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Magnetic Anisotropy and Super-Sensitive Stress-Magnetoimpedance in Microwires with Positive Magnetostriction

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Abstract—In glass-coated $Co_{71}Fe_{5}B_{11}Si_{10}Cr_{3}$ amorphous ferromagnetic microwires subjected to current annealing, a record sensitivity of the magnetoimpedance (MI) to mechanical tensile stresses (stress MI) up to 100% at 100 MPa in the absence of additional magnetic bias fields is achieved. The current annealing, combining the effect of Joule heating and a circular magnetic field, induces a specific helicoidal/circular-type magnetic anisotropy and, thus, allows one to control the behavior of the MI and stress MI, making the wires more suitable for use in sensor devices. As a result of changing the direction of the easy anisotropy axis, external mechanical stresses lead to a change in the direction of the static magnetization, which causes an increase in the sensitivity of stress MI.

Keywords: amorphous microwire, induced anisotropy, current annealing, magnetization processes, stressmagnetoimpedance (S-MI)

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1. INTRODUCTION

The physical properties of amorphous alloys produced by rapid quenching are significantly changed during heat treatment below the crystallization temperature and transition to the paramagnetic state (Curie point), since it is accompanied by the relaxation of internal mechanical stresses [1–3] induced in the process of manufacturing; the short-range order parameters also change. Therefore, in amorphous ferromagnetic alloys, using heat treatment, it is possible to control the direction of magnetic anisotropy axes, the magnetostriction constant, the internal stress distribution, and the Curie temperature [4–7].

Amorphous alloys are good soft magnetic materials, since their effective magnetocrystalline anisotropy is low due to the averaging effect of the exchange interaction. Upon heating below the crystallization temperature, the magnetoelastic anisotropy decreases due to the relaxation of internal stresses. Since the magnetocrystalline and magnetoelastic anisotropies in such a material are sufficiently reduced, the behavior of the magnetization can be controlled by uniaxial anisotropy induced during heat treatment in the presence of a magnetic field or mechanical stress. Thus, induced magnetic anisotropy is of great practical importance,

making it possible to control the magnetic structure in accordance with specific practical requirements.

In this work, the helicoidal magnetic anisotropy induced by current annealing in glass-coated $Co₇₁Fe₅B₁₁Si₁₀Cr₃$ amorphous microwires makes it possible to obtain a high sensitivity of the shape of the hysteresis loop and the magnetoimpedance to tensile mechanical stresses. Amorphous microwires with a high cobalt content are known as materials very suitable for obtaining large and giant values of MI [3, 8– 10]. With a proper choice of the composition, it is possible to produce microwires with a very small negative magnetostriction, in which the internal tensile stresses create a dominantly circular anisotropy. This creates the conditions for achieving a high sensitivity of the MI effect to changes in the external axial magnetic field. In order to further enhance circular anisotropy (by reducing the spread of the anisotropy axes), in [11, 12], various methods of heat treatment by current annealing (using both alternating and direct currents) were proposed, which made it possible to increase the sensitivity to several hundred percent in the region of low magnetic fields on the order of several oersteds, which are typical of the manifestation of the MI effect. On the other hand, traditional annealing, which

reduces quenching stresses, often leads to a decrease in MI sensitivity, which can be associated with a large scatter of the directions of the light anisotropy axes [13, 14].

The direction of easy magnetization, associated with field-induced anisotropy, is determined by the pairwise ordering of transition metal atoms, which occurs during annealing due to changes in the shortrange order microstructure. This was confirmed by a direct observation using HRTEM analysis [15]. An increase in the degree of ordering can even lead to an increase in the saturation magnetization and the Curie temperature. In the case of a cylindrical geometry of the conductor during current annealing, thermal and magnetic factors act simultaneously and the magnetic field favors the magnetic ordering in the circular direction. For example, current annealing and annealing under tensile stresses were used in [16, 17] to change the easy anisotropy axis in Fe-based wires with positive magnetostriction in order to enhance the MI effect in these inexpensive materials.

