Magnetoimpedance effect in single-layered and sandwiched FeCuNbSiB thin films in frequencies up to 500 MHz

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***Abstract* — The magnetoimpedance (MI) effect was studied in single-layered ($Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$) and sandwiched ($Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7/Cu/Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$) thin films. Both single-layered and sandwiched structures were deposited onto glass substrates using the high-power impulse magnetron sputtering (HiPIMS) discharge operated with ultra-short pulses (3 µs). The MI measurements have been performed at room temperature, at frequencies up to 500 MHz, in longitudinal and transverse magnetic fields, before and after thermal / thermo-magnetic treatments. The largest MI ratio was obtained at 100 MHz, for the sandwiched structure, under the action of a longitudinal external magnetic field, after a thermo-magnetic treatment performed at 290°C.**

***Keywords* — Magnetoimpedance effect; Thin film; HiPIMS**

I. INTRODUCTION

SINCE its discovery, the magnetoimpedance (MI) effect has been a topic of intensive research due to its substantial potential applications in recording heads and magnetic field sensor elements with high sensitivity and quick response [1, 2]. From the practical point of view, the operating regime and potential applications of a magnetic sensor are determined especially by its field sensitivity [3]. MI materials have the ability to detect very small magnetic fields, whereas magneto-resistance materials generally require large fields to obtain a response of a few percent. The MI effect has been intensively studied in soft magnetic amorphous and nanocrystalline materials with near zero magnetostriction and different geometries: wires, microwires, ribbons, and single-layered, multilayered, and sandwiched thin films [4-9]. The ever-increasing demands for miniaturization encouraged the fabrication of MI materials in form of thin films. No less important is the fact that MI effect occurs with widely different magnitudes depending on the geometry, component materials and their layering.

The MI effect consists of significant changes in the complex electrical impedance of a ferromagnetic conductor as a response to an external static magnetic field, H_{dc}, when a high frequency alternating current (*ac*) flows through the conductor. If the changes in the complex electrical impedance are large, the effect is called giant magneto-impedance (GMI) effect. The origin of MI effect can be understood in terms of classical electromagnetism, being related, for a uniform single-layered film, to the skin depth, δ, which is a function of transverse permeability, $\mu(I_{ac}, f, H_{dc})$. The skin depth can be defined as [9]:

$$\delta = \frac{1}{\sqrt{\sigma \cdot f \cdot \mu(I_{ac}, f, H_{dc})}}, \tag{1}$$

where σ is the electrical conductivity and f is the drive current frequency.

According to Eq. (1), the transverse permeability can be controlled through the *ac* amplitude and frequency, as well as through the external magnetic field.

The skin effect represents the tendency of an alternating electric current to concentrate near the outer surface of a solid conductor. This effect becomes more and more significant as the frequency increases. As it can be seen from Eq. (1), large values of f and $\mu(I_{ac}, f, H_{dc})$ may lead to small skin depths. The MI effect is also very sensitive to all mechanism involved in the magnetization processes, as well as to the intrinsic and induced anisotropies of the investigated material. The MI ratio can be defined as [1]:

$$\frac{\Delta Z}{Z}(\%) = \frac{Z(H_{dc}) - Z(H_{dc\,\max})}{Z(H_{dc\,\max})} \times 100, \tag{2}$$

where $Z(H_{dc})$ and $Z(H_{dc\ max})$ are the impedances of the investigated sample corresponding to the magnetic field, H_{dc}, and its maximum value, $H_{dc\ max}$, which is the external saturation static magnetic field. In the present work, when $H_{dc\ max}$ value was not high enough to saturate the MI effect of the investigated samples, the MI ratio was calculated for $Z(H_{dc})$ and $Z(H_{dc=0})$.

Morikawa *et al.* [5] showed that it is possible to improve the MI performance without requiring the condition of a strong skin effect, but by using instead of thin films, multilayered structures as F/M/F (F – soft magnetic layer and M – a non-magnetic layer with high conductivity). In this case, due to its low resistance, the non-magnetic layer will carry the majority of the alternating current.

