Tuning of giant magnetoimpedance effect of amorphous and nanocrystalline microwires

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Abstract

Composite structural materials containing periodic or random arrays of metallic wires often referred to as wire metamaterials have a strong dispersion of the dielectric function in the GHz frequency band. These metamaterials ferromagnetic thin wires magnetoimpedance effect present tuneability of the effective permittivity, by a weak magnetic field or a mechanical stress. We present our studies of the factors affecting soft magnetic properties and giant magnetoimpedance effect (GMI) in thin amorphous and nanocrystalline microwires. We showed that the magnetoelastic anisotropy is one of the most important parameters that determine magnetic softness and GMI effect of glass-coated microwires and annealing can be very effective for manipulation the magnetic properties of amorphous ferromagnetic microwires. Considerable magnetic softening and increasing of the GMI effect is observed in Fe-rich nanocrystalline glass-coated microwires FINEMET-type after nanocrystallization.

1. Introduction

Studies of innovative design of electromagnetic materials continue to be an important issue in microwave technology. A recent trend is to achieve adjustability in these structures. One of the perspective materials is the tuneable metamaterial utilising thin ferromagnetic wires exhibiting magnetoimpedance effect (MI) [1,2]. Our and some other groups work demonstrates that this leads to powerful adjustment of their electromagnetic properties, in particular, effective permittivity, by a weak magnetic field or a mechanical stress[1-3]. Composite structural materials containing periodic or random arrays of metallic wires often referred to as wire metamaterials have a strong dispersion of the dielectric function in the GHz frequency band. This behaviour could be of plasmonic or resonance type for long or short-cut wires, respectively [1,2]. In both cases, the real part of the permittivity can be negative in the dispersion region resulting in many fascinating effects including

negative refraction and frequency selective band-gap or band-pass regimes [1-3].

In thin conducting wires the currents that are responsible for effective permittivity are constrained with the associated resonances determined by the geometrical parameters. The current resonances are damped due to the wire impedance which may increase greatly when the wire magnetisation is changed. This is known as giant magnetoimpedance (GMI) effect [4-6]. In soft magnetic amorphous wires subjected to an external magnetic field GMI is in the range of 100% even at frequencies of few GHz [4-6]. Increase in magnetic losses results in increase in the relaxation parameter which determines the frequency dispersion of the effective permittivity. In the case of plasmonic wire arrays, this will result in considerable decrease in the absolute value of the permittivity and will enhance the wave propagation. Similarly, in cut-wire composites, the increase in relaxation broadens the permittivity dispersion which may even show transformation from resonance to relaxation behaviour [1-3]. Therefore, in composites containing ferromagnetic wires exhibiting GMI effect at GHz frequencies the effective permittivity will depend on the wire magnetic properties via the corresponding dependence of its impedance.

Consequently tailoring of the GMI effect in magnetic wires is essential for performance of the tuneable metamaterial utilising thin ferromagnetic wires.

Recent few years studies of glass-coated magnetic microwires exhibiting excellent sift magnetic properties and Giant magnetoimpedance effect gained considerable attention[5-7]. These properties are related to cylindrical symmetry as well as to crystalline structure of magnetic wires

On the other hand GMI effect and its magnetic field dependence are closely related to the internal and applied stresses [8-10]. Therefore both GMI and magnetic softness can be tailored by magnetic wires processing.

Consequently we present our recent studies of the factors affecting soft magnetic properties and giant magnetoimpedance (GMI) effect in thin amorphous wires.

2. Experimental details

For the preparation of Co- and Fe-rich Finemet-type glass-coated microwires we employed modified Taylor-Ulitovsky technique and/or quenching-and-drawing method described elsewhere [5,6]. This method essentially consists of a simultaneous drawing of the composite microwire (metallic nucleus inside the glass capillary) through the quenching liquid (water or oil) jet onto rotating bobbins. More details on microwires preparation can be found elsewhere [5-8].

We studied glass-coated $Co_{50.69}Fe_{8.13}Ni_{17.55}B_{13.29}Si_{10.34}$ microwires (d=12.8 µm, D=15.8 µm, ρ =0.81) and $Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$ (d=10.7 µm, D=16.4 µm, ρ =0.6). We have measured the magnetic field dependence of impedance, Z, and GMI ratio, $\Delta Z/Z$, for as-prepared samples and after heat treatments [5,6].