High MI sensitivity is usually associated with the reorientation of the direction of magnetization under external actions. In this sense, the induced circular anisotropy is optimal for obtaining a highly sensitive MI response to a change in the axial component of weak magnetic fields. If the external action is a mechanical stress (leading to the stress MI effect), the type of anisotropy depends on the sign of magnetostriction. From the viewpoint of the effect of a tensile stress on MI for use in sensors, it is preferable to use wires with axial anisotropy in the case of a negative magnetostriction constant and wires with circular anisotropy in the case of a positive magnetostriction constant. This creates the conditions for rotating the orientation of the magnetization in the wire under the action of a tensile stress. Thus, to implement a stress MI without using a magnetization bias field, a necessary type of anisotropy must be formed, which can be accomplished by appropriate heat treatment in the presence of a magnetic field.

In this work, we propose to use current annealing in order to create a specific magnetic anisotropy of the circular type in amorphous $Co₇₁Fe₅B₁₁Si₁₀Cr₃$ microwires with an almost zero positive magnetostriction coefficient. This is important for increasing the MI sensitivity [18] and makes it possible to expand the applicability of MI, including stress-sensitive MI at microwave frequencies [19] and pulsed MI for magnetic random access memory (MRAM) [20–22].

2. MATERIALS AND MEASURING TECHNIQUES

In this work, we studied the effect of current annealing, in particular, the amplitude and time of current flow, on magnetic anisotropy, hysteresis loops, and magnetoimpedance (MI) in amorphous

 $Co_{71}Fe_{5}B_{11}Si_{10}Cr_{3}$ microwires with an almost zero positive magnetostriction ($\mu_s \sim 10^{-7}$), produced by the Taylor–Ulitovskii method [23]. An amorphous microwire (with a total diameter of 29.5 μm and a metal core diameter of 23.9 μm) with a length of 15 cm was annealed with a current of 50 mA for 5–60 min. To create a reliable electrical connection at the ends of the wires, the glass sheath was removed and the wires were soldered to the contact pads. All current procedures were performed under the same conditions. The current magnitude was chosen so as to achieve a moderate heating effect in the temperature range of 490– 540 K, which lies below the Curie temperature $(T_{\rm C} = 637 \text{ K})$ and crystallization temperature ($T_{\rm cr} =$ 736 K) of the wires.

Differential scanning calorimetry (DSC) analyses were carried out in an Ar atmosphere at a heating rate of 10 K/min using a highly sensitive Netzsch DSC 204 F1 Phoenix calorimeter. The Curie (T_c = 637 K) and crystallization (T_{cr} = 736 K) temperatures of the wires were determined from the DSC graph using standard applications.

The magnetization of wires under the action of tensile stress up to 1000 MPa was studied by the induction method using two miniature differential coils with an inner diameter of 3 mm. The frequency of the current in the magnetizing coils was 500 Hz, and the maximum amplitude of the magnetizing field was 1000 A/m. To plot a hysteresis loop, the induced electrical signal was digitized and integrated as a function of the magnetic field. The applied stress σ_{ex} in the metal core was estimated as

$$
\sigma_{\text{ex}} = \frac{PE_m}{E_m S_m + E_{\text{gl}} S_{\text{gl}}},\tag{1}
$$

where *P* is the applied load, S_m and S_{gl} are the areas of the metal core and the glass sheath, respectively, taking into account Young's modulus of the metal $(E_m \sim 130 \text{ GPa})$ and the glass sheath $(E_{gl} \sim 70 \text{ GPa})$ [24].

The dependence of high-frequency impedance on the magnetic field was studied in the frequency range 1–100 MHz using a vector network analyzer (Hewlett-Packard 8753E) by measuring the parameter S_{21} (transmission coefficient) in a circuit containing a microwire in a specially designed microwave strip cell [25]. The length of the wire for impedance measurements was 11 mm. The sample in a horizontal position was placed inside a Helmholtz coil, creating a slowly varying magnetic field of up to 3000 A/m. A tensile load was applied at the center of the wire using a suspended load on a diamagnetic thread.

3. RESULTS AND DISCUSSION

The study of the structural parameters of microwires before and after current annealing at 50 mA for

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 $-400 -200$ 0 200 400 -1.25 *H*, A/m *H*, A/m $-2000 -1000$ 0 1000 2000 30 80

130

40 min 60 min

Fig. 1. (a) Hysteresis loops and (b) the real part of the impedance vs. magnetic field for different times of annealing by a current of 50 mA in an amorphous $Co_{71}Fe_{5}B_{11}Si_{10}Cr_{3}$ microwire. (Inset) (a) Residual magnetization M_{r}/M_{s} and (b) the effective circular anisotropy field H_K vs. annealing time. M_s is the saturation magnetization.