In nanocrystalline state, beside its high values of saturation magnetization and magnetic permeability, very low values of coercivity and high frequency losses, the FeCuNbSiB alloy is a great candidate for GMI effect [10]. Our group already

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published several studies on amorphous and nanocrystalline $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ thin films. We've studied the topological, structural, magnetic, magnetostrictive, mechanical, and tribological properties of this alloy in form of thin films, as well as the influence of certain deposition parameters (pulse duration, repetition frequency, average power, target-to-substrate distance, working gas pressure, substrate bias and nature) and post-deposition thermal treatments on its properties [11-13].

The high-power impulse magnetron sputtering (HiPIMS) is a relatively newly developed ionized physical vapor deposition technique, characterized by a dense plasma, high pulse power density at the sputtering target and high ionization degree of the sputtered material. Hence, this technique offers a great potential to deposit thin films with enhanced (topological, structural, mechanical) properties.

This paper presents the results of MI effect studies on FeCuNbSiB single-layered and FeCuNbSiB/Cu/FeCuNbSiB sandwiched thin films deposited using the HiPIMS technique. The studies have been performed before and after thermal and thermo-magnetic treatments, at frequencies up to 500 MHz. Some of the obtained samples have also been studied from the point of view of their surface chemical composition, structural and magnetic properties. The novelty and importance of this work are two-fold: first, besides our previous studies, it can be considered a significant contribution to a thorough description of FeCuNbSiB thin films properties and second, the technique used for the deposition of the investigated samples – from our best knowledge, these may be the first results of studies on MI effect in single-layered and sandwiched thin films obtained using the HiPIMS technique. As some of our previous studies reported, HiPIMS is a relatively new and promising technique, which can be used to deposit high-quality thin films of a wide range of materials [11-15].

II. EXPERIMENTAL DETAILS

The films were deposited onto 18×18 mm^2 glass substrates, using 10×18 mm^2 masks (Figure 1), under a base pressure of approximately 2×10^{-6} Torr, using a HiPIMS system equipped with two targets: one made of $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ ribbons (provided by Vacuumschmelze GmbH) and another one made of a high purity (99.995%) circular copper (Cu) target. The targets were sputtered in pure argon atmosphere, under a pressure of 10 mTorr and a constant gas flow rate of 20 sccm, using the following deposition parameters: -1 kV pulse discharge voltage, 4 µs pulse duration and average discharge power of 30 W. The substrate was grounded and the target-to-substrate distance was kept constant at 6 cm. After the deposition, the single-layered samples (thickness of 800 nm) were cut into 3 mm wide and 10 mm long pieces for MI measurements. In the case of sandwiched films, FeCuNbSiB (800 nm)/Cu (800 nm)/ FeCuNbSiB (800 nm), the FeCuNbSiB layers were identical to those of the single-layered samples, while the Cu layer was 1 mm wide and 14 mm long (Figure 1).

The surface composition of the $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ layers

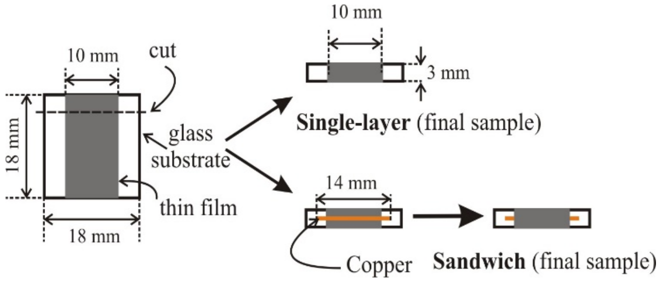

Fig. 1. Simplified schematic of the single-layered and sandwiched thin films obtaining process.

was studied by X-ray photoelectron spectroscopy (XPS), using a PHI 500 VersaProbe spectrometer with mono-chromatic Al K_α radiation (1486.7 eV). The thickness of the samples was estimated based on the deposition rate which was measured by a quartz crystal microbalance. The obtained results showed that the samples have a reasonably uniform thickness (800±20 nm), which will ensure reliable MI measurements and facile interpretation.

