneto-impedance (GMI) effect, thin dimensions and possibility for applications in magnetic micro-sensors. We overview research in the field of development of microwires with high GMI effect and improved features and report novel results on studies of GMI (diagonal and off-diagonal components) at high frequencies (between 10 MHz and 4 GHz) and its correlation with soft magnetic behaviour of thin amorphous microwires (Co-Fe-rich with nearly-zero magnetostriction constant) with metallic diameter between 3 and 20 µm and on optimization of GMI effect. We studied and analyzed low-field hysteresis of GMI effect and its dependence on circular magnetic field and discuss the nature the low-field hysteresis in terms of helical magnetic anisotropy and the effect of the bias field. Choosing samples composition, annealing conditions and geometry we were able to tailor their magnetoelastic anisotropy and respectively magnetic softness and GMI.

1. Introduction
Recently studies of soft magnetic properties and GMI effect of thin glass coated microwires attracted considerable interest [1-3]. Most reported results correspond to the microwires with the diameter about 20-30 µm. As previously reported elsewhere [2,3] the hysteretic magnetic properties of glass-coated microwires are rather different from those of conventional amorphous wires mostly because of additional magnetoelastic energy related with strong internal stresses induced during the rapid solidification of the thin wire surrounded by the glass coating when using Taylor-Ulitovsky method [4]. Recently certain progress has been achieved in enhancement of magnetic softness and GMI effect of glass coated microwires, paying most attention on alloy composition and post-fabrication processing of microwires aiming improvement of the magnetic permeability and GMI effect through the diminishing of the magnetoelastic anisotropy related with strong internal stresses and induction of desirable magnetic anisotropy. Consequently, adequate choosing of the geometric parameters determined by the fabrication procedure and conditions of heat treatment can significantly enhance soft magnetic properties and GMI effect (up to 600%)[5,6] In most of applications high linearity of signal and low hysteresis are desirable. Anti-symmetrical signal with linear region has been obtained in current pulsed scheme excitation of wires using detection of off-diagonal GMI component [7,8]. At the same time we recently showed, that linearity and shape of off-diagonal component in microwires can be tailored by thermal treatment [9]. At the same time, considerable GMI hysteresis has been observed and analyzed in microwires at low field region [10].
In this paper we review and analyze the results on GMI effect in thin microwires paying special attention to tailoring the GMI effect of thin glass-coated amorphous microwires (with the metallic nucleus diameter, \(d\) below 15 \(\mu m\)) by choosing the sample geometry (ratio, \(\rho\), between metallic nucleus diameter, \(d\) and the total microwire diameter, \(D\)) and annealing conditions by the Joule heating with the aim to improve magnetic field and stress sensitivity of the GMI effect and diminish the GMI hysteresis.

2. Tailoring of magnetic properties and GMI effect in thin glass-coated microwires

2.1. Effect of microwire geometry

As mentioned above, the strength of the internal stresses, \(\sigma\), is related with simultaneous solidification of metallic nucleus inside glass coating and most important contribution is induced by the difference in the thermal expansion coefficients of metallic nucleus and outer glass coating solidifying simultaneously [2-4, 11,12]. Chemical composition of the alloy has been fixed on the basis of previous knowledge of magnetically soft behavior of amorphous ribbons [13]. The effect of the \(\rho\) ratio on the hysteresis loops of nearly-zero magnetostrictive \(\text{Co}_{67}\text{Fe}_{3.85}\text{Ni}_{1.45}\text{B}_{11.5}\text{Si}_{14.5}\text{Mo}_{1.7}\) microwires is shown in Fig.1. All studied samples exhibited inclined almost unhysteretic \(M(H)\) loops with extremely low coercivities (up to 4 A/m). Magnetic anisotropy field, \(H_k\), is found to be determined by the \(\rho\)–ratio, increasing with \(\rho\).