60 min was carried out using X-ray diffraction and DSC methods. The X-ray diffraction pattern exhibited a continuous wide halo typical of the amorphous state [26]. It was found that even long-term annealing does not lead to partial crystallization of the samples.

 M_r/M_s

 -0.75

 -0.25

0.25

1.25

0.75

Mr /*Ms* 0.5

1.0

The magnetization and MI processes in amorphous microwires are largely determined by magnetic anisotropy; therefore, current annealing, which causes a change in the character of magnetic anisotropy for λ _s $>$ 0, leads to a change in the behavior of the hysteresis loop and the MI curve [24, 27]. The inset in Fig. 1 shows the dependences of the residual magnetization (M_r) and the effective circular anisotropy field (H_K) , determined from the magnetization and magnetoimpedance curves, on the annealing time at a current of 50 mA. As can be seen from Fig. 1a, the hysteresis loop of the original wires has a rectangular shape, which is typical of samples with axial anisotropy and positive magnetostriction λ _s > 0 , and the MI curves of these wires (in the original form) are characterized by one central peak (Fig. 1b) [28]. After current annealing, the behavior of hysteresis loops changes from rectangular to inclined and the corresponding change in the behavior of the MI from a curve with one peak to a curve with two symmetric peaks (Fig. 1b), corresponding to a change in the easy anisotropy axis, which approaches the circular direction. With increasing annealing time, the slope of the hysteresis loops increases and reaches its maximum at an annealing time of 40 min. The magnitude of the effective magnetic anisotropy field, equal to the field strength at which the MI reaches a maximum, also increases with increasing annealing time. Annealing for 60 min and more leads to an increase in the coercive force, expansion of the MI peaks, and a simultaneous reduction in the sensitivity to a magnetic field.

The change in the behavior of the hysteresis loops and the MI of the wires after current annealing may be caused by the induction of circular magnetic anisotropy due to the ordering of pairs of short-range order atoms upon heat treatment at temperatures below the Curie point in the presence of a circular magnetic field. The magnetic field created by the direct current in the microwire corresponds to 665 A/m at the periphery of the wire. In any case, circular anisotropy cannot be induced in the entire volume of the wire; therefore, near the wire axis, a certain region with axial anisotropy remains, as evidenced by the behavior of the magnetization curves.

It was found that annealing for more than 40 min leads to an increase in the coercive force and a reduction in the MI sensitivity to the magnetic field. As a rule, after a certain time (determined by the activation energy), the kinetic processes are completed and an equilibrium state of the amorphous phase is reached; therefore, a further increase in the annealing time does not change the main magnetic parameters, but crystallization centers may appear, at which domain boundaries can be fixed. To explain the behavior of magnetization, we should also take into account into account the factors such as changes in the microstructure and magnetostriction of samples. The effect of annealing on magnetostriction does not always correlate with a change in magnetic anisotropy. This can be explained in terms of the relaxation of stresses caused by the manufacturing process and changes in the amorphous state of the core material, which reach equilibrium for a longer time. Internal stresses can

Fig. 2. Effect of tensile stress on the behavior of the hysteresis loop in as-prepared amorphous $Co₇₁Fe₅B₁₁Si₁₀Cr₃$ microwire. (Inset) Residual magnetization (*Mr*/*Ms*) vs. tensile stress.

strongly influence the magnetostriction of amorphous wires if its constant λ_s is on the level of 10⁻⁷. The dependence of magnetostriction on mechanical stresses is determined by the formula

$$
\lambda_{s} = \lambda_{s0} - B\sigma_{a}, \quad \sigma_{a} = \sigma_{in} + \sigma_{ex}, \tag{2}
$$

where λ_{s0} is the magnetostriction constant in the absence of mechanical stresses (positive for these wires), *B* is a coefficient having the value of $\sim (1-6) \times 10^{-10} \text{ MPa}^{-1}$ [29]), σ _{*i*} are the internal stresses arising in the process of fast hardening of the amorphous alloy, and σ_{ex} are external tensile stresses. It can be assumed that, with decreasing σ_{in} due to the relaxation of internal stresses, the magnetostriction constant increases along with its contribution to the axial magnetic anisotropy.

