

## INFLUENCE OF VARIOUS HEAT TREATMENTS ON GIANT MAGNETO- IMPEDANCE EFFECT IN NANOCRYSTALLINE FeSiBNbCu RIBBONS

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In this paper we present studies on the frequency dependence of the magneto-impedance in the range of 0.1–2 MHz for  $\text{Fe}_{73.5}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  and  $\text{Fe}_{73.5}\text{Si}_{16.5}\text{B}_6\text{Nb}_3\text{Cu}_1$  nanocrystalline ribbons, which differ in the sign of the magnetostriction constant. As cast samples were annealed in Ar atmosphere at 560°C, with and without an DC and AC magnetic field. At a fixed frequency, an improvement in the field annealed 13.5% Si samples, when compared with the zero field annealed ones, can be observed. On the 16.5% Si field annealed samples only a reduction of magneto-impedance ratio could be observed, when compared to the non-field annealed ones. Analysis of the magnetic properties and X-ray data shows that the observed changes in magneto-impedance effect are consequence of the induced magnetic anisotropy.

*Keywords:* Nanocrystalline ribbons; Heat treatments; Magnetic anisotropy; Magneto-impedance

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## INTRODUCTION

The giant magnetoimpedance (GMT) was recently discovered by Beach and Berkowitz (1994) and Panina and Mohri (1994) and has been intensively studied for the last years in amorphous (Knobel *et al.*, 1995) and nanocrystalline (Knobel *et al.*, 1996) materials with different shapes – ribbons, wires, thin films and more recently microwires (Vázquez and Zhukov 1997). It consists of the large variation of the impedance upon application of DC magnetic field, that appeared when high frequency current is flowing through the sample. The growing number of works concerning GMI is connected with both advanced technological applications and the fundamental interest related with the study of sensitive transport phenomena.

The magneto-impedance of a magnetic conductor is sensitive to peculiarities of magnetic domain structure, which can be modified by induced anisotropy. Annealing, whether with or without field seems adequate techniques to obtain high GMI effect, as they lead to an induced magnetic anisotropy, changing the shape of the hysteresis loop, the magnetic permeability and parameters of the stabilization of the domain structure.

It is well known that an appropriate annealing of amorphous FeSi-BNbCu alloys, the so called FINEMET, leads to a multiphase structure with crystallites having nano-scale grain size (around 10–20 nm) and excellent soft magnetic characteristics (Herzer, 1989). Bearing in mind both potential technological applications and the nature of the magneto-impedance effect, the aim of this paper is to study the effects of field induced anisotropy on the GMI in FeSiBNbCu nanocrystalline ribbons of two different compositions. It was found that field annealing can lead to an improvement of the GMI ratio reaching a value as high as 24%, which is reasonable for technological applications, considering ribbon shape materials. The results are explained in terms of the field induced anisotropy and its effects in the domain structure of the material.

## EXPERIMENTAL METHODS

Two sets of amorphous ribbons were prepared by melt spinning, having nominal compositions  $\text{Fe}_{73.5}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  ( $\lambda_s \approx +10^{-6}$ ) and

TABLE I Composition, length, thickness and cross sections of the samples used

Composition	Length (mm)	Thickness $h$ (mm)	Cross section (mm <sup>2</sup> )
Fe <sub>73.5</sub> Si <sub>13.5</sub> B <sub>9</sub> Nb <sub>3</sub> Cu <sub>1</sub>	90	0.021	0.003
Fe <sub>73.5</sub> Si <sub>16.5</sub> B <sub>6</sub> Nb <sub>3</sub> Cu <sub>1</sub>	90	0.03	0.03

Fe<sub>73.5</sub>Si<sub>16.5</sub>B<sub>6</sub>Nb<sub>3</sub>Cu<sub>1</sub> ( $\lambda_s \approx -10^{-7}$ ). The geometrical sample characteristics are presented in Table I.

