Electronic Surveillance and Security Applications of Magnetic Glass-coated Microwires

Valentina Zhukova, Mihail Ipatov, Paula Corte-León, Alvaro Gonzalez, Alfonso García- Gómez Dept Materials Physics, Dept. Applied Physics and EHU Quantum Center, Univ. Basque Country, UPV/EHU, 20018 San Sebastian, Spain e-mails: valentina.zhukova@ehu.es; mihail.ipatov@ehu.es; paula.corte@ehu.eus; alvaro.gonzalezv@ehu.eus; alfonso.garciag@ehu.eus

Abstract—Applications in security and electronic surveillance require a combination of excellent magnetic softness with good mechanical and anti-corrosive properties and low dimensionality. We overviewed the feasibility of using glasscoated microwires for electronic article surveillance and security applications, as well as different routes of tuning the magnetic properties of individual microwires or microwires arrays making them quite attractive for electronic article surveillance and security applications. We provide the routes for tuning the hysteresis loops non-linearity by the magnetostatic interaction between the microwires in the arrays of different types of amorphous microwires. The presence of neighboring microwire (either Fe or Co-based) significantly affects the hysteresis loop of the whole microwires array. In a microwires array containing magnetically bistable microwires. we observed splitting of the initially rectangular hysteresis loop with a number of Barkhausen jumps correlated with the number of magnetically bistable microwires. Essentially, nonlinear and irregular hysteresis loops have been observed in mixed arrays containing Fe and Co-rich microwires. The obtained non-linearity in hysteresis loops allowed to increase the harmonics and tune their magnetic field dependencies. On the other hand, several routes allowing to tune the switching field by either post-processing or modifying the magnetoelastic anisotropy have been reviewed. The observed unique combination of magnetic properties together with thin dimensions and excellent mechanical and anti-corrosive properties provide excellent perspectives for the use of glasscoated microwires for security and electronic surveillance applications.

Keywords- magnetic microwires; magnetic softness; magnetic bistability, magnetic tags.

I. INTRODUCTION

Soft magnetic materials are highly demanded by several industries, including (but not limited to) microelectronics, electrical engineering, car, aerospace and aircraft industries, medicine, magnetic refrigerators, home entertainment, energy harvesting and conversion, informatics, magnetic recording or security and electronic surveillance [1]-[2]. In Arcady Zhukov

Dept Materials Physics, Dept. Applied Physics and EHU Quantum Center, Univ. Basque Country, UPV/EHU, 20018 San Sebastian and Ikerbasque, Bulbao, Spain e-mail: arkadi.joukov@ehu.es

most cases, like the case of security and electronic surveillance, in addition to excellent magnetic softness, a combination of mechanical and anti-corrosive properties and low dimensionality is required [3].

Almost all department stores, supermarkets, airports, libraries, museums, etc. are provided with different types of security and anti-theft systems. The principle of Electronic Article Surveillance (EAS) systems operation is well established: articles are provided with tags that respond to electromagnetic fields generated by the gates at the store/supermarket/library exits [3]. The response is picked up by the antenna installed on the gate, switching on the alarm. It is estimated that hundreds of thousands of such EAS systems have been installed and millions of tags are produced daily. Considering the great number of tags, they must be small, robust enough and inexpensive. Additionally, the magnetic materials employed in tags must be magnetically soft enough. The magnetic softness of crystalline soft magnetic materials (Permalloy, Fe-Si) is affected by processing. Therefore, amorphous soft magnetic materials, prepared by rapid melt quenching are considered as among the most suitable materials for tags containing soft magnetic materials [3][4].

Indeed, as a rule, amorphous materials present excellent magnetic softness together with superior mechanical properties [3]-[6]. Abrupt deterioration of the mechanical properties (such as tensile yield) upon the devitrification of amorphous precursor is reported [6]. Additionally, the fabrication process of amorphous materials involving rapid melt quenching is fast and inexpensive [1]-[7]. Accordingly, amorphous soft magnetic materials are useful for the design of robust magnetic devices and magnetoelastic sensors [8]-[12].

As discussed elsewhere, soft magnetic materials with squared hysteresis loops and relatively low coercivities are the preferred candidates for the EAS systems using magnetic tags [3]. The rectangular hysteresis loops can be easily implemented in different families of amorphous magnetic wires [4]. Therefore, considerable attention has been paid to applications of amorphous wires for magnetic tags for different kinds of EAS systems [4].