After the deposition process, the single-layered samples were thermally treated for 60 min., in furnace, under high vacuum (10^{-6} Torr), at temperatures between 200 and 525°C (no magnetic field applied). The sandwiched samples were subjected to a thermo-magnetic treatment (magnetic field of 500 Oe, applied along the short direction of the samples, in their plane), for 30 min., performed at 290°C. The choice of an annealing temperature lower than in the case of single-layered samples is due to the fact that, at high temperatures, Cu may diffuse into the surrounding magnetic layers. To see how the annealing process influences the magnetic behaviour of the samples, room temperature in-plane hysteresis loops were recorded before and after each thermal treatment, using a differential inductive method at 50 Hz and an integrating fluxmeter. The structure of both types of as-deposited and thermally treated samples was analyzed by X-ray diffraction, using a SHIMADZU LabX XRD-6000 diffractometer (CuK_α radiation, Bragg-Bretano configuration).

The MI measurements were performed at room temperature, using an Agilent E4991A impedance analyzer, in the frequency range from 1 to 500 MHz, varying the *ac* amplitude in the range from 0.1 to 10 mA. These values are low enough to avoid Joule heating of the samples. A pair of Helmholtz coils was used to provide the *dc* magnetic field in the range from -70 to 70 Oe. The MI effect was studied in two configurations: longitudinal – LMI (H_{dc} was applied along the *ac* direction) and transverse – TMI (H_{dc} was applied perpendicular to the *ac* direction, in film's plane).

III. RESULTS AND DISCUSSION

According to the XRD results, in as-deposited state, both single-layered and sandwiched samples are amorphous. The XPS analysis results showed that the composition of the FeCuNbSiB layers is very close to the target one.

The MI measurements results indicated that, even if the *ac* amplitude, selected in the range of previously mentioned values, has no significant influence on the MI response, a maximum value was obtained when the probe current amplitude was set at 2 mA.

Figure 2 presents room temperature in-plane hysteresis loops for as-deposited single-layered $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$

thin film, measured by applying the magnetic field along (a) and perpendicular (b) to its short direction, in film's plane. The sample presents in-plane uniaxial anisotropy, with the easy axis (EA) along the transverse direction. For the as-deposited single-layered samples, the coercive magnetic field, H_c, value is about 1.9 Oe, with no significant changes for the easy or hard axis (HA) directions.

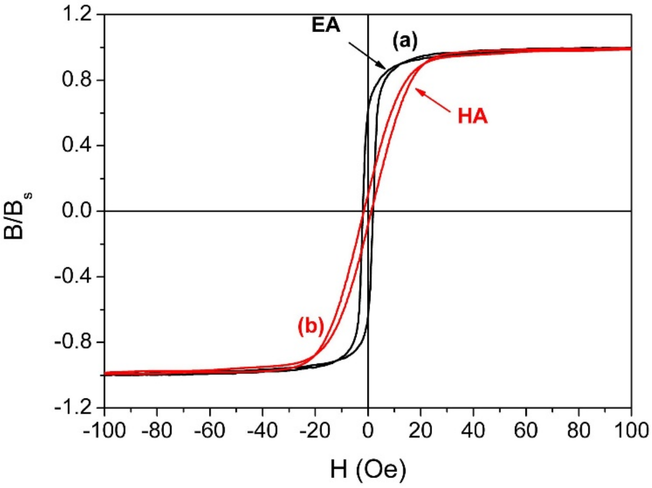

Fig. 2. Room temperature in-plane hysteresis loops for as-deposited single-layered $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ thin film: (a) along and (b) perpendicular to its short direction, in film's plane.

Figure 3 shows the coercive magnetic field and average grains size dependence on the annealing temperature (200–525°C) for single-layered $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ thin films. The XRD results revealed that after the thermal treatment performed at 450°C, the films structure consists of randomly oriented crystalline α-Fe(Si) grains (approximately 11 nm in size), embedded in a residual amorphous matrix. The nanocrystals size was estimated from the (110) diffraction peak present in the recorded diffractograms, using the Debye-Scherrer formula. According to the obtained results, increasing the annealing temperature from 450 to 525°C leads to an increase in average grains size from 11 to 18 nm. The study of the H_c dependence on the annealing temperature showed that 475°C is an optimum annealing temperature in that it leads to a minimum H_c value (0.5 Oe), about 4 times lower than the value of the as-deposited state.