The dependence of the magnetization curves on the tensile stress for the wires in the original form and after current annealing is shown in Figs. 2 and 3, respectively. The hysteresis loops of unannealed wires remain rectangular up to a load of σ_{ex} < 500 MPa; in this case, the coercive force (H_c) increases. A further increase in the load leads to a change in the type of hysteresis loop from rectangular to inclined; in this case, *Hc* decreases. This behavior is caused by a change in the sign of λ _s from positive to negative, as follows from Eq. (2). A change in the sign of magnetostriction under the action of external stresses in similar amorphous alloys was earlier observed in [30]. Indeed, the magnetic anisotropy energy E_{ma} has the form

$$
E_{\text{ma}} = -(K + K_{\text{me}}) \cos^2 \theta, \quad K_{\text{me}} = \frac{3}{2} \lambda_s (\sigma_{\text{in}} + \sigma_{\text{ex}}), (3)
$$

Fig. 3. Effect of tensile stress on the behavior of the hysteresis loop in an amorphous $Co₇₁Fe₅B₁₁Si₁₀Cr₃$ microwire after current annealing at 50 mA for 60min: (solid curves) $\sigma_{ex} \leq 500 \text{ MPa}$ and (dotted curves) $\sigma_{ex} > 500 \text{ MPa}$. (Inset) Residual magnetization (M_r/M_s) vs. tensile stress.

where K is the anisotropy constant and θ is the angle between the magnetization and wire axes. As follows from formulas (2) and (3), in the presence of a tensile stress $\sigma_{\rm ex}$, the magnetoelastic anisotropy constant $K_{\rm me}$ increases, reaching its maximum when $\sigma_{ex} = (\lambda_{s0} B\sigma_{\text{in}}/2B$), and can become negative when $\sigma_{\text{ex}} = (\lambda_{\text{so}} B\sigma_{\text{in}}$ /*B*. If *K* is negligibly small, the easy direction of easy magnetization of the wire has a circular direction.

The shape of the hysteresis loops of the annealed wires under the action of the load up to σ_{ex} < 500 MPa transforms from inclined to rectangular (Fig. 3). In this case, the induced anisotropy constant *K* is negative (K < 0), while K_{me} increases (λ_s > 0), and the tensile stress leads to an increase in the contribution of the axial anisotropy. A further increase in the load (above 500 MPa) leads to a reverse change in the magnetization processes, and the hysteresis loops transform from a rectangular to inclined.

It should be noted that a moderate tensile stress applied to annealed microwires almost completely restores the hysteresis loop observed in untreated wires without load (compare with Fig. 2). Consequently, in wires with positive magnetostriction, current annealing and a tensile stress have opposite effects on the formation of the direction of easy magnetization of the wire.

Thus, the resulting anisotropy is a consequence of both factors (current-induced anisotropy and mechanical stresses). These observations are consistent with previous results demonstrating that current heating significantly enhances circular anisotropy in wires with a negative magnetostriction [31], but, at the

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Fig. 4. The real part of the impedance in an amorphous $Co_{71}Fe_{5}B_{11}Si_{10}Cr_{3}$ microwire vs. external magnetic field strength for different values of the tensile stress: (a) in as-prepared state and (b) after current annealing at 50 mA for 60 min. (Insets) The corresponding changes in the effective circular anisotropy field.

same time, leads to partial relaxation of internal stresses [32]. The importance of the results obtained is associated with the possibility of the formation of an almost circular anisotropy in wires with a positive magnetostriction, since external tensile stresses restore the axial anisotropy in them. This opens up a way to obtain microconductor materials with a highly stress-sensitive MI effect.

The main objective of this work is to achieve a highly sensitive stress-MI. The dependence of the effective circular anisotropy field on the tensile stress in untreated wires and in wires after current annealing is shown in Fig. 4. The same figure also shows the magnetoimpedance curves. In unannealed wires under the action of a tensile stress, a noticeable change in the behavior of MI is observed at σ_{ex} = 300 MPa, when the central peak splits into two small symmetric peaks. Very large changes are observed at stresses above 500 MPa due to a change in the sign of the magnetostriction constant and a corresponding change in the orientation of the easy anisotropy axis from axial to circular.