All samples were annealed at temperature  $T = 560^\circ\text{C}$  in Ar atmosphere for 1 h using four different procedures, named A, B, C and D:

- (A) Annealing without magnetic field (conventional heat treatment in order to obtain a nanocrystalline state (Yoshisawa *et al.*, 1988).
- (B) Heat treatment under an axial DC magnetic field,  $H_{\text{HT}} = 15 \text{ kA/m}$ .
- (C) Heat treatment under an axial AC magnetic field,  $A_{\text{AC}} = 15 \text{ kA/m}$ , frequency  $f = 50 \text{ Hz}$ ,  $10 \text{ kHz}$  or  $2 \text{ MHz}$ .
- (D) Heat treatment under transversal AC magnetic field,  $A_{\text{AC}} = 15 \text{ kA/m}$ ,  $f = 50 \text{ Hz}$ .

The samples' structure was checked by X-ray diffraction in the as quenched state (amorphous) and after the heat treatments (nanocrystalline structure). Magnetic characterization of the samples was also performed using hysteresis loops measured by the conventional fluxmetric method. Domain structure was observed using the Bitter technique.

The impedance was measured registering the current induced AC voltage across the sample using a digital oscilloscope. This current was monitored by means of a series resistor, and kept constant during the measurements. The current was fixed at  $I_{\text{AC}} = 7.5, 10$  or  $20 \text{ mA}$ , with frequencies ranging from  $0.1$  to  $2 \text{ MHz}$ . This current intensity is small enough to avoid any heating coming from Joule effects. An axial DC magnetic field ( $H_{\text{DC}} \leq 1720 \text{ A/m}$ ) was applied to the sample and the impedance changes were measured. The relative change of impedance ( $\Delta Z/Z$ ), or GMI ratio, with  $H$  was defined as  $\Delta Z/Z(H) = [Z(H) - Z(H_{\text{max}})]/Z(H_{\text{max}})$ .

## EXPERIMENTAL RESULTS AND DISCUSSION

The field dependence of the GMI ratio, measured at three different frequencies, is shown in Fig. 1 for the sample Fe<sub>73.5</sub>Si<sub>13.5</sub>B<sub>9</sub>Nb<sub>3</sub>Cu<sub>1</sub> after

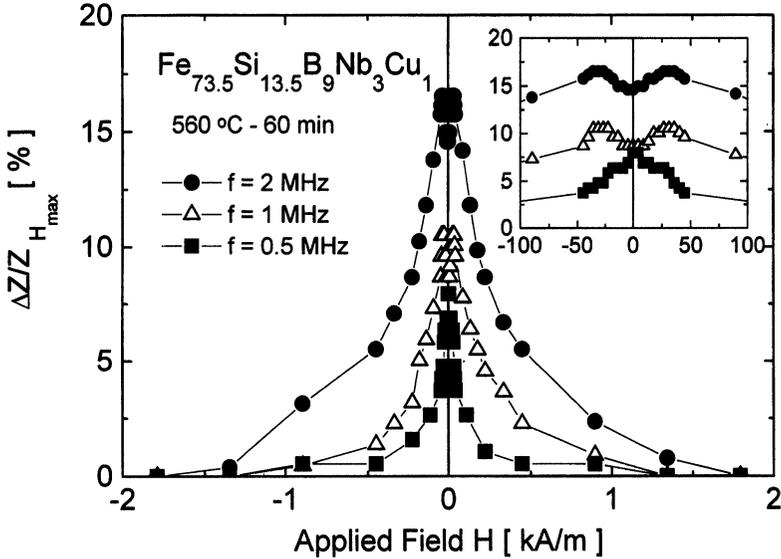


FIGURE 1 Field dependence of the magneto-impedance ratio for the  $\text{Fe}_{73.5}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  sample annealed without field at  $T = 560^\circ\text{C}$  for  $t = 60$  min.

the heat treatment A. For  $f = 0.5$  MHz there is a monotonous decay of the GMI ratio with the field intensity, with a maximum at  $H = 0$ . Increasing the frequency ( $f = 1$  and 2 MHz),  $\Delta Z/Z(H)$  increases, and the maximum shifts to higher magnetic field value. A  $\Delta Z/Z_{H_{\max}}(H)$  value of 17% at 2 MHz is achieved for this sample.

The increase of  $\Delta Z/Z_{H_{\max}}(H)$  with frequency is related to the changes in the penetration depth ( $\delta$ ) of the current. It is well known that a high frequency AC current flowing in a conductor will remain at the surface, the penetration depth being proportional to the circular permeability ( $\mu$ ) and to the current frequency by the following relation (Chikazumi, 1964), where  $\rho$  is the sample resistivity:

$$\delta = \sqrt{\frac{\rho}{\pi\mu f}} \quad (1)$$

The shift of the maximum can be explained in terms of the transverse permeability behaviour with frequency, as already pointed out by Panina and Mohri (1996).