Such \(H_k(\rho)\) dependence has been attributed to the magnetoeelastic energy contribution given by

\[
K_{me} = \frac{3}{2} \lambda_s \sigma_i,
\]

(1)

\[
\lambda_s = \frac{\mu_0 M_s}{3} \left( \frac{dH_k}{d\sigma} \right),
\]

(2)

where \(\lambda_s\) is the saturation magnetostriction and \(\sigma_i\) is the internal stress. The magnetostriction constant is mostly determined by the chemical composition and achieves almost nearly-zero values in amorphous alloys based on Fe-Co with Co/Fe \(\approx 70/5\) \(\lambda_s \approx 0\) [2]. On the other hand, the estimated values of the internal stresses in these glass coated microwires arising from the difference in the thermal expansion coefficients of metallic nucleus and glass coating are of the order of 100-1000 MPa, depending strongly on the ratio between the glass coating thickness and metallic core diameter [2-4, 11,12], increasing with decreasing \(\rho\)–ratio. Considering elevated internal stresses stress dependence of magnetostriction constant should be taken into account as well [2-4]:

Fig.1. Hysteresis loops of \(\text{Co}_{67}\text{Fe}_{3.85}\text{Ni}_{1.45}\text{B}_{11.5}\text{Si}_{14.5}\text{Mo}_{1.7}\) microwires with different geometry (a) and dependence of \(H_k\) on \(\rho\)–ratio (b).
where $\mu_0 M_s$ is the saturation magnetization.

Considering abovementioned we should assume that any method allowing change the internal stresses (by using thermal treatment, chemical etching, controlling microwire geometry) can change drastically magnetic anisotropy and consequently the hysteresis loops and the GMI behavior of glass coated microwires.

2.2. Tailoring of magnetic properties by heat treatment and induced magnetic anisotropy

Heat treatment resulting in structural and stress relaxation in amorphous alloys is a commonly used method for tailoring the magnetoelastic energy [2]. In the case if the alloy composition contains more than one transition metal the pair ordering mechanism of induced magnetic anisotropy especially if magnetic field and/or stress is applied during the heat treatment should be taken into account [2,4,14].

Effect of current annealing (CA) and magnetic field current annealing (FCA) on hysteresis loop of Co$_{67}$Fe$_{3.85}$Ni$_{1.45}$B$_{11.5}$Si$_{14.5}$Mo$_{1.7}$ is shown in Fig.2. Axial magnetic field during FCA (about 8000A/m) has been much higher than circular magnetic field created by the current. As can be appreciated from comparison of Figs 2a and 2b, application of magnetic field during annealing results in induction of axial magnetic anisotropy.

Application of stress during stress annealing of Fe$_{74}$B$_{13}$Si$_{11}$C$_2$ microwires resulted in induction of considerable stress induced anisotropy [15]. In this case rectangular hysteresis loop associated with the strong axial magnetic anisotropy induced by residual stresses of mostly tensile origin after stress annealing converted into inclined hysteresis loop with large enough magnetic anisotropy field [15]. In fact the hysteresis loops can be tailored varying the time or temperature of stress annealing and consequently a variety of hysteresis loop with different magnetic anisotropy can be obtained in Fe$_{74}$B$_{13}$Si$_{11}$C$_2$ microwires, as can be observed in Fig.3a. Such behavior should be attributed to the compressive stresses induced by the SA compensating axial stresses due to the simultaneous solidification of the metal and glass.

The strength of such compressive stresses depends on both time and temperature of annealing.

Under applied stress, the hysteresis loop changes drastically, exhibiting enhanced stress sensitivity (Fig.3b).

Another interesting effect related with such stress sensitivity of hysteresis loop is stress-impedance, i.e. impedance change under applied stress observed in samples with induced stress anisotropy (see Fig.4)
2.3. Tailoring of the GMI effect (including off-diagonal component)

Like in the case of hysteresis loops the sample geometry strongly affects the GMI effect, i.e. magnetic field dependences of impedance (Fig.5). Here we present magnetic field, $H$, dependence of real part, $Z_1$, of the longitudinal wire impedance $Z$ ($Z = Z_1 + iZ_2$). Indeed we observe (see Fig.5) that the sample geometry strongly affects the DC axial magnetic field dependence of the real part of GMI, $Z_1$ of $\text{Co}_{67}\text{Fe}_{3.85}\text{Ni}_{1.45}\text{B}_{11.5}\text{Si}_{14.5}\text{Mo}_{1.7}$ microwires measured at different frequencies $f$. 

