



## Research articles

# Monocrystalline Heusler $\text{Co}_2\text{FeSi}$ alloy glass-coated microwires: Fabrication and magneto-structural characterization

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## ABSTRACT

Large scale production of single crystalline phase of Heusler  $\text{Co}_2\text{FeSi}$  alloy microwire is reported. The long microwire ( $\sim 1$  km) with the metallic nucleus diameter of about  $2 \mu\text{m}$  is characterized by well oriented monocrystalline structure ( $B2$  phase, with the lattice parameter  $a = 5.615 \text{ \AA}$ ). Moreover, the crystallographic direction  $[101]$  is parallel to the wire's axis along the entire length. Additionally, the wire is characterized by exhibiting a high Curie temperature ( $T_c > 800 \text{ K}$ ) and well-defined magnetic anisotropy mainly governed by shape. Electrical resistivity measurement reveals the exponential suppression of the electron-magnon scattering which provides strong evidence on the half-metallic behaviour of this material in the low temperature range.

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## 1. Introduction

The production of spintronic devices has intensified the search for new materials, especially half-metals, exhibiting room temperature ferromagnetism [1,2]. This extraordinary behaviour can be understood via distinct character of energy bands for spin up and spin down states. While spin up band overlaps the Fermi energy level ( $E_f$ ) and demonstrates a metallic character, the spin down band shows a semiconductor-like gap at the  $E_f$  [3,4]. Heusler alloys, which are well-known for their diverse and multifunctional magnetic behaviour in various fields of application, have recently attracted much attention as they are theoretically predicted to exhibit 100% spin polarization at  $E_f$  [5–7].

Full-Heusler alloys with the compositional stoichiometry ( $X_2YZ$ ) crystallize in  $L2_1$  structure. Here  $X$  and  $Y$  are transition or rare earth metals (lanthanides) and  $Z$  represents a main group element [8,9]. Among half-metallic alloys, Co-based full-Heusler compounds are most promising materials due to their high Curie temperatures ( $T_c \sim 1100 \text{ K}$ ), high magnetic moment ( $\sim 6 \mu_B/\text{f.u.}$ ) and low Gilbert damping constant ( $\gamma = 0.004$ ) [10].

Although the  $L2_1$  structure is a highly ordered structure, the formation of disordered phases ( $B2$ ,  $A2$ , etc.) may occur during the fabrication of alloys. The exchange between atomic positions of elements for the  $X_2YZ$  Heusler alloy in  $L2_1$  lattice can result in different structural disorders: a) exchange between  $Y$  and  $Z$  atoms results in  $B2$  disorder, b) exchange between  $X$  and  $Y$  atoms results in  $DO_3$  disorder and c) exchange between  $X$ ,  $Y$  and  $Z$  atoms results in the  $A2$  disorder. It was predicted by theoretical calculations [11,12], that the  $Y$ - $Z$  disorder ( $B2$  type structure) has a much lower influence on the spin polarization in comparison to the  $X$ - $Y$  ( $DO_3$  type structure) and  $X$ - $Y$ - $Z$  disorder ( $A2$  type structure) which strongly reduces this feature.

Due to the above mentioned disorder, the production of Heusler alloys brings some disadvantages [13]. Heusler alloys are usually prepared by arc-melting method. Moreover, they require further suitable thermal treatment for a long time (from several hours up to entirely weeks) for obtaining the correct – high ordered phase [14]. It was shown earlier [15], that rapid quenching by Taylor-Ulitovsky technique offers the production of glass-coated metallic microwires without the necessity of long thermal treatments. This single-step method with a high quenching rate of up to  $10^7 \text{ K/s}$  allows to prepare in a short time (of the order of few minutes) up to kilometres of the wire [16]. It is worth mentioning, that due to its high quenching rate it is possible to produce

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Heusler-based microwires with single crystalline phase and well-defined magnetic anisotropy [17–19].

In the given contribution, we present the basic structural and magnetic characterization of glass-covered Heusler  $\text{Co}_2\text{FeSi}$  alloy microwires prepared by Taylor-Ulitovsky technique. We show that it is possible to produce up to kilometres of monocrystalline Heusler-based microwires in few minutes using this simple production technique that may enhance the suitability of  $\text{Co}_2\text{FeSi}$  for spintronic applications.

## 2. Material and methods

The starting master alloy with nominal composition of  $\text{Co}_{50}\text{Fe}_{25}\text{Si}_{25}$  was prepared using arc-melting of pure elements (99.99%) under argon atmosphere. Magnetic glass-coated microwires were fabricated by the Taylor-Ulitovsky technique, which consists on drawing and casting directly from the melted master alloy [20]. The diameter of inner metallic nucleus of microwire sample is around of  $2\ \mu\text{m}$ , while the diameter of external Pyrex coating is around  $20\ \mu\text{m}$ .

