

# Tailoring of Magnetic Properties and Magnetoimpedance Effect in Thin Amorphous Wires

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We have studied the effect of annealing conditions on magnetic properties of amorphous CoFeNi-based glass-coated microwires. We show that annealing can be very effective for manipulation the magnetic properties of amorphous ferromagnetic glass-coated microwires. Low coercivity and high giant magnetoimpedance (GMI) effect have been observed in as-prepared Co-rich microwires. After annealing of Co-rich microwires we can observe transformation of inclined hysteresis loops into rectangular and coexistence of fast magnetization switching and GMI effect in the same sample. We demonstrate that the switching field value of microwires can be tailored by annealing in the range from 4 to 200 A/m.

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## 1. Introduction

Thin magnetically soft amorphous wires presenting magneto-impedance, GMI effect and fast magnetization switching have been already proposed for a number of promising applications, especially in the field of sensors, electronic devices and smart materials [1, 2]. From the view point of emerging applications the most promising are glass-coated microwires prepared by Taylor-Ulitovsky method, mostly because of the thinnest diameters among the cast wires [2]. Diameter reduction is a crucial factor for miniaturization of the microsensors.

On the other hand soft magnetic properties of amorphous ferromagnetic glass-coated microwires are affected by the magnetoelastic energy related to the presence of glass coating. Therefore minimization of the internal stresses i.e. employing the annealing can considerably improve magnetic softness and GMI effect of these materials [2–3].

Aforementioned GMI effect is one of most promising features for the magnetic sensor applications [2–5]. Recently published papers reported on development of magnetic sensors using GMI effect allowing achieving of pT magnetic field resolution [4, 5]. The origin of the GMI effect is related to the skin effect in soft magnetic conductor [2, 4–6]. High circumferential permeability usually exhibited by amorphous wires with vanishing magnetostriction constant is essential for observation of high GMI effect [4–6].

The magnetic field dependence of the GMI effect is intrinsically related to the magnetic anisotropy and peculiar surface domain structure of amorphous wires [2, 7].

Shape magnetic anisotropy for thin micrometric wires (with metallic nucleus diameters around 10  $\mu\text{m}$ ) becomes

irrelevant for the sample length above 2–3 mm. In the absence of typical magnetocrystalline anisotropy and defects, typical for crystalline materials, it is commonly assumed, that the magnetic field dependence of GMI effect and overall magnetic properties are determined by the magnetoelastic anisotropy,  $K_{\text{me}}$ . For the glass-coated magnetic microwires the presence of non-magnetic glass-coating considerably affects the internal stresses distribution and strength and hence magnetoelastic anisotropy.

This magnetoelastic anisotropy,  $K_{\text{me}}$ , is affected by internal stresses,  $\sigma_i$ , and magnetostriction coefficient,  $\sigma_s$  [2, 4]:

$$K_{\text{me}} = 3/2\lambda_s\sigma_i. \quad (1)$$

The magnetostriction coefficient,  $\lambda_s$ , depends on the chemical composition of the amorphous metallic alloy taking nearly-zero values in amorphous Fe-Co based alloys with Co/Fe $\approx$ 70/5 [8, 9].

The internal stresses originate from the fabrication process involving rapid quenching from the melt of the composite glass-coated microwires consisting of metallic nucleus surrounded by the glass coating. Consequently both thermal stresses induced by the solidification of the metallic nucleus from the surface layer as well as the internal stresses associated to the difference in the thermal expansion coefficients between the glass coating and the ferromagnetic nucleus affect the magnetoelastic anisotropy [10–12]. The strength of the internal stresses induced by the difference of thermal expansion coefficients depends on the  $\rho$ -ratio between the metallic nucleus diameter,  $d$ , and total microwire diameter,  $D$  ( $\rho = d/D$ ) [10–12] increasing with decreasing the  $\rho$ -ratio, i.e. with increasing of the relative volume of the glass coating.

