Proceedings of the 16th Czech and Slovak Conference on Magnetism, Košice, Slovakia, June 13–17, 2016

# Ni<sub>2</sub>FeSi Heusler Glass Coated Microwires

# L. GALDUN<sup>a,b,\*</sup>, T. RYBA<sup>a</sup>, V.M. PRIDA<sup>b</sup>, B. HERNANDO<sup>b</sup>, V. ZHUKOVA<sup>c</sup>, A. ZHUKOV<sup>c</sup>, Z. VARGOVÁ<sup>d</sup> AND R. VARGA<sup>a</sup>

<sup>a</sup>Institute of Physics, Faculty of Sciences, P.J. Safarik University, Park Angelinum 9, 041 54 Košice, Slovakia

<sup>b</sup>Departamento de Física, Facultad de Ciencias, Universidad de Oviedo, Calvo Sotelo s/n, 33007 Oviedo, Spain

<sup>c</sup>Departamento de Física de Materiales, Facultad de Ciencias, UPV/EHU, 20009 San Sebastian, Spain

<sup>d</sup>Institute of Chemistry, Faculty of Sciences, P.J. Safarik University, Moyzesova 11, 040 01 Košice, Slovakia

We report on fabrication, structural and magnetic properties of novel Heusler-type glass coated Ni<sub>2</sub>FeSi microwires that were prepared by the Taylor–Ulitovsky method, having a metallic nucleus diameter about 3.9 µm and total sample diameter of 39 µm. This single step and low cost fabrication technique offers to prepare up to km of glass-coated microwires starting from few g of cheap elements for diverse applications. The X-ray diffraction data from the metallic nucleus indicates  $L_{21}$  crystalline structure (a = 5.563 Å), with a possible DO3 disorder. Magnetic measurements determined the Curie temperature well above the room temperature (770 K) together with uniform easy magnetization axis of the metallic core, which predisposes this material to a suitable candidate for spintronic applications.

DOI: 10.12693/APhysPolA.131.851

PACS/topics: 71.20.Be, 72.20.Lp

## 1. Introduction

The great discovery by Heusler in 1903 of a ferromagnetic alloy made from nonmagnetic elements have attracted intensive interest in remarkable materials known as the Heusler alloys. Due to the wide range of extraordinary multifunctionalities exhibited by this kind of materials, there are currently known more than thousands of alloys belonging to this classification of materials [1].

Several Heusler systems have also been reported to be ideal materials for spintronic applications due to their half-metallic behavior, which shows at the Fermi energy level  $(E_f)$  the conducting nature for one spin band whereas the other spin band (usually minority spin) remains insulating. The possibility to exhibit 100% spin polarized conduction electrons attracts a great deal of attention for spintronic applications [2].

Indeed, it was found that one of the most crucial property to achieve high spin polarization in full-Heusler materials lies in the highly ordered  $L_{21}$  crystal phase. Thus, the Heusler alloys prepared by arc melting or grown as thin films are required to initiate the necessary structural ordering through performing long time and high temperature annealing treatments [3]. To avoid this disadvantageous step, alternative rapid quenching production of the Heusler alloys has been recently carried out [4].

Therefore, thin glass-coated magnetic microwires, made by rapid quenching Taylor–Ulitovsky technique, allow to fabricate promising materials exhibiting the high structural order necessary for obtaining high spin polarization. This single-step fabrication method allows to

Ladislav.galdun@student.upjs.sk

produce metallic microwires from arc-melted master alloy of up to km in length and with core diameters ranging from 1 to 30  $\mu$ m coated by insulating glass with thicknesses between 0.5 and 20  $\mu$ m [5].

Theoretical calculations have shown that  $Ni_2Mn$ -based and  $Ni_2Fe$ -based full-Heusler alloys have been predicted to exhibit spin polarization even though their do not show half-metallic character [6]. In their case, the spin polarization is given by a strong asymmetry in the majority and minority density of state close to the Fermi level. A noteworthy example is  $Ni_2MnSn$ , which was experimentally proved to exhibit up to 70% of spin polarization [4].

From a commercial point of view, new research about easy preparation method of thin Heusler wires as spintronic devices is desired. There are many applications in spintronics based on thin magnetic wires like race track memory, domain wall logic, oscillators, etc. [7]. In all of these applications, having a thin magnetic wire with high spin polarization is important. Latest results show that the Taylor–Ulitovsky method is also suitable for the production of thin wire with its diameter down to 90 nm [8].

