Correlation Between the Magneto-Impedance and Ferromagnetic Resonance Responses in Nanocrystalline Microwires

Tibor-Adrian Óvári, Horia Chiriac, Manuel Vázquez, and Antonio Hernando

***Abstract*—Results on the correlation between ferromagnetic resonance and magneto-impedance responses in soft magnetic Fe-based microwires with nanocrystalline structure are reported for the first time. The evolution of the magneto-impedance effect with structural changes starting from the as-cast amorphous state up to nanocrystallization and further crystallization is interpreted through changes induced in the surface magnetic anisotropy and magnetization process of such wires, that are studied by means of ferromagnetic resonance.**

***Index Terms*—Ferromagnetic resonance, magneto-impedance effect, microwires, nanocrystalline materials.**

I. Introduction

THE MAGNETO-IMPEDANCE (MI) effect consists of the variation of the high frequency impedance of a soft magnetic conductor subjected to a small dc magnetic field due to the change in the skin depth originating in the change in magnetic permeability with the applied field [1]. The MI effect has been widely studied in the last few years, and found to be suitable for sensor applications. The magnitude of the MI effect at frequencies of the order of MHz depends on the sample's surface magnetic anisotropy and magnetization process. At frequencies of the order of GHz, the origin of the MI effect is connected to the ferromagnetic resonance phenomenon (FMR) [2]. We have recently reported that FMR is a sensitive method of investigation for the surface anisotropy and magnetization process of amorphous microwires [3]. Measurements of microwave losses of such materials also reflect accurately their magnetization processes [4].

The aim of this paper is to investigate the evolution of the MI effect in $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ glass-coated microwires starting from the as-cast amorphous state up to nanocrystallization and further crystallization, and to explain the results based on the changes induced by structural modifications in their surface magnetic anisotropy and magnetization process. These changes have been monitored by means of microwave absorption FMR measurements.

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II. Experiment

$Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ amorphous glass-coated microwires were prepared by glass-coated melt spinning. Samples with diameter of the metallic core of 32 μm and glass coating thickness of 4 μm, cut to 10 mm long pieces, have been used in the experiments. The amorphous state was checked by X-ray diffraction. Samples were annealed in vacuum for 1 h at temperatures up to 600°C, starting from the as-cast amorphous state in each case.

MI measurements were performed after each annealing stage, at frequencies of the ac driving current between 100 kHz and 10 MHz, using a digital oscilloscope coupled with a computer, that allowed automatic data acquisition and processing. The amplitude of the ac current was 5 mA. The impedance behavior as function of frequency and an axial dc field were investigated. The maximum value of the applied dc field, $H_{\max}$, was 1700 A/m, enough to roughly saturate the sample along the axial direction. In order to analyze the magnitude of the MI effect, we used the MI ratio (MIR):

$$\Delta Z/Z = 100 \times [Z(H) - Z(H_{\max})]/Z(H_{\max}) \qquad (1)$$

where $Z(H)$ is the impedance under a dc applied field H.

Microwave absorption FMR spectra were determined in parallel configuration—dc magnetic field parallel to the wire axis—with an X band spectrometer. The frequency of the microwave field was 10.5 GHz, and its power was 200 μW. Due to the high frequency, the FMR response is given by the microwires' surface region, which is also of interest from the point of view of the MI effect. The derivatives of the resonance curves were obtained by the dc magnetic field modulation technique. The dc magnetic field was modulated with an alternating field having a frequency of 1 kHz.

Circumferential permeability was calculated from circumferential magnetization curves obtained at 10 kHz by integrating the voltage induced across the sample ends due to flux variation determined by the circumferential magnetization through the longitudinal cross section.

III. Results and Discussion

Fig. 1 illustrates the frequency dependence of the maximum MIR, $(\Delta Z/Z)_{\max}$, with the annealing temperature as a parameter. One observes that the as-cast microwire displays a very

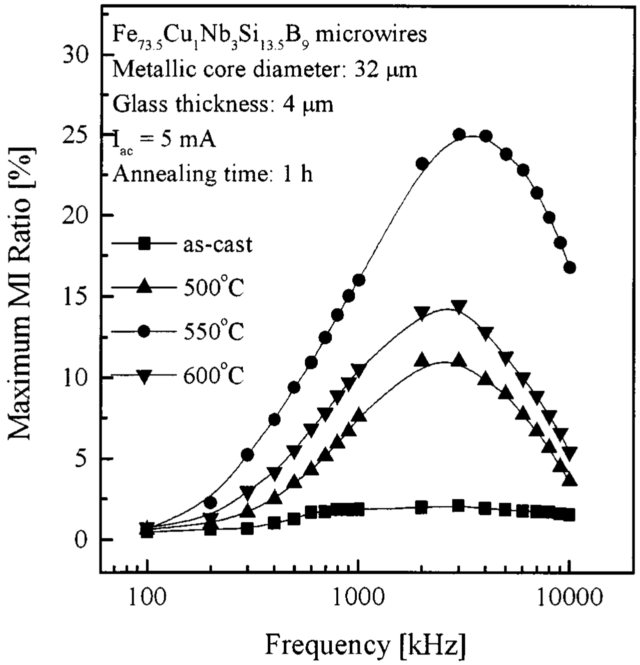

Fig. 1. Frequency dependence of the maximum MIR for as-cast and annealed $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ microwires.