Additionally, previously has been reported that in amorphous alloys the magnetostriction coefficient exhibits stress dependences that can be expressed as [13, 14]:

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$$\lambda_{s,\sigma} = \lambda_{s,0} - B\sigma, \quad (2)$$

where  $\lambda_{s,\sigma}$  is the magnetostriction coefficient under stress,  $\lambda_{s,0}$  is the magnetostriction coefficient under zero-stress and  $B$  is a positive coefficient of order  $10^{-10}$  MPa $^{-1}$ . This stress dependence of the magnetostriction coefficient is expected to be relevant for nearly zero magnetostriction microwire compositions, i.e. for Co-rich microwires. Consequently in the case of Co-rich compositions one can expect considerable influence of the thermal treatment on magnetic properties of glass-coated microwires.

For successful implementation of the microwires in modern devices it is quite important to find the conditions to control and optimize the magnetic properties of amorphous microwires. It can be realized by changing the microwire parameters (composition, diameter of metallic nucleus and thickness of glass shell) and by changing treatment conditions (temperature, annealing, etc.) [2, 4].

Accordingly, the purpose of this paper is to study the effect of annealing of low magnetostrictive Co-rich microwires on magnetic properties and GMI effect.

## 2. Materials and methods

### 2.1. Materials

We have studied glass-coated microwires of Fe<sub>8.13</sub>Co<sub>50.69</sub>Ni<sub>17.55</sub>B<sub>13.29</sub>Si<sub>10.34</sub> ( $d = 12.8$   $\mu$ m,  $\rho = 0.81$ ,  $D = 15.8$   $\mu$ m) prepared by Taylor-Ulitovsky technique (also called in some publications as the drawing and quenching technique), described elsewhere [4].

### 2.2. Magnetic properties and magnetoimpedance

Hysteresis loop of as-prepared and annealed microwires were measured by the induction method [9]. We represent the normalized magnetization,  $M/M_0$  versus magnetic field,  $H$ , where  $M$  is the magnetic moment at given magnetic field and  $M_0$  is the magnetic moment of the sample at the maximum magnetic field amplitude,  $H_0$ .

We have measured magnetic field dependences of impedance,  $Z$ , and GMI ratio,  $\Delta Z/Z$ . We have used a specially designed micro-strip sample holder placed inside a sufficiently long solenoid that creates a homogeneous magnetic field,  $H$ . There, one end of the wire was connected to the inner conductor of a coaxial line through a matched microstrip line, while the other was connected to the ground plane. This sample holder allows measurement of the samples of 6 mm in length. This sample length is sufficiently long allowing to neglect the effect of the demagnetizing factor [3, 15]. We have determined the impedance  $Z$  using the vector network analyzer from reflection coefficient  $S_{11}$ . The employed method has allowed extending the frequency range up to GHz-range [2, 15].

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

$$\Delta Z/Z = [Z(H) - Z(H_{\max})] 100/Z(H_{\max}). \quad (3)$$

The axial DC-field with maximum value,  $H_{\max}$ , up to 20 kA/m was supplied by magnetization coils.

The microwires have been annealed in conventional furnace at temperatures,  $T_{\text{ann}}$  of 200–300 °C for a duration,  $t_{\text{ann}}$ , from 5 to 60 minutes. After annealing the magnetic properties of microwires were studied again.

All samples (as-prepared and annealed microwires) present amorphous structure of metallic core similar to recently studied other Co-rich samples subjected to annealing at similar temperatures [4].

## 3. Results and discussion

Before annealing the studied samples present an inclined hysteresis loop with low coercivity, typical for Co-rich microwires, with low and negative magnetostriction coefficient (Fig. 1a). As we observed, annealing even for quite short time and at low temperature leads to a significant change of the magnetic properties (Fig. 1b–d).