Here we present glass-coated Ni<sub>2</sub>FeSi microwires prepared by the Taylor–Ulitovsky method. Based on the promising results obtained in Ni<sub>2</sub>MnSn samples [4], we decided to produce microwires of similar composition. However, the production process of microwires brings some restrictions due to evaporation of Mn that can be replaced by Fe. It was shown that it may decrease the size of the band gap in the  $E_f$ , but also increase the Curie temperature [9]. Secondly, Sn has been replaced by Si to avoid the chemical reactions with the glass of the Pyrex during the fabrication of the wires.

Consequently, the aim of this work is to present and discuss some results about the morphological and magnetic properties in full-Heusler  $Ni_2FeSi$  glass-coated microwires.

<sup>\*</sup>corresponding author; e-mail:

# 2. Experimental

Full-Heusler master alloy with nominal composition of  $Ni_{50}Fe_{25}Si_{25}$  was obtained after arc-melting of starting pure elements (99.99%) under argon atmosphere. To obtain the high homogeneity the ingot was four times remelted. Further, ferromagnetic glass-coated microwires were fabricated by drawing and casting directly from the melted master alloy using the Taylor–Ulitovsky method. More information can be found in [10]. Final wire consists of a metallic nucleus with a diameter of 3.9 µm and a total diameter of the glass-coated microwire of 39 µm.

The morphological characteristics and chemical composition of the Ni<sub>2</sub>FeSi microwire were determined by scanning electron microscope (SEM) with energydispersive X-ray spectroscopy (EDX) option. Structure and crystalline phase were characterized by single-crystal X-ray diffraction (XRD) analysis on Oxford Diffraction Xcalibur Nova diffractometer with Cu  $K_{\alpha}$  radiation ( $\lambda =$ 1.5418 Å). Hysteresis loops along the parallel and perpendicular directions with respect to the wire's axis were measured using a vibrating sample magnetometer VSM Versalab (QD), in the temperature range from room temperature (RT) down to 50 K. A single piece of microwire with length of 2.2 mm was used for the characterization of the volume magnetic properties. Thermomagnetic measurements were performed in a PPMS (Physical Property Measurement System — Quantum Design Model 6000) platform in the temperature range of RT up to 1000 K and at the low applied field value of 100 Oe.

#### 3. Results and discussion

Figure 1 shows a typical SEM image of a fracture crosssection of the metallic nucleus together with the surrounding Pyrex glass coating. The metallic core with smooth surface and diameter of about 3.9  $\mu$ m is coated by glass with total diameter of 39  $\mu$ m.

The SEM/EDX chemical analysis of the metallic nucleus in the as-quenched microwires was performed after the removing of the Pyrex glass-cover by chemical etching with diluted HF acid. Subsequently the analysis of three different sample pieces revealed slightly out of stoichiometric Heusler alloy with the average composition of  $Ni_{51.0}Fe_{23.6}Si_{25.4}$  and with the variation of  $\pm 1\%$  of each element.

The X-ray diffraction analysis of glass-coated Ni<sub>2</sub>FeSi microwires carried out at RT (Fig. 1), revealed a wide plateau corresponding to the amorphous glass-coating together with the crystalline patterns, indicating the most representative high ordered  $L2_1$  cubic structure. On the other hand, chemical analysis (which shows that our sample is a slightly out of nominal 2:1:1 composition) points to the fact that some structural disorder may be present. The mutual exchange between X and Y atoms positions of elements for the X<sub>2</sub>YZ Heusler alloy in  $L2_1$  lattice can result in the DO3 disorder, which is difficult to detect by X-ray diffraction analysis with an ordinary Cu  $K_{\alpha}$  source alone. The lattice parameter of crystalline structure was estimated from the angle position of the Bragg peaks and their Miller index giving a value of a = 5.563 Å, which is similar to the lattice parameter for Co<sub>2</sub>FeSi alloy [11].

Figure 2 displays the thermomagnetic evolution of the Ni<sub>2</sub>FeSi alloy microwire from RT up to 1000 K (black curve) and cooling down (red curve), for the static magnetic field value of 100 Oe applied in the parallel direction with respect to the wire axis. The stress-induced relaxation process causes the increase of the magnetization of the metallic nucleus with a maximum about 574 K. Further, the magnetization of the Heusler phase decreases with increase of the temperature up to the Curie temperature  $\approx$  770 K. After annealing the magnetization shows a plateau most probably due to the crystallization of new non-Heusler crystalline phases or due to the various chemical or structural ordering [12].



Fig. 1. X-ray diffraction profile and SEM micrograph of glass-coated Ni<sub>2</sub>FeSi microwire.