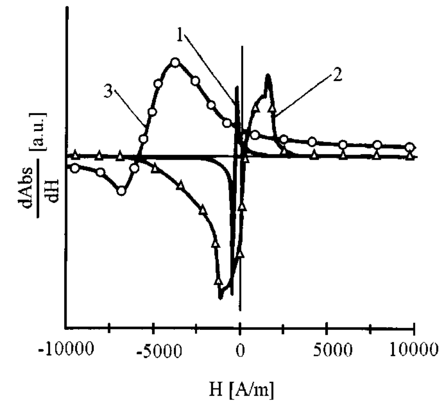

Fig. 2. Low field FMR peaks of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ microwires: as-cast (curve 1), annealed at 550°C (curve 2—triangles), and at 600°C (curve 3—circles).

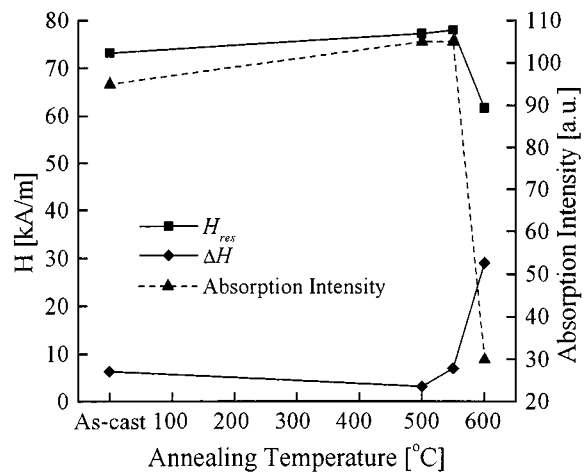

Fig. 3. Dependence of the resonance field, H_{res}, line breadth, ΔH, and absorption intensity, on the annealing temperature for $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ microwires.

small MI effect, with the maximum MIR of ~2%. Annealing for 1 h at 500°C leads to an increase of the MI response, the maximum MIR reaching to 11%. Nevertheless, the largest MI effect is obtained after annealing at 550°C, when the maximum MIR is larger than 25%. Annealing over this temperature leads to a decrease of the MI response, i.e. the maximum MIR for a sample annealed at 600°C decreases below 15%.

The composition of these microwires is one of the most suitable for the achievement of a soft magnetic nanocrystalline structure after appropriate annealing—1 h at around 550°C [5]. This magnetically soft structure is responsible for the largest MI response displayed in Fig. 1. However, structural relaxation within the amorphous structure occurs for annealing below this temperature, accounting for the moderate enhancements of MI and magnetic softening. Finally, important structural changes (namely, crystallization) that result in the corresponding magnetic hardening, take place especially when samples are annealed at temperatures over 550°C.

The MI response is altered by structural relaxation and changes through corresponding modifications induced in the surface magnetic anisotropy and magnetization process. To obtain a large and sensitive MI effect in this frequency range, either an appropriate domain structure (i.e., circumferential in this case) or soft magnetic behavior in the surface region of the sample are required.

The derivative resonance spectra of microwires display besides the main resonance peak, a smaller low field peak, whose origin has been related to the low field surface magnetization process [3].

Fig. 2 shows the low field peaks of the FMR spectra for an as-cast amorphous microwire (curve 1), and after annealing at 550°C (curve 2), and at 600°C (curve 3). It was shown, by correlation with the low field hysteresis loops, that these peaks are determined by the low field magnetization process from the surface region of the microwire, and the analysis of their shapes allows us to obtain information about the magnetization process mechanisms, whether they are mainly rotations or domain wall movements [3]. Therefore, they are related to the MI characteristics, since the latter strongly depends on the type of the preponderant magnetization process (domain wall motion or magnetization rotation).

The shape of the low field peak for the as-cast microwire is very sharp, fact consistent with the axial surface magnetization process achieved by a single and large Barkhausen jump (LBJ). The low field peak is detected at the same value of the applied field at which the LBJ occurs for the axial magnetization process of the whole sample (30 A/m). The LBJ appears due to the favorable domain structure, i.e., single domain with longitudinal easy axis of magnetization, typical for long amorphous microwires with positive magnetostriction and low demagnetizing field, as the $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ as-cast one $(\lambda_S = 25 \times 10^{-6})$ [3], [6].

Fig. 3 illustrates the dependence of the resonance field, H_{res}, absorption intensity, and line breadth, ΔH, of the main resonance peak, on the annealing temperature.