We used a specially designed micro-strip sample holder described elsewhere [5]. The sample holder was placed inside a sufficiently long solenoid that creates a homogeneous magnetic field, *H*. The sample impedance, *Z*, was measured using a vector network analyzer from reflection coefficient S₁₁.

The magneto impedance ratio, $\Delta Z/Z$, has been defined as:

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

An axial *DC*-field with a maximum value H_{max} up to 8 kA/m was supplied by magnetizing coils.

The frequency range for the diagonal impedance component has been measured from 1 MHz up to 7 GHz.

Hysteresis loops have been determined by the flux-metric method described elsewhere [5,8].

Crystalline structure and phase composition have been checked using a BRUKER (D8 Advance) X-ray diffractometer with $CuK\alpha$ (λ =1.54 Å) radiation.

The magnetostriction coefficient has been measured using the small angle magnetization rotation method (SAMR) method described elsewhere [11] and recently modified for the case of thin magnetic wires [12].

3. Experimental results and discussion

3.1. Tailoring of magnetic properties and GMI effect in Co-rich amorphous microwires

As can be seen from Fig. 1 typically as-prepared Corich microwires presents typical for Co-rich microwires quasi-linear hysteresis loops and XRD typical for amorphous structure (consisted of a diffuse halo without observation of any crystalline peak). A coercive field of both samples is about $4 \, A/m$ that is typical for amorphous Co-based glass-coated microwires with low negative magnetostriction constant. The origin of such magnetic softness is related to the internal stresses arising during preparation of glass-coated microwires[8,14]. These stresses give rise to an easy magnetization direction

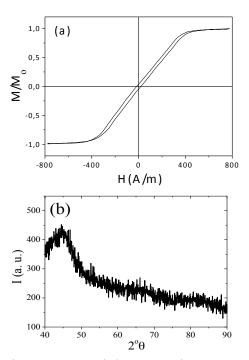


Figure 1: Hysteresis loop (a) and XRD patterns (b) of as-prepared $Co_{50.69}Fe_{8.13}Ni_{17.55}B_{13.29}Si_{10.34}$ microwire

perpendicular to the wire axis, leading to an alignment of the magnetic moments in the direction which is perpendicular (circumferential) to the wire axis.

The annealing even for quite short time and at low temperature leads to significant changing of the magnetic properties of both studied Co-rich microwires (Figs 2a-c).

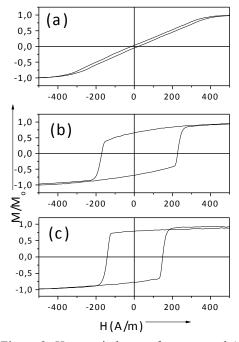


Figure 2: Hysteresis loops of as-prepared (a) and annealed for 5 min at 200° C (b) and 250° C (c) $Co_{50.69}Fe_{8.13}Ni_{17.55}B_{13.29}Si_{10.34}$ microwires

Increasing of the annealing temperature the hysteresis loop becomes more rectangular: remanent magnetization rises with increasing of T_{ann} , although coercivity, H_c , remains almost the same for all annealing conditions.

In spite of considerable magnetic hardening (a remarkable increasing of coercivity from 4 to 200 A/m for $\text{Co}_{50.69}\text{Fe}_{8.13}\text{Ni}_{17.55}B_{13.29}\text{Si}_{10.34}$ sample), both as-prepared and annealed microwires at different annealing conditions present considerable GMI effect as shown in Fig 3.

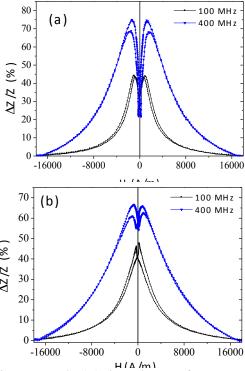


Figure 3: $\Delta Z/Z$ (H) dependences of as-prepared (a) and annealed at 200° C for 5 min (b) $Co_{50.69}Fe_{8.13}Ni_{17.55}B_{13.29}Si_{10.34}$ microwires measured at different frequencies

The difference of $\Delta Z/Z$ (H) dependences measured for as-prepared (Fig.3a) and annealed (Fig.3b)samples is the value of the magnetic field, H_m , at which $\Delta Z/Z$ maximum takes place: for annealed samples H_m –values are lower than for as-prepared samples for all measured frequencies. Increasing the frequency, f, the maximum GMI ratio, $\Delta Z/Z_m$, rises (see Fig.3).