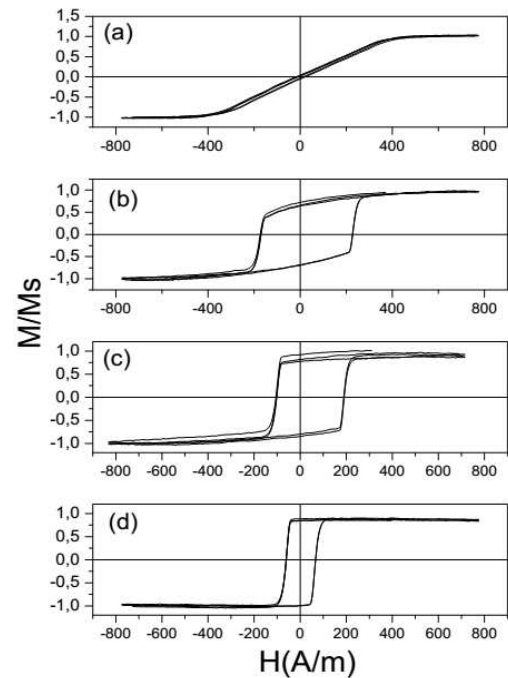


Fig. 1. Hysteresis loops of as-prepared (a) and annealed for 5 min at 200 °C (b), 250 °C (c), and 300 °C (d) Fe<sub>8.13</sub>Co<sub>50.69</sub>Ni<sub>17.55</sub>B<sub>13.29</sub>Si<sub>10.34</sub> microwires.

In spite of considerable magnetic hardening (increasing of coercivity from 4 to 200 A/m), both as-prepared and annealed microwires present considerable GMI effect (Fig. 2a–b). The main difference of observed  $\Delta Z/Z(H)$  dependences for as-prepared and annealed samples is the value of the magnetic field,  $H_m$ , at which  $\Delta Z/Z$  maximum takes place: for annealed samples values of  $H_m$  are much lower than those of the as-prepared samples for all measured frequencies (see Fig. 2a–b).

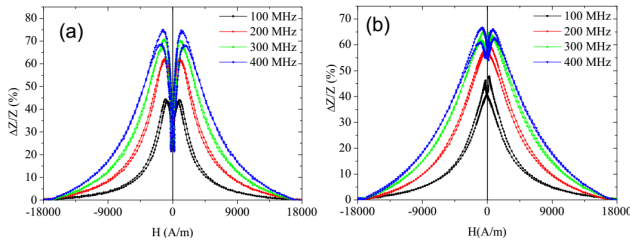


Fig. 2.  $\Delta Z/Z(H)$  dependences of as-prepared (a) and annealed at 200 °C for 5 min (b) microwires.

With the increase of the annealing time and temperature the hysteresis loop becomes more rectangular: remanent magnetization rises with the increasing  $T_{\text{ann}}$ , although coercivity,  $H_c$ , remains almost the same for all annealing conditions (Fig. 3a–b).

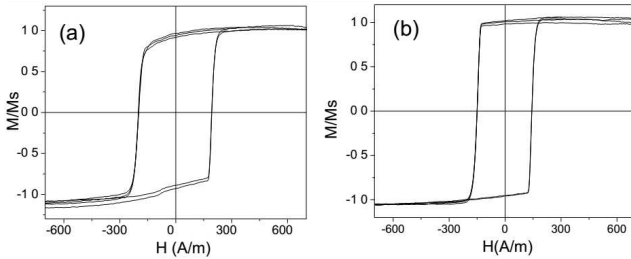


Fig. 3. Hysteresis loops of microwires of  $\text{Fe}_{8.13}\text{Co}_{50.69}\text{Ni}_{17.55}\text{B}_{13.29}\text{Si}_{10.34}$  annealed at 250 °C for 60 min (a) and at 300 °C for 60 min (b).