Figure 2 shows the  $\Delta Z/Z_{H_{\max}}(H)$  for the  $\text{Fe}_{73.5}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  submitted to the treatment B in DC field. The  $\Delta Z/Z_{H_{\max}}(H)$  is larger at higher frequencies, like for the sample annealed without field. Comparing both procedures, an improvement of the magneto-impedance in the field annealed sample can be observed, reaching 24% for  $f=2$  MHz. However, for all current frequencies,  $\Delta Z/Z_{H_{\max}}(H)$  shows a decay with the measuring field, with a maximum at  $H=0$ . No shifts for higher fields could be observed as the frequency increases. Data on frequency dependence of the magneto-impedance has already been published in several papers (Beach and Berkowitz, 1994; Knobel *et al.*, 1996), but another independent parameter, which defines the shape of  $\Delta Z/Z(H)$  curve is a driving current value. From the results shown in the Fig. 3 we can conclude that the driving current value dependence of the magneto-impedance ratio is rather weak for the samples after annealing without field. But in near zero field the relationship between the value of the driving current and the shape of  $\Delta Z/Z(H)$  curve can be reliably defined for the samples after heat treatment without field by regime A, having a two-peak form. With increase of the driving current the GMI ratio is

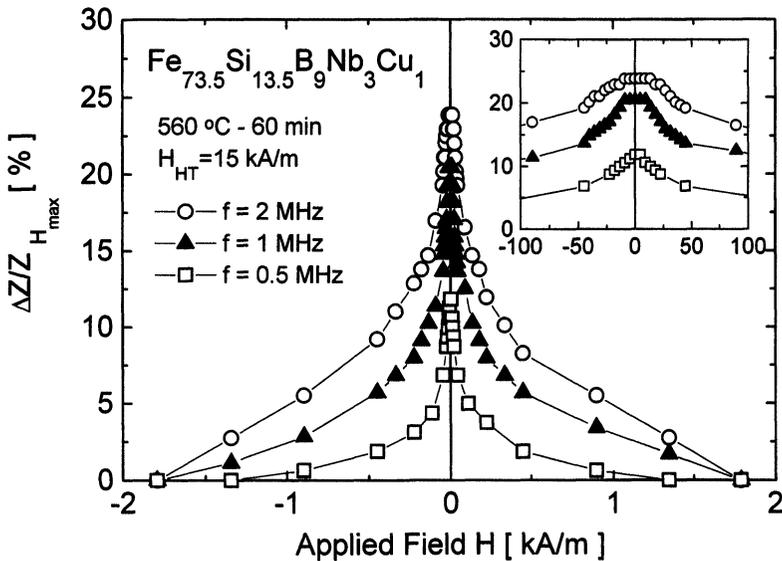


FIGURE 2 Field dependence of the magneto-impedance ratio for  $\text{Fe}_{73.5}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  sample annealed in a DC field  $H_{\text{HT}} = 15000$  A/m, at  $T = 560^\circ\text{C}$ ,  $t = 60$  min.