Observed changes of the hysteresis loops after annealing must be associated with the stress relaxation. Indeed the magnetostriction constant depends on stresses (both applied and internal) in according the following equation:

$$\lambda_s(\sigma) = \lambda_s(0) - B\sigma \tag{2}$$

where $\lambda_s(\sigma)$ is the magnetostriction constant under stress; $\lambda_s(0)$ is the zero-stress magnetostriction constant; B is a positive coefficient of order 10^{-10} MPa, and σ stresses. Furthermore, as experimentally shown in Fig. 4, the magnetostriction considerably change after annealing to

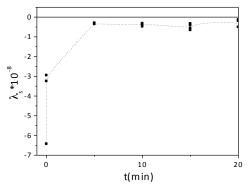


Figure 4: Dependence of the magnetostriction coefficient on annealing time for annealed $Co_{69.2}Fe_{4.1}B_{11.8}Si_{13.8}C_{1.1}$ microwire samples at $T_{ann}=300^{\circ}C$

nearly zero for annealed $Co_{69.2}Fe_{4.1}B_{11.8}Si_{13.8}C_{1.1}$ microwire samples after annealing at $t_{ann} \ge 5$ min.

In addition, if the magnetostriction coefficient is low and negative, we must consider two opposite consequences of the internal stresses. The first contribution is an increase of the total magnetoelastic energy given by eq (3):

$$K_{me} = 3/2 \lambda_s \sigma_i, \tag{3}$$

where λ_s is the saturation magnetostriction and σ_i is the internal stress induced by the glass coating layer during the fabrication technique of glass-coated microwires.

The second one must be related to the stress dependence (either applied or internal stresses: $\sigma = \sigma_{applied} + \sigma_i$) on magnetostriction coefficient described by eq. 2, which is quite relevant for the case of low magnetostriction constant, $\lambda_{s,\theta}$. Accordingly, the magnetostriction constant under stress, $\lambda_{s,\sigma}$, must decrease. Based on the dependencies observed in Figs.2-3, the increase of coercivity and rectangular character of hysteresis loops after annealing must be attributed to the stress relaxation and corresponding magnetostriction changes. We can also assume that the outer domain shell of the annealed Co-rich microwire that exhibits both rectangular hysteresis loop and a GMI effect has high circumferential magnetic permeability.

3.2. Tailoring of GMI effect in nanocrystalline Fe-rich microwires

Usually amorphous Fe-rich microwires present rectangular hysteresis loop as shown in Fig. 5 typical for Ferich amorphous microwires.

Typically Fe-rich amorphous microwires present relatively low GMI effect (1-2 order smaller than Co-rich microwire).

The conventional way of formation of the nanocrystalline structure from the amorphous state, is the heat treatment (with appropriate annealing conditions as temperature, annealing time, heating rate, etc.) [14,15]. The evolution of structural and magnetic properties of Fe_{70.8}Cu₁Nb_{3.1}Si_{14.5}B_{10.6} microwire has been studied in at different annealing temperature in the range between 400-650°C for 1 hour in order to investigate the devitrification process (Fig. 6).

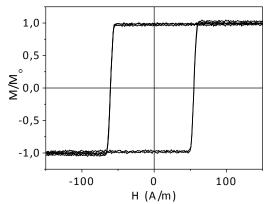


Figure 5: Hysteresis loops of as-prepared $Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$ microwires .

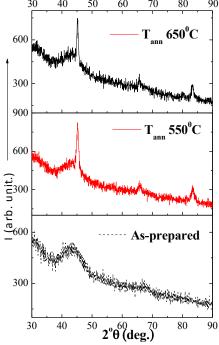


Figure 6: XRD patterns of as-prepared and annealed at different temperature for 1 hour Fe_{70.8}Cu₁Nb_{3.1}Si_{14.5}B_{10.6} glass-coated microwires.

Starting from 550° C to 650° C a main crystalline peak is appearing in the range between 42° to 45° which is correspond to the existence of α -Fe (Si) BCC crystal structure [13,14], as well as another two weak peaks appear in the range between 65° to 85° .

The grain size has been estimated based on Scherrer equation as we previously reported [16]. From this analysis we can underline that the grain size of studied microwires increases from 17 to 22 nm upon increasing the annealing temperature from 550 to 650°C, respectively.