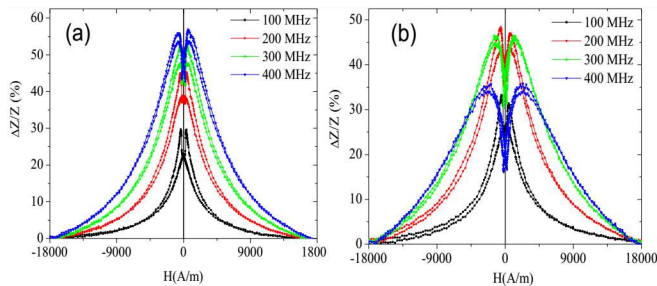


Fig. 4.  $\Delta Z/Z(H)$  dependences of the annealed for 60 min at 250 °C (a) and 300 °C (b)  $\text{Fe}_{8.13}\text{Co}_{50.69}\text{Ni}_{17.55}\text{B}_{13.29}\text{Si}_{10.34}$  microwires, measured at different frequencies.

On the other hand, all annealed samples of  $\text{Fe}_{8.13}\text{Co}_{50.69}\text{Ni}_{17.55}\text{B}_{13.29}\text{Si}_{10.34}$  present a considerable GMI effect (Fig. 4a–b).

For an interpretation of the unusual magnetic hardening observed after annealing (Fig. 1a–d) we must consider not only the stress relaxation (that usually results in decreasing coercivity) but also the change of the character of the remagnetization process, induced by the heat treatment. The observed tendency allows us to assume that annealing induces an axial magnetic anisotropy, which is confirmed by the perfectly rectangular hysteresis loops exhibited by the annealed microwire which are typical

for microwires with a positive magnetostriction constant presenting an axial easy magnetization axis. To explain this unusual dependence of the magnetic properties upon the annealing temperature, we must consider that the stress relaxation affects the magnetostriction coefficient, as also recently described for other Co-rich microwires [3]. Similarly to recently observed changes induced by annealing we can assume that the stress relaxation induced by annealing can change the sign of the magnetostriction constant. Indeed the reported internal stresses values inside the metallic nucleus are between 200 MPa and 5 GPa [10–12]. Experimentally measured values of the  $B$ -coefficient from Eq. (2) reported for similar  $(\text{Co}_{0.94}\text{Fe}_{0.06})_{75}\text{Si}_{15}\text{B}_{10}$  amorphous alloy are about  $1.8 \times 10^{-10} \text{ MPa}^{-1}$  [13]. Moreover recently we have confirmed this assumption by direct measurements of the magnetostriction coefficient in as-prepared and annealed Co-rich microwires [16, 17].

We can also assume that the outer domain shell of the annealed Co-rich microwire, which exhibits both rectangular hysteresis loop and a GMI effect has high circumferential magnetic permeability. This assumption is deduced by observing the much higher GMI ratio of Co-rich microwires that exhibit a rectangular hysteresis loop after annealing, than those of the Fe-rich amorphous microwires also exhibiting similar bulk hysteresis loop characteristics but a much lower GMI effect (usually about 1–5%) [18, 19].

As mentioned above, amorphous Co-rich glass-coated microwires with excellent magnetic softness are quite interesting for microsensor applications. On the other hand there are various typical features related to the composite character of these materials. Most discussed are the internal stresses described above. There are other typical features related to the composite character of glass-coated microwires such as the defects [20]. More detailed studies reveal existence of the bubbles in the glass-coating and of the interfacial layer between the glass coating and metallic nucleus [20, 21]. In spite of the considerable difference in thermal expansion coefficients of the glass and the metallic nucleus the separation between two different materials is usually not observed. Formation of the interfacial layer between the glass and metallic nucleus promotes the adhesion between the glass and the metallic nucleus.

#### 4. Conclusions

We have investigated the possibility to control the magnetic properties of amorphous ferromagnetic microwires by the annealing. We have demonstrated that annealing conditions drastically affect the magnetic properties. We have observed that annealing of amorphous  $\text{Fe}_{8.13}\text{Co}_{50.69}\text{Ni}_{17.55}\text{B}_{13.29}\text{Si}_{10.34}$  microwire considerably affects its hysteresis loop, GMI effect and allows simultaneous observation of a GMI effect and fast magnetization switching in the microwire. The observed dependences of these characteristics are attributed to stress relaxation and changes in the magnetostriction after sample annealing.