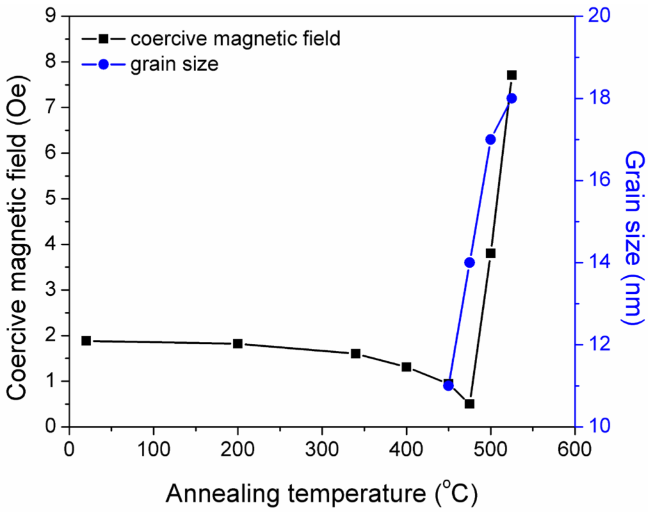

Fig. 3. Annealing-temperature dependence of coercive magnetic field and average grain size for single-layered $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ thin films.

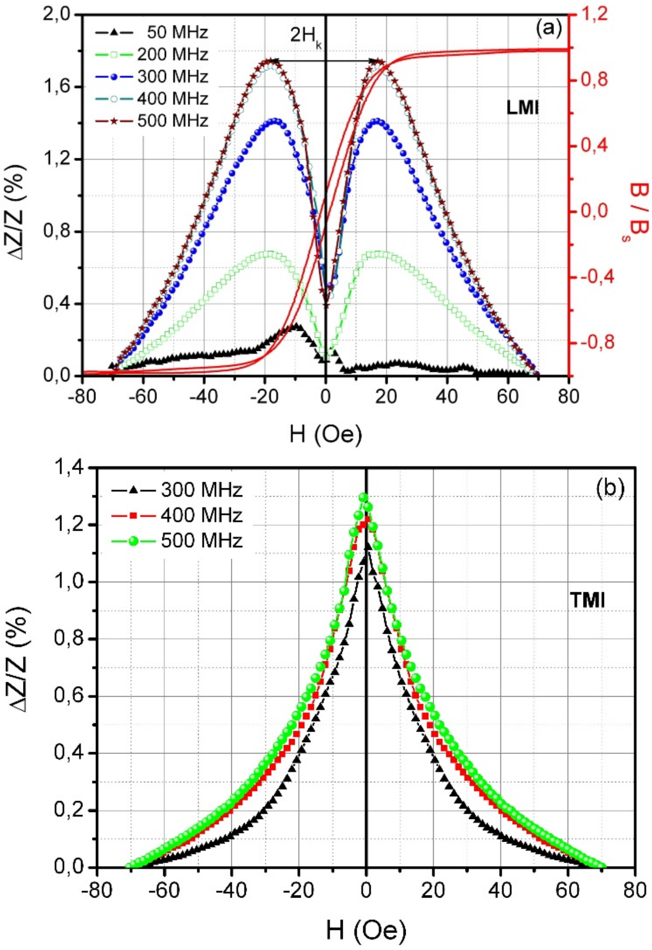

Fig. 4. Dependence of MI ratio on *ac* frequency an H_{dc} for $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ single-layered thin films in as-deposited state: (a) longitudinally and (b) transversely applied magnetic field.