The formation of fine nanograins within the amorphous matrix allows reducing the net magnetostriction, according to this equation:

$$\lambda_{s,eff} = V_{cr} \lambda_{s,cr} + (I - V_{cr}) \lambda_{s,am}$$
(4)

where $\lambda_{s,eff}$ the saturation magnetostriction coefficient and V_{cr} the crystalline volume fraction.

Consequently, the enhanced soft magnetic properties of these kinds of material can be achieved. As can be observed from Fig.7 considerable magnetic softening is observed after samples annealing.

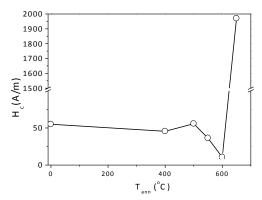


Figure 7: Coercivity dependence on annealing temperature measured in $Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$ microwire with ρ =0.6

Magnetic softening (optimum softest behavior) with quite low value of coercivity is obtained in the samples treated at $T_{ann}\approx\!500\text{--}600~^\circ\text{C}$ which is ascribed to the fact that the first crystallization process has been developed, leading to fine nanocrystals $\alpha\text{-Fe}$ (Si) of grain size around 10-20 nm, such it has been widely reported for FINEMET- ribbons (see for example ref. 17). The precipitation of the second phase results in abrupt increase of the coercivity observed for annealing temperature above 600°C .

Consequently together with magnetic magnetic softening we observed great enhancement of the GMI effect (see Fig.8) after devitrification of studied sample: maximum

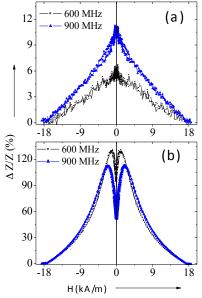


Figure 8: $\Delta Z/Z$ (H) dependences of as-prepared (a) and annealed at $T_{ann} = 550^{\circ}C(b)$ $Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$ microwires measured at different frequencies

GMI ratio increases more than one order from ≈7% up to 130 %. Observed values of GMI effect in less expensive Ferich microwires with nanocrystalline structure are of practical interest for GMI related-applications.

It is worth mentioning that the main interest in amorphous soft magnetic materials is related to their liquidlike structure characterized by the absence of long range ordering and defects typical for crystalline magnetic materials. Therefore, the main parameter determining magnetic softness of amorphous materials is the magnetoelastic anisotropy. The role of the magnetostriction coefficient, λ_s , is already discussed above. But the other contribution is related to the internal stresses, σ_i . In the case of glass-coated microwires the main part of internal stress is induced by the glass coating during the simultaneous rapid solidification of metallic nucleus surrounded by glass coating during the glass-coated microwires preparation [6, 8,18]. The strength of the internal stresses depends on the ρ ratio between the metallic nucleus diameter, d, and total microwire diameter, D ($\rho=d/D$) [8,18] increasing with the the ρ -ratio decreasing. Therefore ρ -ratio is the other relevant parameter determining the magnetoelastic anisotropy.

On the other hand, magnetic softness of amorphous materials can be improved in the case of nanocrystallization at which a formation of two-phase systems consisting of nanocrystalline grains randomly distributed in a soft magnetic amorphous phase is observed [14-17]. After nanocrystallization, such material consists of small (around 10 nm grain size) nanocrystallites embedded in the residual amorphous matrix. Aforementioned magnetic softness is thought to be originated because the magnetocrystalline anisotropy vanishing and decreasing of the magnetostriction coefficient value which can achieve quite low magnetostriction value when the grain size approaches 10 nm [14-17].

Consequently we demonstrated that both annealing allowing internal stresses relaxation, modification of the magnetostriction coefficient by stress relaxation or by devitrification of the amorphous precursor can be used for engineering of magnetic properties and GMI effect of glass-coated micrtowires.

4. Conclusions

We showed that the GMI effect of magnetic wires can be tuned by annealing. We observed that annealing of amorphous Co-rich microwire considerably affects its hysteresis loop, GMI effect. The observed dependences of these characteristics are attributed to stress relaxation and changes in the magnetostriction after sample annealing.

Considerable magnetic softening and increasing of the GMI effect is observed in less expensive Fe-rich nanocrystalline FINEMET-type glass-coated microwires after the devitrification process caused by thermal annealing.

Acknowledgements

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