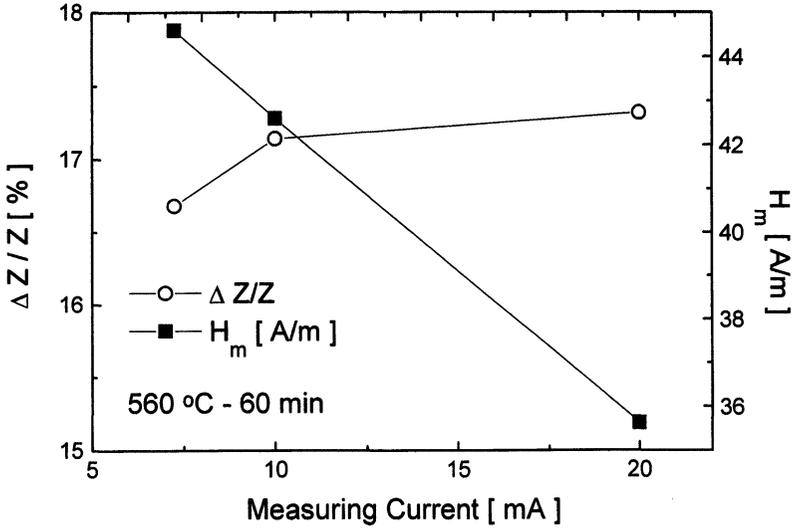


FIGURE 3 The dependence of the  $\Delta Z/Z$  ratio and position of the  $\Delta Z/Z$  peaks ( $H_{\max}$ ) on the driving current value for  $\text{Fe}_{73.5}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  nanocrystalline ribbon after conventional annealing at  $T = 560^\circ\text{C}$  for 1 h.

slightly increased and a displacement of the  $\Delta Z/Z$  peaks towards a smaller fields can be seen. The same behaviour of the magneto-impedance ratio for microwires had been observed earlier by Vázquez and Zhukov (1997).

The  $\Delta Z/Z_{H_{\max}}(H)$  curves reflect the circular permeability behavior with axial applied field, the maximum observed being related to the circular anisotropy field. DC field annealing induces an easy magnetization along the ribbon's axis, so that the circular anisotropy field is reduced, and the maximum of  $\Delta Z/Z_{H_{\max}}(H)$  shifts to lower field values. In fact, the hysteresis loops of the field annealed samples become narrower and more rectangular than those for the as cast and after heating without field. Heat treatment in AC field leads to formation of both induced magnetic anisotropy and different type of magnetic domain structure without stabilization of the domain walls.

Figure 4 shows the maximum  $\Delta Z/Z_{H_{\max}}(H)$  as a function of the current frequency for both compositions. For the  $\text{Fe}_{73.5}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  ribbon an increase of the  $(\Delta Z/Z_{H_{\max}})_{\max}$  can be observed for all frequencies. One should note that these samples have a much smaller cross

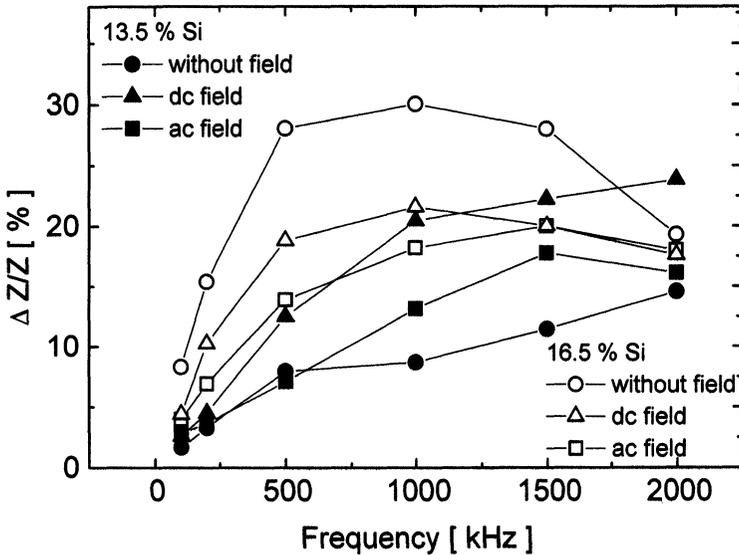


FIGURE 4 Frequency dependence of the maximum  $\Delta Z/Z_{H_{\max}}(H)$  for the annealed (with and without field)  $\text{Fe}_{73.5}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  and  $\text{Fe}_{73.5}\text{Si}_{16.5}\text{B}_6\text{Nb}_3\text{Cu}_1$  samples.

section. In this case, the maximum variation of impedance related to the penetration depth should be observed only for higher frequencies. The AC field annealed sample shows a larger magneto-impedance ratio than the one annealed without field, which might be ascribed to larger values of the circular permeability.

The largest GMI values were found for the  $\text{Fe}_{73.5}\text{Si}_{16.5}\text{B}_6\text{Nb}_3\text{Cu}_1$  samples. For these ribbons there is a reduction of the  $(\Delta Z/Z_{H_{\max}})_{\max}$  after various regimes of the field annealing. The domain structure observation shows that the domains are arranged in a quite regular axial pattern for the field annealed samples. Although MI is not directly related with domain structure this implies that the transversal permeability in the field annealed samples is smaller, and so should be the magneto-impedance ratio. There is a maximum of the  $(\Delta Z/Z_{H_{\max}})_{\max}$  as a function of the frequency, related to the penetration depth frequency dependence. As the frequency increases, even if the permeability changes with applied field, the current will penetrate only inside the thin surface layer, and so, for a high frequency limit the magneto-impedance effect should be smaller.