Figure 4(a) and (b) and Figure 5(a) and (b) present ΔZ/Z *vs.* *H* LMI (Fig. 4(a) and Fig. 5(a)) and TMI (Fig. 4(b) and Fig. 5(b)) curves, at selected frequencies, for a $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ single-layered thin film in as-deposited state (Fig. 4(a) and (b)) and after annealing, for 60 min., at the optimum temperature of 475°C (Fig. 5(a) and (b)), respectively. In Fig. 4(a) and 5(a), the corresponding HA normalized hysteresis loops were over-laid.

A general observation is the difference between the shape of the LMI and TMI curves. For all the samples, before and after thermal treatment, the LMI curves present two peaks of nearly the same intensity, symmetric located with respect to the external magnetic field. Instead, the TMI curves present a single peak located at 0 Oe. These differences in shape may be understood in terms of the measurements' geometry effect on the effective differential permeability [16]. Another difference is that for the LMI measurements, the MI effect occurs in a field range almost twice larger than for the TMI ones. In both configurations, the increasing-field curves (shown) and the decreasing-field curves (not shown) were essentially the same, indicating a non-hysteretic MI effect for the obtained $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ single-layered thin films.

As it can be observed, the LMI peaks location coincides with the effective anisotropy field, H_k, which can also be obtained from the corresponding hysteresis loops: ±18 Oe for the as-deposited samples and ±11 Oe for the thermally treated ones.

Therefore, after the heat treatment, due to both

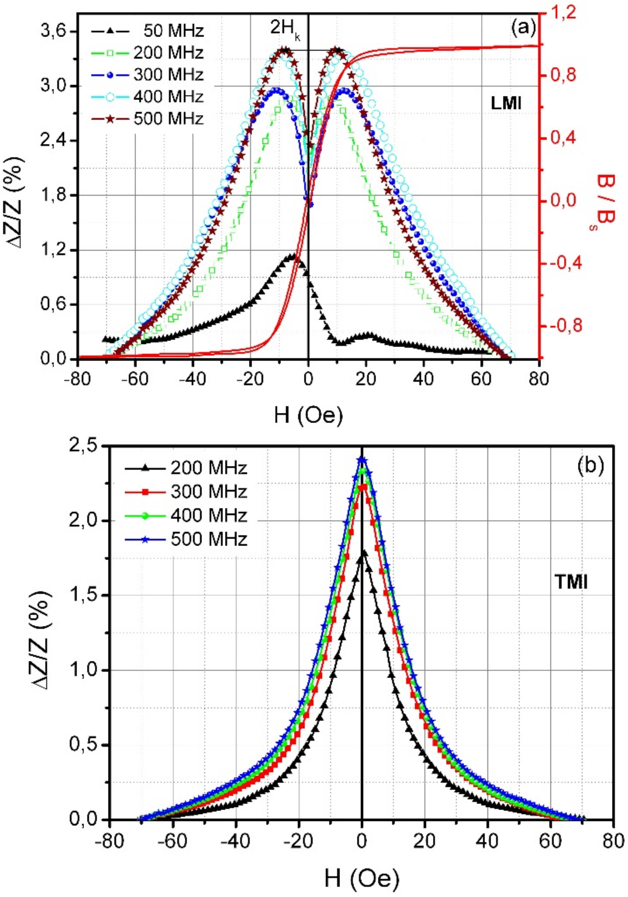

Fig. 5. Dependence of MI ratio on *ac* frequency an H_{dc} for thermally treated $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ single-layered thin films: (a) longitudinally and (b) transversely applied magnetic field.

relaxation of residual stress induced during the deposition process and structural changes in whole thin film's volume, H_k value decreases with approximately 40%. For both types of measurements, the thermal treatment doesn't change the geometry of the curves, leading to an enhancement of the MI ratios, especially in the low-field region, and to a slight narrowing of the peaks.

In both longitudinally and transversally magnetized cases, before and after the thermal treatment, the MI ratio increases with the *ac* frequency, the largest MI response being found when the *ac* frequency was set to 500 MHz. Based on the existing literature, it can be assumed that the MI ratio might increase for frequencies above 500 MHz, in the present case this value being limited by the measuring system.