In fact one should take into account that both magnetic permeability as well as magneto-impedance have tensor character. The off-diagonal components possess the asymmetrical dependence on magnetic field, that is a necessary condition for determination the magnetic field direction [7,8]. Figs. 6,7 show field dependence of the off-diagonal voltage response, $V_{\text{out}}$ measured using pulsed scheme as described elsewhere [7-9] in $\text{Co}_{67}\text{Fe}_{3.85}\text{Ni}_{1.45}\text{B}_{11.5}\text{Si}_{14.5}\text{Mo}_{1.7}$ ($\alpha_5 = 3 \times 10^{-7}$) microwire with different geometry: metallic nucleus diameter and total diameter with glass coating are 6.0/10.2, 7.0/11.0 and 8.2/13.7 µm. Such considerations regarding dependence of the GMI response on the samples geometry should be taken into account for development of extremely thin microwires. It is important for microminiaturized sensor application since the demagnetizing factor starts to affect the domain structure and magnetic properties when geometric ratio of microwire diameter to its longitude becomes lower [10]. It should be assumed that the internal stresses relaxation after heat treatment should drastically change both the soft magnetic behavior and $\Delta Z/Z(H)$ dependence mostly because of the stress dependence of the magnetostriction described in ref [1]:

Fig.3 Hysteresis loops of $\text{Fe}_{74}\text{B}_{13}\text{Si}_{11}\text{C}_{2}$ microwire annealed under applied stress of 500 MPa (a) at (1) – 300 °C 3 hours, (2) – 280 °C 40 min, (3)- 265 °C 40 min, (4) 235 °C 40 min and (5)- 215 °C 40 min and (b) stress induced changes of hysteresis loops of the same microwires (1- measured under applied stress, 2- measured without stress).

Fig.4 Stress impedance effect of stress annealed $\text{Fe}_{74}\text{B}_{13}\text{Si}_{11}\text{C}_{2}$ glass-coated microwire under stress (468 MPa) at 275°C for 0.5h measured at frequency, $f=10$ MHz for the driving current amplitude of 2 mA.

2.3. Tailoring of the GMI effect (including off-diagonal component)

Like in the case of hysteresis loops the sample geometry strongly affects the GMI effect, i.e. magnetic field dependences of impedance (Fig.5). Here we present magnetic field, $H$, dependence of real part, $Z_1$ of the longitudinal wire impedance $Z$ ($Z = Z_1 + iZ_2$). Indeed we observe (see Fig.5) that the sample geometry strongly affects the DC axial magnetic field dependence of the real part of GMI, $Z_1$ of $\text{Co}_{67}\text{Fe}_{3.85}\text{Ni}_{1.45}\text{B}_{11.5}\text{Si}_{14.5}\text{Mo}_{1.7}$ microwires measured at different frequencies $f$. 

In fact one should take into account that both magnetic permeability as well as magneto-impedance have tensor character. The off-diagonal components possess the asymmetrical dependence on magnetic field, that is a necessary condition for determination the magnetic field direction [7,8]. Figs. 6,7 show field dependence of the off-diagonal voltage response, $V_{\text{out}}$ measured using pulsed scheme as described elsewhere [7-9] in $\text{Co}_{67}\text{Fe}_{3.85}\text{Ni}_{1.45}\text{B}_{11.5}\text{Si}_{14.5}\text{Mo}_{1.7}$ ($\alpha_5 = 3 \times 10^{-7}$) microwire with different geometry: metallic nucleus diameter and total diameter with glass coating are 6.0/10.2, 7.0/11.0 and 8.2/13.7 µm. Such considerations regarding dependence of the GMI response on the samples geometry should be taken into account for development of extremely thin microwires. It is important for microminiaturized sensor application since the demagnetizing factor starts to affect the domain structure and magnetic properties when geometric ratio of microwire diameter to its longitude becomes lower [10]. It should be assumed that the internal stresses relaxation after heat treatment should drastically change both the soft magnetic behavior and $\Delta Z/Z(H)$ dependence mostly because of the stress dependence of the magnetostriction described in ref [1]:
\[ \lambda_{s}(\sigma) = \lambda_{s}(0) - \Lambda\sigma \]  

where \( \lambda_{s}(0) \) is the saturation magnetostriction constant without applied stresses and \( \Lambda \) is the positive coefficient of the order of \( 10^{-10} \) MPa.