The aforementioned squared hysteresis loops of magnetic wires are linked to the peculiar remagnetization process of magnetic wires running through a single and large Barkhausen jump [4] [14].

Glass-coated magnetic microwires prepared by the socalled Taylor-Ulitovsky technique present the widest metallic nucleus diameters range (from 200 nm up to 100 μm) [4][15][16]. In this way, the Taylor-Ulitovsky method is the unique technique allowing fabrication of nanowires by rapid melt quenching [15]. On the other hand, the preparation of amorphous magnetic wires with diameter of about 100 µm coated by glass has recently been reported [16]. The presence of a flexible, thin, bio-compatible and insulating glass coating allows to enhance the corrosive resistance and, therefore, makes these microwires suitable for novel applications including biomedicine, electronic article surveillance, non-destructive monitoring external stimuli (stresses, temperature) in smart composites or construction health monitoring through the microwire inclusions [17][18].

Accordingly, considering dimensionality and combination of physical properties (magnetic, mechanical, corrosive), amorphous soft magnetic microwires are potentially suitable materials for electronic article surveillance and security applications [4][19][20]. There are several original papers dealing with rather different (multibit or single-bit) security and EAS applications of magnetic microwires [19][20]. In this paper, we will provide an overview of the trends related to EAS and security applications of glass-coated magnetic microwires.

This paper is organized as follows. In Section 2, the experimental methods as well as the microwires characteristics analyzed in this paper are provided. Section 3 deals with results on the feasibility of using magnetic microwires for magnetic tags followed by an overview of tuning of hysteresis loop non-linearity by the magnetostatic interaction between microwires.

II. EXPERIMENTAL SYSTEM DETAILS

Generally, we analyzed two different types of magnetic amorphous microwires: i) amorphous microwires with high positive magnetostriction coefficients, λ s, (Fe-Si-B-C, Fe-Ni-Si-B-C or Fe-Ni-Si-B) and ii) amorphous microwires with vanishing λ s (Co-Fe-Ni-B-Si-Mo, Co-Fe-Ni-B-Si-Mo, Co-Fe-B-Si-Cr-Ni or Co-Fe-B-Si-C). We studied microwires with metallic nucleus diameters, *d*, ranging from 10 up to 100 µm prepared using the Taylor-Ulitovsky method described elsewhere [4][21]. The Taylor-Ulitovsky method allows preparation of thinnest metallic wires (with typical diameters of the order of 0.1 to 100 µm) covered by an insulating glass coating [5][21].

The amorphous structure of all the microwires has been proved by the X-ray Diffraction (XRD) method. Typically, the crystallization of amorphous microwires was observed at Tann ≥ 500 ° C [4].

The induction method has previously been used for the hysteresis loops measurements. The details of the experimental set-up are described elsewhere [22]. The hysteresis loops were represented as the magnetic field, H, dependence of the normalized magnetization, M/M_0 , being M - the magnetic moment at a given magnetic field, and M_0 the magnetic moment at the maximum magnetic field amplitude Hm. Such hysteresis loops are useful for comparison of the samples with different chemical compositions (and, hence, different saturation magnetization).

In several cases, the hysteresis loops were measured with a conventional Super-conducting Quantum Interference Device (SQUID).

III. EXPERIMENTAL RESULTS AND DISCUSSION

Magnetic tags applications require a non-linear hysteresis loop that contains the characteristic distribution of harmonic frequencies. It is believed that the steeper the magnetization reversal, the higher the harmonic content of the signal. Accordingly, perfectly rectangular hysteresis loops with low coercivity observed in Fe-rich microwires (Figure 1) are attractive for use as magnetic tags.

On the other hand, the non-linearity of the hysteresis loop of the magnetic microwires can be further improved using the magnetostatic interaction of microwires. Below, we will present several experimental results on magnetic response of two kinds of individual microwires $(Co_{67}Fe_{3.9}Ni_{1.5}B_{11.5}Si_{14.5}M_{0.6}$ and $Fe_{74}B_{13}Si_{11}C_2)$ as well as the arrays containing either microwires of the same type or arrays containing two different kinds of microwires.

The hysteresis loops of such microwires are rather different: $Fe_{74}B_{13}Si_{11}C_2$ microwire with high and positive magnetostriction coefficient, λ s, exhibits perfectly rectangular hysteresis loops with Hc \approx 100 A/m (Figure 1a), while and inclined hysteresis loop with quite low Hc (Hc \approx 5 A/m) is observed in $Co_{67}Fe_{3.9}Ni_{1.5}B_{11.5}Si_{14.5}M_{0.6}$ microwire (see Figure 2b).