The thermal treatment leads to an increase in the MI ratios, especially in the low-field region, where, for example, at 500 MHz, the LMI and TMI ratios almost doubled. Anyway, it's well known that, in many cases, soft magnetic amorphous single-layered films exhibit no measurable or weak MI effect. Suitable annealing processes release the mechanical stresses induced in the films during the deposition process, improve the soft magnetic properties and, consequently, the MI response.

Figure 6(a) and (b) and Figure 7(a) and (b) present ΔZ/Z *vs. H* LMI (Fig. 6(a) and Fig. 7(a)) and TMI (Fig. 6(b) and Fig. 7(b)) curves for a $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7/Cu/Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ sandwiched film in as-deposited state (Fig. 6(a) and (b)) and after a thermo-magnetic treatment, performed for 30 min., at 290°C (Fig. 7(a) and (b)).

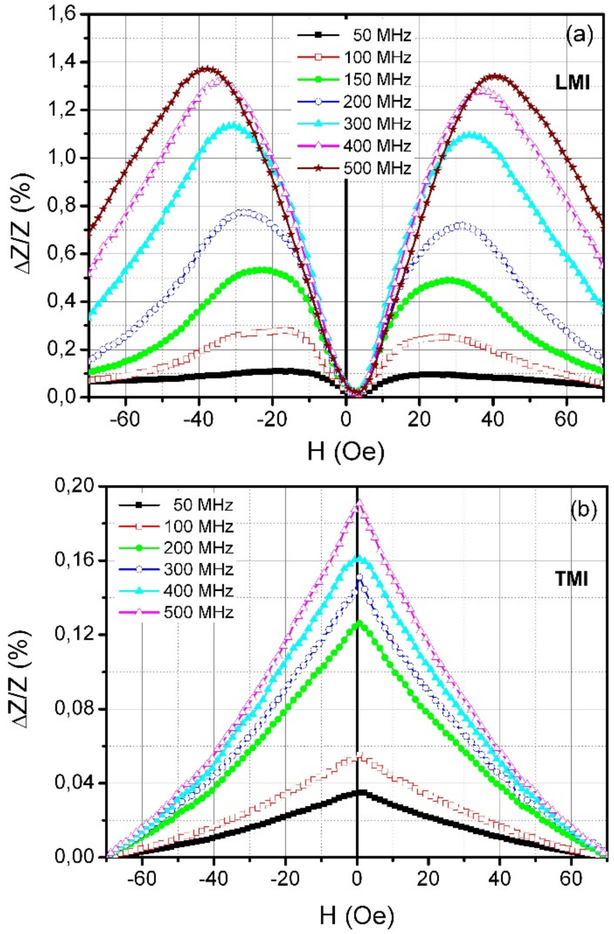

Fig. 6. The dependence of MI ratio on *ac* frequency an H_{dc} for a $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7/Cu/Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ sandwiched film (as-deposited state): (a) longitudinally and (b) transversely applied magnetic field.

In the case of as-deposited sandwiched films, for both LMI and TMI configurations, the MI response increases with the *ac* frequency: in the low-field region, when the frequency value increases from 50 to 500 MHz, the LMI ratio value increases about 14 times, while the TMI ratio value increases about 6 times.

After the thermo-magnetic treatment, in both LMI and TMI configurations, the MI ratio value increases in the frequencies range from 30 to 100 MHz, reaches its maximum value at 100 MHz and decreases in the frequencies range from 100 to 500 MHz. It's obvious that, compared with the single-layered films, the sandwiched samples exhibit their maximum MI effect at lower frequencies.

The obtained results showed that, after the thermo-magnetic treatment, in the low-field region, for the MI measurements performed for an *ac* frequency value of 100 MHz, the LMI ratio value increases about 60 times, while the TMI ratio value increases about 13 times with respect to the as-deposited state.