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### References

- [1] F. Qin, H.-X. Peng, *Prog. Mater. Sci.* **58**, 183 (2013).
- [2] P.A. Ekstrom, A. Zhukov, *J. Phys. D: Appl. Phys.* **43**, 205001, (2010).
- [3] A. Zhukov, A. Talaat, M. Ipatov, J.M. Blanco, V. Zhukova, *J. Alloys Compd.* **615**, 610 (2014).
- [4] L.V. Panina, K. Mohri, *Appl. Phys. Lett.* **65**, 1189 (1994).
- [5] T. Uchiyama, K. Mohri, Sh. Nakayama, *IEEE Trans. Magn.* **47**, 3070 (2011).
- [6] R. Beach, A. Berkowitz, *Appl. Phys. Lett.* **64**, 3652 (1994).
- [7] N.A. Usov, A.S. Antonov, A.N. Lagar'kov, *J. Magn. Magn. Mater.* **185**, 159 (1998).
- [8] Y. Konno, K. Mohri, *IEEE Trans. Magn.* **25**, 3623 (1989).
- [9] A. Zhukov, J.M. Blanco, M. Ipatov, A. Chizhik, V. Zhukova, *Nanoscale Res. Lett.* **7**, 223 (2012).
- [10] H. Chiriac, T-A. Ovari, A. Zhukov, *J. Magn. Magn. Mater.* **254–255**, 469 (2003).
- [11] A. Zhukov, M. Ipatov, M. Churyukanova, S. Kaloshkin, V. Zhukova, *J. Alloys Compd.* **586**, S279 (2014).
- [12] A.S. Antonov, V.T. Borisov, O.V. Borisov, A.F. Prokoshin, N.A. Usov, *J. Phys. D: Appl. Phys.* **33**, 1161 (2000).
- [13] J.M. Barandiaran, A. Hernando, V. Madurga, O.V. Nielsen, M. Vazquez, M. Vazquez-Lopez, *Phys. Rev. B* **35**, 5066 (1987).
- [14] V. Zhukova, J.M. Blanco, M. Ipatov, A. Zhukov, *J. Appl. Phys.* **106**, 113914 (2009).
- [15] A. Zhukov, A. Talaat, M. Ipatov, V. Zhukova, *IEEE Magn. Lett.* **6**, 2500104 (2015).
- [16] A. Zhukov, K. Chichay, A. Talaat, V. Rodionova, J.M. Blanco, M. Ipatov, V. Zhukova, *J. Magn. Magn. Mater.* **383**, 232 (2015).
- [17] A. Zhukov, M. Churyukanova, S. Kaloshkin, V. Sudarchikova, S. Gudoshnikov, M. Ipatov, A. Talaat, J.M. Blanco, V. Zhukova, *J. Electr. Mater.* **45**, 226 (2016).
- [18] M. Vázquez, J.M. García-Beneytez, J.M. García, J.P. Sinnecker, A. Zhukov, *J. Appl. Phys.* **88**, 6501 (2000).
- [19] M. Churyukanova, V. Zhukova, A. Talaat, J.J. del Val, S. Kaloshkin, E. Kostitcyna, E. Shuvaeva, V. Sudarchikova, A. Zhukov, *J. Alloys Compd.* **615**, S242 (2014).
- [20] A. Zhukov, E. Kostitcyna, E. Shuvaeva, S. Kaloshkin, M. Churyukanova, V. Sudarchikova, A. Talaat, V. Zhukova, *Intermetallics* **44**, 88 (2014).
- [21] A. Zhukov, E. Shuvaeva, S. Kaloshkin, M. Churyukanova, E. Kostitcyna, V. Sudarchikova, A. Talaat, M. Ipatov, V. Zhukova, *J. Appl. Phys.* **115**, 17A305 (2014).