The hysteresis loop of an array containing two $Fe_{74}B_{13}Si_{11}C_2$ microwires is rather different from that of a single $Fe_{74}B_{13}Si_{11}C_2$ microwire. Two Barkhausen jumps can be observed at magnetic field amplitude, $H_0>80$ A/m (see Figure 3a). Such peculiar hysteresis loop shape has been explained considering the magnetostatic interaction in the two-microwire array [4]. Such magnetostatic interaction is a consequence of stray fields created by magnetically bistable microwires: the superposition of external and stray fields causes magnetization reversal in one of the samples, when the external field is below the switching field of a single microwire. A single rectangular hysteresis loop (similar to the case of single microwire shown in Figure 1) is observed for 60 A/m < $W_0 < 80$ A/m (see Figure 3b).



Figure 1. Hysteresis loops of as- prepared (a), and annealed at T_{ann} = 400 °C for 180 min (b) Fe₇₅B₉Si₁₂C₄ microwires.



Figure 2. Hysteresis loops of $Fe_{75}B_9Si_{12}C_4$ microwires with positive (a) and $Co_{67}Fe_{3.9}Ni_{1.5}B_{11.5}Si_{14.5}M_{0.6}$ with vanishing (b) magnetostriction coefficients.

Increasing the magnetic field amplitude (approximately at H0 > 250 A/m), this splitting of the hysteresis loop disappears (Figure 3b). Such dependence of the hysteresis loop of two microwires array can be understood from the counterbalance between the dH/dt and the switching time determined by the velocity of the DW propagation along the whole wire [4].

As discussed elsewhere [4], coercivity, H_c , is also affected by the frequency, *f*. Accordingly, Hc, as well as overall hysteresis loops of two microwires array, are affected by f in a similar way as by H_0 (see Figure 3b). For a two microwires array, two-steps hysteresis loops are observed for *f*< 150 Hz. At *f* > 150 Hz, the hysteresis loop splitting disappears, and at 150 < *f* <1000 Hz, a single smooth magnetization jump is observed.



Figure 3. Hysteresis loops measured at different magnetic field amplitudes H_0 (a) and at different magnetic field frequencies f (b) for as array with two Fe₇₅B₉Si₁₂C₄ microwires, dependences of odd harmonics (c) and even harmonics (d) on magnetic field amplitude in linear array of two Fe₇₄B₁₃Si₁₁C₂ microwires.

Accordingly, the odd and even harmonics of the signal of two Fe-rich microwires array are affected by H_0 and f (see Figure 3c,d).

A sharp increase in the harmonics amplitudes is observed when H_0 exceeds Hc (see Figures 3c,d). The even harmonics amplitudes are significantly inferior to the odd harmonics amplitudes. The field dependences of odd harmonics have a "plateau" between 60 and 90 A/m, which reflects the hysteresis loops splitting (see Figure 3a).

Another example of tuning the non-linearity of hysteresis loops and harmonics is the magnetostatic interaction of microwires with different character of hysteresis loops. Rather non-linear hysteresis loops can be obtained in an arrav consisting of one Co₆₇Fe_{3.9}Ni_{1.5}B_{11.5}Si_{14.5}M_{0.6} one Fe₇₄B₁₃Si₁₁C₂ and microwires (see Figure 4a). In such array, at $H_0 < 90$ A/m (which corresponds to Hc of Fe₇₄B₁₃Si₁₁C₂ microwire) the hysteresis loops character is typical of those for a single Co67Fe3.9Ni1.5B11.5Si14.5M0.6 microwire. Essentially, nonlinear hysteresis loops have been observed at H₀ >110 A/m (Figure 4a). Such peculiar hysteresis loops can be interpreted as the superposition of two hysteresis loops: one from magnetically bistable Fe₇₄B₁₃Si₁₁C₂ microwire (shown Figure 2a) and in the other one from Co₆₇Fe_{3.9}Ni_{1.5}B_{11.5}Si_{14.5}M_{0.6} microwire with linear hysteresis loop (shown in Figure 2b).