IV. SUMMARY AND CONCLUSION

The magneto-impedance effect was studied in FeCuNbSiB single-layered and FeCuNbSiB/Cu/FeCuNbSiB sandwiched thin films deposited using the HiPIMS technique. In almost all cases, the LMI curves presented two peaks of nearly the same intensity, symmetric located with respect to the external magnetic field, while the TMI curves presented a single peak. The TMI effect was found to be weaker than the

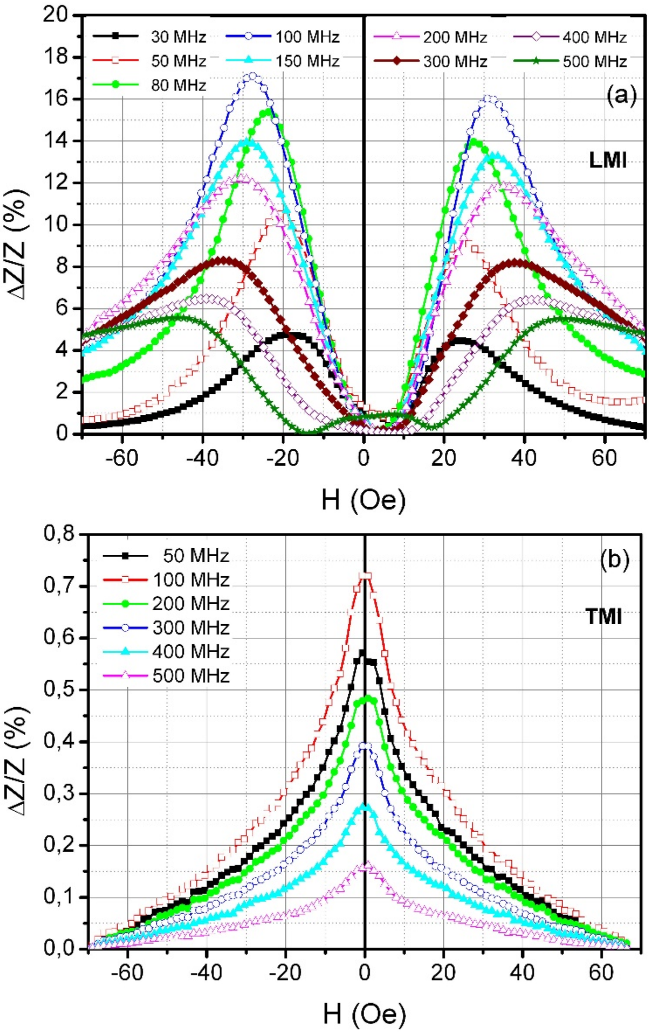

Fig. 7. The dependence of MI ratio on *ac* frequency an H_{dc} for a $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7/Cu/Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ sandwiched film after a thermo-magnetic treatment (30 min., 290ºC): (a) longitudinally and (b) transversely applied magnetic field.

LMI one, regardless the layering or the microstructure of the investigated samples.

In as-deposited state, for all the obtained samples, in both TMI and LMI configurations, the MI ratio increases with increasing the alternating current frequency. After the annealing process, the MI response keeps its increasing trend only for the FeCuNbSiB single-layered films. In the case of sandwiched films, the largest MI response was found when the current frequency was set at 100 MHz. As compared with the as-deposited state, this value is approximately 60 times higher for the LMI configuration and 13 times higher for the TMI one, respectively.

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References

[1] C. Tannous, J. Gieraltowski, "Giant magneto-impedance and its applications", *Journal of Materials Science: Materials in Electronics*, 15, 2004, pp. 125-133, DOI:10.1023/B:JMSE.0000011350.93694.91;

[2] M. H. Phan, Hua-Xin Peng, "Giant Magnetoimpedance Materials: Fundamentals and Applications", *Progress in Materials Science*, 53, 2008, pp. 323-420, DOI:10.1016/j.pmatsci.2007.05.003.