This behavior is consistent with the magnetization processes, as illustrated by the graphs shown in Fig. 2. In the annealed wires, the reverse behavior of MI under the action of a tensile stress is observed. Under the action of a moderate stress (σ_{ex} = 280 MPa), the graphs of MI significantly change their shape from a dependence with two symmetric peaks to a curve with one peak, as shown in Fig. 4b. An increase in the tensile stress causes a reverse trend in the behavior of the MI. This is also consistent with changes in the nature of anisotropy and the type of hysteresis loops.

Since the studies of MI were performed at a frequency of 50 MHz, due to the skin effect, they primarily reflect the processes occurring in a microwire's surface layer. In the original wires, the character of the magnetic anisotropy of this layer is similar to that in the rest of the wire, where the magnetization is oriented in the axial direction. In the annealed wires, circular anisotropy is stronger in the surface layer, which is mainly responsible for the MI effect. As a result, mechanical stresses have a stronger effect on the behavior of MI than on the course of the hysteresis loop.

The discovered possibility of changing the direction of the easy anisotropy axis using current annealing in wires with a positive magnetostriction opens up broad prospects for use in mechanical stress sensors operating in the high-frequency range. Such induced magnetic anisotropy is also of great interest for increasing the MI sensitivity [33] and, even more importantly, creates the conditions for the manifestation of the effect of ultrahigh stress-sensitive MI without using additional bias magnetic fields (or currents).

The dependence of the impedance on the applied tensile stress in the absence of a magnetic field $(H=0)$ for wires in the original state and after current annealing at 50 mA for 60 min is shown in Fig. 5. As can be seen from the graph, the MI of the original wires under the action of a tensile stress of less than 250 MPa does not show a significant change. When the load exceeds 340 MPa, a sharp decrease in the MI begins. This is due to the change in the sign of the magnetostriction constant under the action of tensile stress from positive to negative.

Fig. 5. The real part of the impedance in an amorphous $Co_{71}Fe_{5}B_{11}Si_{10}Cr_{3}$ microwire vs. applied tensile stress in the absence of a magnetic field $(H=0)$: (a) in as-prepared state wire and (b) after current annealing at 50 mA for 60 min. The impedance is normalized to its maximum value.

Thus, the behavior of the MI in annealed wires in the absence of a magnetic field $(H = 0)$ under the action of tensile stress exhibits two opposite trends, caused by a circular magnetic anisotropy induced in the surface layer of the wire and an axial-type magnetoelastic anisotropy. The MI increases linearly at loads $\sigma_{\rm ex}$ < 250 MPa and reaches a maximum at $\sigma_{\rm ex}$ < 300 MPa. A further increase in the load leads to a decrease in both the MI and its sensitivity to changes in the magnetic field. These trends can be explained within the balance between the induced current and the magnetoelastic anisotropy, taking into account the behavior of the magnetostriction constant under the action of a tensile stress. The sensitivity to external mechanical stresses achieved in the absence of a magnetic field exceeds 260% under loads σ_{ex} < 250 MPa; this possibility is realized without using additional bias magnetic fields (or currents).

4. CONCLUSIONS

The current-induced circular anisotropy in wires with a positive magnetostriction creates conditions for a significant change in the MI under the action of tensile stress, which can be called the giant stress-magnetoimpedance (stress-MI) effect. Stress-MI opens up broad prospects for the creation of stress sensors operating in the microwave range. In particular, we have demonstrated the feasibility of control over the direction of the easy anisotropy axis in current-annealed amorphous ferromagnetic microwires by means of a tensile stress. This makes it possible to manipulate the behavior of the magnetoimpedance and the magnetization of wires by external actions. Thus, the current annealing of amorphous ferromagnetic microwires makes it possible to produce sensor elements with the required type of magnetic anisotropy. In addition, wires with a positive magnetostriction and induced circular anisotropy can be used to create smart construction materials with a possibility of remote control of mechanical stresses.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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