The evolution of maximum value of  $\Delta Z/Z_{H_{\max}}$  with frequency of AC current for the samples annealed in AC axial or transverse magnetic field (see details in the caption of the figure) is presented in Fig. 5. The obtained dependences indicate that conventional annealing is the best for the sample  $\text{Fe}_{73.5}\text{Si}_{16.5}\text{B}_6\text{Nb}_3\text{Cu}_1$  from the point of view of the GMI effect. Besides, the  $\Delta Z/Z$  ratio is rather sensitive to the conditions of the field annealing. Generally, transverse field annealing results in drastic decrease of the magneto-impedance and the frequency of the axial field annealing is an important parameter too. Axial hysteresis loops for the same regimes of heat treatments as in Fig. 5 are presented in Fig. 6. Significant changes of magnetic anisotropy induced by AC axial and/or transverse field annealing can be outlined.

Recently published papers by Hoffmann and Kronmuller (1996), Lukshina *et al.* (1996) and Herzer (1996) show that application of the magnetic field or stress during devitrification results in induction of rather strong magnetic anisotropy. X-ray patterns presented in Fig. 7 for the samples annealed at different conditions (see figure caption) and exhibiting different  $\Delta Z/Z(f)$  dependences show also certain difference

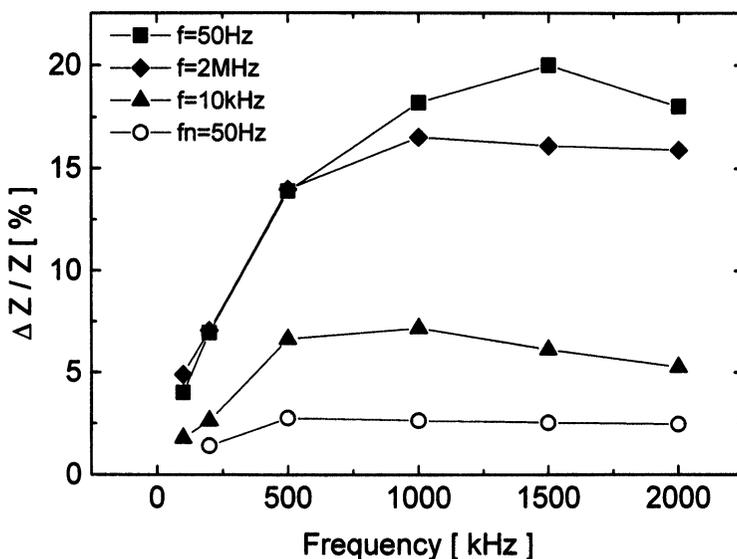


FIGURE 5 Frequency dependence of the maximum  $\Delta Z/Z_{H_{\max}}(H)$  for  $\text{Fe}_{73.5}\text{Si}_{16.5}\text{B}_6\text{Nb}_3\text{Cu}_1$  nanocrystalline ribbon annealed in AC field with various frequencies.