[3] A. Chaturvedi, N. Laurita, A. Leary, M.-H. Phan, M. E. McHenry, H. Srikanth, "Giant magnetoimpedance and field sensitivity in amorphous and nanocrystalline (Co1−xFex)89Zr7B4 (x = 0, 0.025, 0.05, 0.1) ribbons", *Journal of Applied Physics*, 109, 2011, pp. 07B508(3), DOI: 10.1063/1.3540403;

[4] L. V. Panina, K. Mohri, "Magneto-impedance Effect in Amorphous Wires", *Applied Physics Letters*, 65, 1994, pp. 1189-1191, DOI: 10.1063/1.112104;

[5] T. Morikawa, Y. Nishibe, H. Yamadera, Y. Nonomura, M. Takeuchi, Y. Taga, "Giant magneto-impedance effect in layered thin films", *IEEE Trans. Magn.*, 33, 1997, pp. 4367-4372, DOI: 10.1109/20.620448;

[6] L.V. Panina and K. Mohri, "Magneto-impedance in multilayer films", *Sens. Actuators*, 81, 2000, pp. 71-77, DOI:10.1016/S0924-4247(99)00089-8;

[7] S. Xiao, Y. Liu, Y. Dai, L. Zhang, "Giant magnetoimpedance effect in sandwiched films", *Journal of Applied Physics*, 85, 1999, pp. 4127-4130, DOI:10.1063/1.370321;

[8] Y. Han, X. Li, W.X. Lv, W.H. Xie, Q. Zhao, Z.J. Zhao, "Magnetoimpedance effect of FINEMET ribbons coated with Fe20Ni80 permalloy film", Journal of Alloys and Compounds, 678, 2016, pp. 494-498, DOI:10.1016/j.jallcom.2016.04.021;

[9] M. A. Corrêa, A. D. C. Viegas, R. B. da Silva, A. M. H. de Andrade, R. L. Sommer, "Magnetoimpedance of Single and Multilayered FeCuNbSiB Films in Frequencies up to 1.8 GHz", *Journal of Applied Physics*, 101, 2007, pp. 043905, DOI:10.1063/1.2512867;

[10] M.E. McHenry, D.E. Laughlin, "Amorphous and Nanocrystalline Materials for Applications as Soft Magnets", *Progress in Materials Science*, 44, 1999, pp. 291-433, DOI:10.1016/S0079-6425(99)00002-X;

[11] I. L. Velicu, M. Neagu, H. Chiriac, V. Tiron, M. Dobromir, "Structural and Magnetic Properties of FeCuNbSiB Thin Films Deposited by HiPIMS", *IEEE Transactions on Magnetics*, 48, 2012, pp. 1336-1339, DOI:10.1109/TMAG.2011.2173561;

[12] I. L. Velicu, M. Kowalczyk, M. Neagu, V. Tiron, H. Chiriac, J. Ferenc, "FINEMET-type thin films deposited by HiPIMS: influence of growth and annealing conditions on the magnetic behaviour", *Materials Science & Engineering B*, 178, 2013, pp. 1329-1333, DOI:10.1016/j.mseb.2013.03.002;

[13] I. L. Velicu, Maria Neagu, Lucian Costinescu, Vasile Tiron, Daniel Munteanu, "Nanomechanical characterization of amorphous and nanocrystalline FeCuNbSiB thin films", *Applied Surface Science*, 352, 2015, pp. 5-9, DOI:10.1016/j.apsusc.2014.12.143;

[14] I.-L. Velicu, V. Tiron, B.-G. Rusu, G. Popa, "Copper thin films deposited under different power delivery modes and magnetron configurations: A comparative study", *Surface and Coatings Technology*, 2016, in press, DOI:10.1016/j.surfcoat.2016.11.001;

[15] I.-L. Velicu, V. Tiron, C. Porosnicu, I. Burducea, N. Lupu, G. Stoian, G. Popa, D. Munteanu, "Enhanced properties of tungsten thin films deposited with a novel HiPIMS approach", *Applied Surface Science*, 2016, in press, DOI:10.1016/j.apsusc.2017.01.067;

[16] R. L. Sommer, C. L. Chien, "Longitudinal and Transverse Magneto-Impedance in Amorphous $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ Films", *Appl. Phys. Lett.*, 67, 1995, pp. 3346-3348, DOI:10.1063/1.115242.