It should be noted from Fig. 6 that the \( V_{out}(H) \) curves have asymmetrical shape exhibiting close to linear growth within the field range from \(-H_m\) to \(H_m\). The \( H_m \) limits the working range of MI sensor to 240 A/m and should be associated with the anisotropy field.

Fig. 5. \( Z(H) \) dependence Co\(_{67}\)Fe\(_{3.85}\)Ni\(_{1.45}\)B\(_{11.5}\)Si\(_{14.5}\)Mo\(_{1.7}\) microwires with different geometry measured at frequencies \( f = 10, 200 \) and 500 MHz

It should be noted from Fig. 6 that the \( V_{out}(H) \) curves have asymmetrical shape exhibiting close to linear growth within the field range from \(-H_m\) to \(H_m\). The \( H_m \) limits the working range of MI sensor to 240 A/m and should be associated with the anisotropy field.

Fig. 6. \( V_{out}(H) \) response of Co\(_{67}\)Fe\(_{3.8}\)Ni\(_{1.4}\)Si\(_{14.5}\)B\(_{11.5}\)Mo\(_{1.7}\) microwires with different \( d \).

Fig. 7. \( V_{out}(H) \) of Joule-heated Co\(_{67}\)Fe\(_{3.85}\)Ni\(_{1.45}\)B\(_{11.5}\)Si\(_{14.5}\)Mo\(_{1.7}\) microwire annealed with 50 mA currents for different time.
The influence of Joule heating on off-diagonal field characteristic of nearly zero magnetostriction Co$_{67.1}$Fe$_{3.8}$Ni$_{1.4}$Si$_{14.5}$B$_{11.5}$Mo$_{1.7}$ microwire with diameters 9.4/17.0 µm is shown in Fig. 7. One can see that the thermal annealing with 50 mA DC current reduces the $H_m$ from 480 A/m in as-cast state to 240 A/m after 5 min annealing.

Recent applications and reduced dimensionality of developed microwires require extending frequency range for GMI studies and applications. This particularly induced us to extend frequency range for GMI studies. Fig.8 presents results on magnetic field dependence of real part, $Z_1$ of the longitudinal wire impedance $Z$ ($Z = Z_1 + iZ_2$) till 4 GHz. General features of these dependences is that the magnetic field of maximum shifts to the higher field region increasing the $f$.

2.4. GMI hysteresis

Another relevant feature is low field hysteresis observed even at high $f$ (Fig.8b). Recently we developed a model for magnetization reversal and MI field dependence for nearly-zero-magnetostrictive amorphous microwires with consideration that low-field GMI hysteresis arises from deviation of the anisotropy easy axis from transversal direction and explaining why the circular bias magnetic field $H_B$ produced by DC current $I_B$ running through the wire affects the hysteresis and asymmetry of the MI dependence. The validity of the model has been confirmed by the experiments [10]. Thus it was demonstrated that the low-field hysteresis originates from deviation of easy axis from circular direction (helical magnetic anisotropy) and that the application of the bias field leads to suppressing of this hysteresis (see Fig.9, where effect of bias voltage on $S_{21}$ parameter, proportional to off-diagonal GMI component is shown). Using the developed model, the main characteristics of the studied ferromagnetic microwire such as anisotropy field $H_k$, angle between the anisotropy easy axis and the transversal direction were obtained. In fact in pulsed exciting scheme when the sharp pulses with pulse edge time about 5 ns are produced by passing square wave multi-vibrator pulses through the differentiating circuit, overall pulsed current contains a DC component that produces bias circular magnetic field [6,7]. In this way low field hysteresis can be surpassed selecting adequate pulse amplitude.
In fact low field hysteresis can be also used for data storage in wire MI element. Thus, independently on the initial state, after applying of a constant current pulse $I_B$ in one direction (right to left in Fig 10 a), the magnetic moments will orient along the easy axis in up direction (Fig. 10 b) which corresponds to the store logical ’1’ state. Then, submitting the wire to external magnetic field makes the magnetic moments rotate in close-to-circumferential direction which characterized by low impedance (Fig. 10c) that can be easily detected. To write logical ’0’ one need to pass current pulse $I_B$ in the opposite direction (left to right in d) that makes the magnetization orient along the easy axis in down direction (Fig 10e). In this case, the external magnetic field induces the magnetic moments rotation in close-to-longitudinal direction characterized by high impedance as shown in Fig. 10f. Similarly, to rotate the magnetic moments from equi-impedance state (with the same impedance value), a small static current can be applied instead of external magnetic field $H_E$.