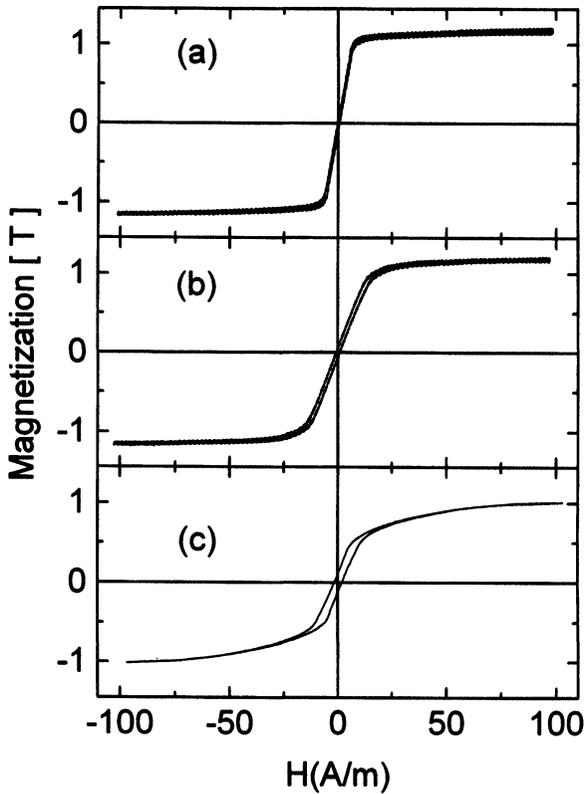


FIGURE 6 Quasistatistical axial hysteresis loops of the nanocrystalline  $\text{Fe}_{73.5}\text{-Si}_{16.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  ribbons: (a) after conventional heat treatment at  $T=560^\circ\text{C}$ ; (b) after AC axial field annealing  $f=10\text{ kHz}$ ,  $T=560^\circ\text{C}$ ; (c) after AC transversal field annealing  $f=50\text{ Hz}$ ,  $T=560^\circ\text{C}$ .

of the relative intensities of the diffraction peaks. This indicates in our opinion difference in grain alignment which finally results in different magnetic anisotropy and, therefore magneto-impedance behaviour.

The origin of the magnetic anisotropy induced by magnetic field or stress annealing needs a special separate study. Among the possible explanations we can point out the pair ordering inside nanograins and internal stresses arising during devitrification owing to difference in specific volumes of amorphous matrix and precipitating crystalline phases. Anyway, the effect of magnetic field annealing on previously introduced thin structure close to the nanograin boundaries as well as

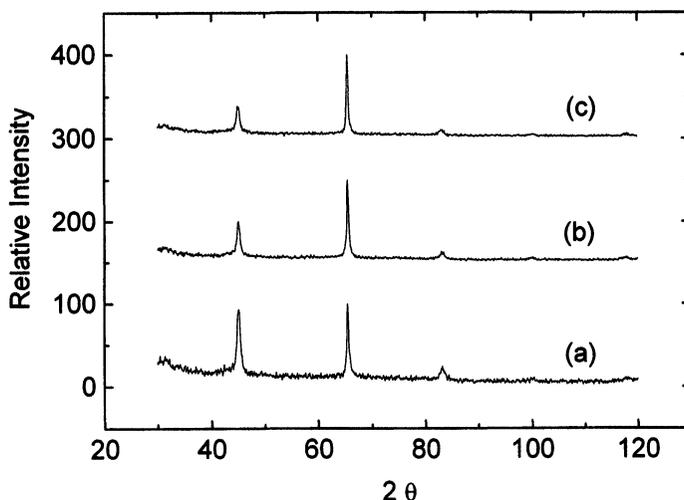


FIGURE 7 X-ray diffraction for nanocrystalline  $\text{Fe}_{73.5}\text{Si}_{16.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  ribbons: (a) after conventional heat treatment at  $T=560^\circ\text{C}$ ; (b) after AC axial field annealing  $f=10\text{ kHz}$ ,  $T=560^\circ\text{C}$ ; (c) after AC transversal field annealing  $f=50\text{ Hz}$ ,  $T=560^\circ\text{C}$ .

the inelastic polarization of the amorphous matrix need further experimental study.

## CONCLUSIONS

Axial field dependence of magneto-impedance of Finemet ribbons with Si content 13.5% and 16.5% after different kinds of heat treatments was studied. It was found generally, that conventional heat treatment results in higher GMI ratio in ribbons with 16.5% Si.

Application AC or DC axial magnetic field during annealing modifies both axial field and frequency dependences of magneto-impedance in both ribbons. Generally, field annealing is favorable in case of ribbon  $\text{Fe}_{73.5}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_1$  while the magneto-impedance of  $\text{Fe}_{73.5}\text{Si}_{16.5}\text{B}_6\text{Nb}_3\text{Cu}_1$  decreases as a result of any magnetic field annealing.

Driving current dependence of the axial magnetic field dependence of magneto-impedance was found for the ribbons with 13.3% Si after conventional heat treatment. The observed dependences were interpreted in a framework of the conventional phenomenology of GMI

effect taking into account change of the skin depth after different heat treatments through the induction of magnetic anisotropy, with the AC driving current frequency and circular magnetic field.

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