The peculiar hysteresis loop character at $H_0 \leq 120$ A/m can be explained by the partial magnetization reversal of the magnetically bistable wire under the influence of the stray field from the Co-based wire. The stray field is affected by the sample demagnetizing factor and the sample magnetization [23] [24]. In the case of Co-rich microwire the magnetization and hence, the stray field are affected by the applied magnetic field (as can be appreciated from the hysteresis loops shown in Figure 2b). In contrast, the magnetization of Fe-rich sample change by abrupt jump and below and above Hc is almost independent of the magnetic field (see Figure 2a).

Accordingly, such microwire array consisting of two microwires (Fe-rich and Co-rich) with different hysteresis loops presents odd and even harmonics quite different from the case of the array with two Fe-rich microwires (see Figures 4 b,c). A single sharp jump of odd and even harmonics is observed at $H_0 \approx Hc$. There is also a change in the odd and even harmonics in the weak (H_0 <Hc) field region (see Figures 4 b, c).

Thus, the use of arrays consisting of magnetic microwires allows us to create a complex and unique spectrum of magnetic harmonics in magnetic microwires.

Essentially, non-linear and irregular hysteresis loops have been observed in mixed arrays containing Fe and Corich microwires. The observed non-linear hysteresis loops allowed to increase the harmonics and to tune their magnetic field dependencies.

The aforementioned examples provide the routes for optimization of the response of magnetic microwires by



Figure 4. (a) Hysteresis loops of the $Fe_{74}B_{13}Si_{11}C_2 + Co_{67}Fe_{3.9}Ni_{1.5}B_{11.5}Si_{14.5}M_{0.6}$ array; (b) dependences of odd harmonics on magnetic field amplitude and (c) dependences of even harmonics on magnetic field amplitude. Reprinted with permission from ref. (4).

tuning the non-linearity of the hysteresis loops through the magnetostatic interaction. Such magnetic microwires can easily be incorporated into magnetic tags capable to respond to magnetic fields generated by the gates at the store/supermarket/library exits.

IV. CONCLUSIONS

In this paper, we showed that the presence of a neighbouring microwire (either Fe- or Co-based) significantly affects the hysteresis loop of the whole microwires array. In a microwires array containing magnetically bistable microwires, we observed splitting of the initially rectangular hysteresis loop with a number of Barkhausen jumps correlated with the number of magnetically bistable microwires. Essentially, non-linear and irregular hysteresis loops have been observed in mixed

arrays containing Fe and Co-rich microwires. The observed non-linear hysteresis loops allowed to increase the harmonics and to tune their magnetic field dependencies.

The observed unique combination of magnetic properties, together with thin dimensions and excellent mechanical and anti-corrosive properties, provide excellent perspectives for the use of glass-coated microwires for security and electronic surveillance applications.

ACKNOWLEDGMENT

This work was supported by Spanish MCIU under PGC2018-099530-B-C31 (MCIU/AEI/FEDER, UE), by EU "INFINITE" (Horizon Europe Framework under Programme) project, by the Government of the Basque Country, under PUE_2021_1_0009 and Elkartek (MINERVA and ZE-KONP) projects, by the University of the Basque Country, under the scheme of "Ayuda a Grupos Consolidados" (Ref.: GIU18/192) and under the COLAB20/15 project and by the Diputación Foral de Gipuzkoa in the frame of Program "Red guipuzcoana de Ciencia, Tecnología e Innovación 2021" under 2021-CIEN-000007-01 project. The authors thank for technical and human support provided by SGIker of UPV/EHU (Medidas Magnéticas Gipuzkoa) and European funding (ERDF and ESF). We wish to thank the administration of the University of the Basque Country, which not only provides very limited funding, but even expropriates the resources received by the research group from private companies for the research activities of the group. Such interference helps keep us on our toes.

REFERENCES

[1] P. Corte-Leon et al., "Magnetic Microwires with Unique Combination of Magnetic Properties Suitable for Various Magnetic Sensor Applications", Sensors, vol. 20, p. 7203, 2020.

[2] M. Vázquez, J. M. García-Beneytez, J. M. García, J. P. Sinnecker, and A. Zhukov, "Giant magneto-impedance, vol. 88, pp. 6501-6505, 2000.

[3] G. Herzer, "Magnetic materials for electronic article surveillance", J. Magn. Magn. Mater., Vol. 254–255, pp. 598–602, 2003.

[4] V. Zhukova et al., "Electronic Surveillance and Security Applications of Magnetic Microwires", Chemosensors, Vol. 9, p.100, 2021.

[5] T. Goto, M. Nagano, and N. Wehara, "Mechanical properties of amorphous $Fe_{80}P_{16}C_3B_1$ filament produced by glass-coated melt spinning", Trans. JIM, vol. 18, pp. 759–764, 1977.