3. Conclusions

Thin amorphous wires with enhanced magnetic softness and GMI effect can be produced by the Taylor-Ulitovski technique. Magnetic properties and GMI effect of such microwires can be tailored by an appropriate selection of the metallic nucleus diameter, glass-coating thickness and chemical composition of the metallic nucleus and even by the heat treatment under magnetic field or without it. There are a number of interesting effects, such as induction of the transversal anisotropy in Fe-rich microwires allowing creating extremely stress sensitive elements. The investigation of diagonal and off-diagonal MI tensor component with pulse excitation in 6—12 µm amorphous glass-coated microwires has shown the great potential of these materials for microminiaturized magnetic field sensor application. Their main advantages are high sensitivity low-hysteresis field dependence, low power consumption, and very simple sensor scheme are very promising for technological application. By varying the alloys composition and applying post fabrication processing it is possible to control the sensor's operating range. The off-
diagonal impedance component in microwires with nearly zero magnetostriction is anti-symmetrical with close to linear behavior within the working range. The Joule heating of such microwires tends to decreasing of magnetoelastic coupling and anisotropy field. Thermal treatment is an additional effective factor for tuning of MI dependence as well as the alloy composition and geometric parameters. Low field GMI hysteresis has been observed and explained in terms of helical magnetic anisotropy of microwires. Data storage in MI microwire element using low field hysteresis has been proposed.

ACKNOWLEDGMENTS

The work was supported by the EU ERA-NET program under Project DEV MAG MI WIRTEC “MANUNET-2007-Basque-3” and by the Basque Government under Saiotek 09 Mic Magn project. A. Zh. and V. Zh. wish to acknowledge the support of the Basque Government under Program of Mobility of the Investigating Personnel of the Department of Education, Universities and Investigation Grants MV-2009-2-21 and MV-2009-2-24.

[1] Jiles DC 2003 Acta Materialia 51 5907
[2] Zhukova V, Ipatov M and Zhukov A 2009 Sensors 9 9216
[3] Zhukov A and Zhukova V 2009 Magnetic properties and applications of ferromagnetic microwires with amorphous and nanocrystalline structure, (New York: Nova Science Publishers, ISBN: 978-1-60741-770-5)
[4] Antonov A S, Borisov V T, Borisov OV, Prokoshin A F and Usov N A 2000 J. Phys. D: Appl. Phys. 33 1161
[5] Zhukov A, Gonzalez J, Blanco J M, Prieto M J, Pina E and Vazquez M 2000 J. Appl. Phys. 87 1402
[6] Zhukova V, Chizhik A, Zhukov A, Torcunov A, Larin V and Gonzalez J 2002 IEEE Trans Magn 38 3090
[7] Sandacci S I, Makhnovskiy D P, Panina L V, Mohri K and Honkura Y 2004 IEEE Trans Magn., 35 3505
[8] Mohri K and Honkura Y 2007 Sensors letters, 5 267
[9] Zhukov A, Ipatov M, Gonzalez J, Blanco J M, Zhukova V 2009 J. Magn. Magn. Mater. 321 822
[10] Ipatov M, Zhukova V, Zhukov A, Gonzalez J, and Zvezdin A 2010 Phys. Rev. B 81 134421
[11] Velázquez J, Vazquez M and Zhukov A 1996 J Mater Res. 11 2499.
[12] Chiriac H and Ovari TA 1996 Progress in Material Science 40 333
[13] O’Handley R C, Hasegawa R, Ray R and Chou C-P 1976 Appl. Phys. Lett. 29 330
[14] Morita H, Fujimori H and Obi Y, 1980 J. Magn. Magn. Mater. 15-18 1359
[15] Zhukov A 2006 Adv Func Mat 16 675.