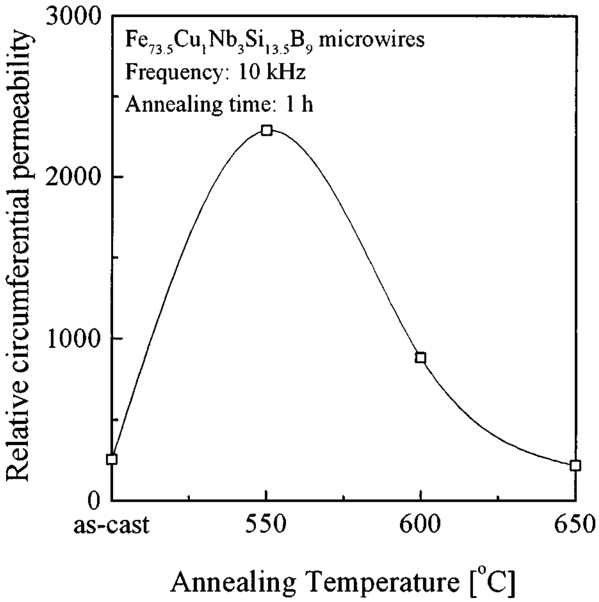

Fig. 4. Dependence of the circumferential relative permeability measured at 10 kHz on the annealing temperature for FeCuNbSiB microwires.

The uniaxial anisotropy of the as-cast sample is of magnetoelastic origin and results from the coupling between positive magnetostriction and axial tensile stresses. The surface region also exhibits longitudinal easy axis [6], and it does not display soft magnetic properties on the circumferential direction. In fact, the axial anisotropy is large as it results from the value of H_{res}, and the circumferential permeability is expected to be small. Thus, one cannot expect a sensitive MI effect in this case. The dependence of the circumferential permeability on annealing temperature, shown in Fig. 4, confirms this explanation, although the results are obtained at 10 kHz.

The sample annealed at 550°C, corresponding to the nanocrystalline phase formation, displays a wider low field peak, whose shape indicates the superposition of two peaks originating in the response of two regions with different magnetic characteristics. In this case, the low field peak appears at an applied field of 500 A/m, that is larger than the coercivity of the axial hysteresis loop. The larger width of the peak shows that the surface magnetization process in this case is mainly achieved through magnetization rotations. The complex aspect of the peak originates in the presence of two distinct phases: the nanosized α-FeSi crystalline grains embedded in a residual amorphous matrix, each one having a different preponderant anisotropy (magnetocrystalline and magnetoelastic, respectively). H_{res} slightly increases for samples annealed at temperatures up to 550°C, while for samples annealed at temperatures over this value it decreases below the value that corresponds to the as-cast sample. The evolution of H_{res} for annealed samples shows a decrease of the axial anisotropy field with annealing temperature up to 550°C, associated with stress relief and structural relaxation. The largest value of H_{res} and consequently the smallest axial anisotropy field are obtained for the sample annealed at 550°C (78 kA/m), that corresponds to the nanocrystalline phase formation and thus to the improvement of soft magnetic properties. Accordingly, the surface region of the nanocrystalline sample displays soft magnetic properties even on the circumferential direction (see also Fig. 4) and the condition for having a large MI effect is accomplished.

The low field peak of the sample annealed at 600°C appears at a larger value of the applied field (6 kA/m), and it is very wide, showing a deterioration of the wire's soft magnetic properties determined by crystallization. In fact, the increasing size of the α-FeSi crystalline grains together with the appearance of FeB phases make the magnetocrystalline anisotropy become preponderant. Therefore, H_{res} decreases and ΔH increases. The surface magnetization process takes place through magnetization rotations, but a strong decrease of the circumferential permeability occurs (see Fig. 4). Hence, the MI response decreases, but not so drastically due to the decrease of resistivity with crystallization.

The absorption intensity displays a similar behavior like H_{res}—a small increment up to 550°C followed by an abrupt decrease for the sample annealed at 600°C. This quantity can be used to predict the MI response at frequencies of the order of GHz, since the resonance response is proportional to the real part of the surface impedance, and it has been demonstrated that for longitudinal MI effect the FMR and MI responses are identical [2]. As expected, at very high frequencies, the differences between the MI responses of the as-cast, annealed, nanocrystalline, and crystalline samples are moderate, although the largest effect is still displayed by the nanocrystalline microwire. In the crystallized sample the mobility of the magnetic moments is reduced, and the fulfillment of the resonance condition is more difficult, leading to a decrease of the absorption intensity and of the MI response at GHz frequencies.

IV. Conclusion

FMR allows, along with the identification of structural phases with different magnetic properties, an accurate explanation of the changes induced in the surface anisotropy and magnetization process of Fe-based microwires during the annealing steps performed to induce the nanocrystalline phase. On these grounds, the changes in the MI response of such materials during similar annealing are explained. The results uniquely correlate FMR and MI with nanocrystalline structure and magnetic softness, being of interest for the application of nanocrystalline microwires in MI sensors.

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