[6] V. Zhukova et al., "Correlation between magnetic and mechanical properties of devitrified glass-coated Fe71.8Cu1Nb3.1Si15B9.1 microwires", J. Magn. Magn. Mater., vol. 249, pp. 79–84, 2002.

[7] A. Zhukov et al., Giant magnetoimpedance in rapidly quenched materials", J. Alloys Compound., vol. 814, pp. 152225, 2020.

[8] K. Mohri, T. Uchiyama, L. P. Shen, C. M. Cai, and L. V. Panina, "Amorphous wire and CMOS IC-based sensitive micromagnetic sensors (MI sensor and SI sensor) for intelligent measurements and controls", J. Magn. Magn. Mater., vol. 249, pp. 351-356, 2001.

[9] T. Uchiyama, K. Mohri, and Sh. Nakayama, "Measurement of Spontaneous Oscillatory Magnetic Field of Guinea-Pig Smooth Muscle Preparation Using Pico-Tesla Resolution Amorphous Wire Magneto-Impedance Sensor", IEEE Trans. Magn., vol. 47, pp. 3070-3073, 2011.

[10] Y. Honkura, "Development of amorphous wire type MI sensors for automobile use", J. Magn. Magn. Mater., vol. 249, pp. 375-381, 2002.

[11] A. Zhukov et al., Magnetoelastic sensor of level of the liquid based on magnetoelastic properties of Co-rich microwires, Sens. Actuat. A Phys., vol. 81(1-3) pp.129-133, 2000.

[12] V. Zhukova et al., "Development of Magnetically Soft Amorphous Microwires for Technological Applications", Chemosensors, vol. 10, p. 26, 2022

[13] L. Ding, S. Saez, C. Dolabdjian, L. G. C. Melo, A. Yelon, and D. Ménard, "Development of a high sensitivity GiantMagneto-Impedance magnetometer: comparison with a commercial Flux-Gate", IEEE Sensors, vol. 9 (2), pp. 159-168, 2009.

[14] K. Mohri, F. B. Humphrey, K. Kawashima, K. Kimura, and M. Muzutani, "Large Barkhausen and Matteucci Effects in FeCoSiB, FeCrSiB, and FeNiSiB Amorphous Wires", IEEE Trans. Magn., vol. 26, pp. 1789–1781, 1990.

[15] H. Chiriac, S. Corodeanu, M. Lostun, G. Ababei, and T.-A. Óvári, "Rapidly solidified amorphous nanowires", J. Appl. Phys., vol. 107, 09A301, 2010.

[16] P. Corte-Leon et al., "The effect of annealing on magnetic properties of "Thick" microwires", J. Alloys Compound., vol. 831, p.150992, 2020.

[17] D. Kozejova et al., "Biomedical applications of glass-coated microwires", J. Magn. Magn. Mater., vol. 470, pp. 2-5, 2019.

[18] A. Talaat et al., "Ferromagnetic glass-coated microwires with good heating properties for magnetic hyperthermia", Sci. Reports, vol. 6 p. 39300, 2016.

[19] D. Makhnovskiy, N. Fry, and A. Zhukov, "On different tag reader architectures for bistable microwires", Sens. Actuat. A Phys., vol. 166, pp. 133-140, 2011.

[20] S. Gudoshnikov, et.al., "Evaluation of use of magnetically bistable microwires for magnetic labels", Phys. Stat. Sol. (a), vol. 208, No. 3, pp. 526–529, 2011.

[21] L. Gonzalez-Legarreta et al., "Optimization of magnetic properties and GMI effect of Thin Co-rich Microwires for GMI Microsensors", Sensors, vol. 20, p.1558, 2020.

[22] A. Zhukov et al., "Advanced functional magnetic microwires for technological applications", J. Phys. D: Appl. Phys., vol. 55, p. 253003, 2022.

[23] V. Rodionova et al., "Design of magnetic properties of arrays of magnetostatically coupled glass-covered magnetic microwires" Phys. Stat. Sol. (a), vol. 207(8), pp. 1954–1959, 2010.

[24] A. Chizhik, A. Zhukov, J. M. Blanco, R. Szymczak, and J. Gonzalez, "Interaction between Fe-rich ferromagnetic glass coated microwires." J. Magn. Magn. Mater., vol. 249/1-2, pp. 99-103, 2002.