Chemical composition, size and shape of the microwire were characterized by scanning electron microscopy (SEM, JEOL-6100) with energy-dispersive X-ray spectroscopy (EDX) option. The microstructural and crystallographic characteristics were investigated by electron backscatter diffraction analysis (EBSD) using SEM TESCAN MIRA 3. The structure and crystalline phase were characterized by single-crystal X-ray diffraction analysis using Oxford Diffraction Xcalibur Nova diffractometer with  $\text{CuK}\alpha$  radiation ( $\lambda = 1.5418\ \text{\AA}$ ). Magnetic hysteresis loops and temperature dependence of magnetization were measured on a single piece of the microwire with the length of  $10\ \text{mm}$  using a SQUID MPMS Quantum Design, along both, the parallel and perpendicular direction with respect to the axis of the wire, in the temperature range from  $10$  to  $400\ \text{K}$  and in the external fields up to  $10\ \text{kOe}$ . The resistivity was measured using standard four probe method in the temperature range from  $8\ \text{K}$  to  $300\ \text{K}$  by employing a Physical property measuring system (PPMS) Quantum Design platform. In order, to make the contacts the glass coating was released mechanically from the ends of the microwire. Afterwards, the contacts were made on metallic nucleus.

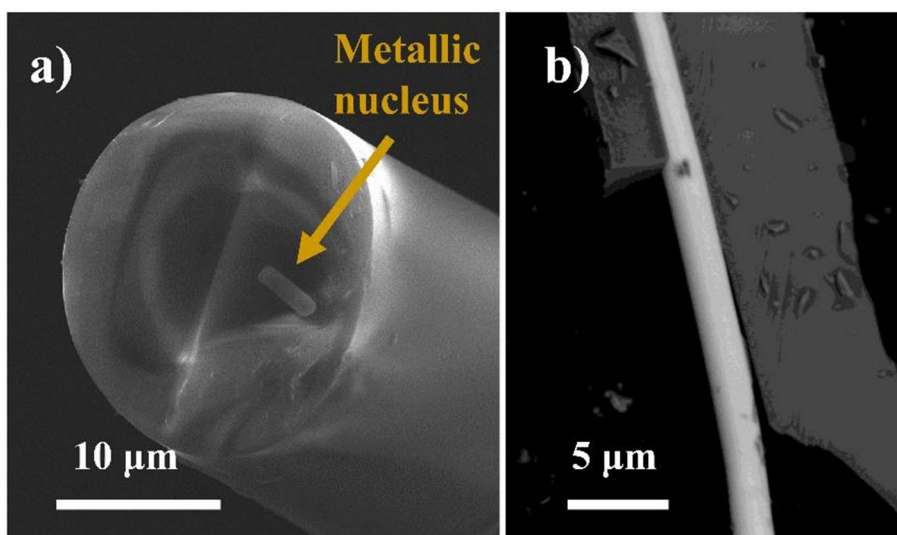
## 3. Results and discussion

Cross-section SEM image of glass-coated microwire shown in Fig. 1(a) displays small metallic core with smooth surface and diameter around of  $2.0\ \mu\text{m}$  that is covered by glass-coating with total diameter of  $20.4\ \mu\text{m}$ . Chemical composition of the metallic nucleus  $\text{Co}_{49.0}\text{Fe}_{25.6}\text{Si}_{25.4}$  checked by SEM EDX is in a good agreement with the expected one.

Fig. 2 displays the X-ray diffraction analysis of the glass-coated  $\text{Co}_2\text{FeSi}$  microwires showing a wide plateau that corresponds to the glass-coating together with the crystalline patterns of the metallic nucleus corresponding to the  $B2$  phase with a lattice parameter  $a = 5.615\ \text{\AA}$ , which is similar to the values presented in other works ( $5.66\ \text{\AA}$ ) [13].

It has been shown earlier [21] that the production of glass coated microwires leads to the formation of oligocrystalline structure, where large crystals (few tens of  $\mu\text{m}$ ) appear along the wire. However, when the production is provided carefully with well selected diameter of the metallic nucleus and glass coating, the monocrystalline structure of the inner metallic alloy can appear. Fig. 3 shows the EBSD analysis performed on an area of the surface of  $\text{Co}_2\text{FeSi}$  microwire after mechanical removal of the external glass coating. Firstly, no grain boundary has been found along the entire wire. Additionally, various analysis of randomly selected pieces of the microwire revealed the same results, which points to the monocrystalline structure of entire microwire with the crystalline  $[101]$  direction lying parallel to the wire's axis. Such preferred orientation can be understood in terms of the tensile stresses induced by drawing and quenching of metallic nucleus during microwire production. During crystallization, the metallic nucleus undergoes strong axial stresses induced by the surrounding glass on the inner metallic alloy due to different thermal expansion coefficient between Pyrex and metallic nucleus.

Volume hysteresis loops (Fig. 4) measured along both, the parallel and perpendicular direction of the applied magnetic field with respect to the wire's axis, revealed well-defined magnetic anisotropy of the metallic nucleus. The comparison between both hysteresis loops points out that the easy magnetization axis is parallel aligned to the microwire's axis. The hysteresis loop for parallel direction shows higher coercivity ( $118.8\ \text{Oe}$ ) comparing to the perpendicular one ( $5.6\ \text{Oe}$ ). This points to the fact that the domain



**Fig. 1.** (a) SEM micrograph showing the cross section of a  $\text{Co}_2\text{FeSi}$  glass-coated microwire. The metallic core is marked with yellow arrow and (b) SEM micrograph showing the metallic core after being released from the glass coating without any grain boundaries. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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