In order to determine the effective magnetic anisotropy of the glass-coated Ni<sub>2</sub>FeSi microwire, the magnetic hysteresis loops were measured in both, the parallel and perpendicular directions of the applied magnetic field with respect to the wire's axis at 50 K (Fig. 2). Besides the soft magnetic behavior of the metallic nucleus, the comparative analysis of hysteresis loops indicates a clear anisotropic behavior with the easy magnetization axis parallel oriented to the wire's axis. The magnetic hysteresis loop measured in the perpendicular direction shows a gradual increase in magnetization up to the higher magnetic field value of 3700 Oe, comparing with the magnetization saturation field value of the magnetic hysteresis loop measured along the parallel direction to the wires axis (2700 Oe). In addition, the squared shape of magnetic hysteresis loop observed in the parallel direction, with coercive field  $H_C = 17.1$  Oe, points to the fact that the domain wall propagation dominates in the parallel direction to the easy magnetization axis, exhibiting a more bistable magnetic behavior, while the hysteresis loop measured in the perpendicular direction reveals dominant magnetization rotation process. It is worth mentioning



Fig. 2. Temperature dependence of magnetization measured in the parallel direction at the magnetic field of 100 Oe and hysteresis loops measured at 50 K in parallel (black) and perpendicular (red) direction with respect to the axis of Ni<sub>2</sub>FeSi glass-coated microwire.

that well-defined effective magnetic anisotropy of glasscoated  $Ni_2FeSi$  microwires, oriented parallel to the wire's axis is another of the crucial parameters for spintronics applications.

# 4. Conclusions

In this work, we report on the fabrication together with the characterization of structural and magnetic properties for novel full-Heusler-type of glass coated Ni<sub>2</sub>FeSi microwires with metallic nucleus diameter about 3.9  $\mu$ m. Through this cost effective rapid quenching method it becomes possible to prepare glass coated alloys with homogeneous chemical composition and highly ordered crystalline structure, which is one of the crucial parameters for obtaining spin polarization in full-Heusler compounds. The structural analysis indicates highly ordered  $L2_1$  crystalline phase with a possible DO3 disorder and lattice parameter a = 5.563 Å. Magnetic hysteresis loops measured along both in parallel and perpendicular direction point to the well-defined easy magnetization axis oriented along the wire axis. These properties, together with the high Curie temperature about 770 K, predispose this alloy for its application in spintronics devices. Therefore, easily and low cost preparation of the Heusler-type glass-coated microwires from cheap elements by using the Taylor–Ulitovsky technique might be crucial from the point of view of future novel technological applications.

### Acknowledgments

This work has been financially supported by NanoCEXmat ITMS 26220120035, VEGA 1/0060/13, APVV-0027-11, VVGS-PF-2015-495, VVGS-PF-2016-72614, Spanish MINECO research funds under project No. MAT2013-48054-C2-2-R and Principado de Asturias by FICyT under GIC-FC-15-GRUPIN14-085 research project. The scientific support from University of Oviedo SCT's, particularly SEM and XRD units are also acknowledged.

#### References

- T. Graf, C. Felser, S.S.P. Parkin, *Prog. Solid State Chem.* 39, 17 (2011).
- [2] S. Ishida, S. Mizutani, S. Fujii, S. Asano, *Mater. Trans.* 47, 31 (2006).
- [3] M. Belmeguenai, H. Tuzcuoglu, M.S. Gabor, T. Petrisor Jr., C. Tiusan, F. Zighem, S.M. Chérif, P. Moch, J. Appl. Phys. 115, 043918 (2014).
- [4] N. Nazmunnahar, T. Ryba, J.J. del Val, M. Ipatov, J. Gonzalez, V. Hašková, P. Szabó, P. Samuely, J. Kravcak, Z. Vargova, R. Varga, J. Magn. Magn. Mater. 386, 98 (2015).
- [5] H. Chiriac, T.-A. Óvári, *Progr. Mater. Sci.* 40, 333 (1996).
- [6] Y. Qawasmeh, B. Hamad, J. Appl. Phys. 111, 033905 (2012).
- [7] D.A. Allwood, G. Xiong, C.C. Faulkner, D. Atkinson,
  D. Petit, R.P. Cowburn, *Science* **309**, 1688 (2005).
- [8] T.-A. Óvári, N. Lupu, S. Corodeanu, H. Chiriac, *IEEE Trans. Magn.* 50, 11 (2014).
- [9] B. Balke, G.H. Fecher, H.C. Kandpal, C. Felser, *Phys. Rev. B* 74, 104405 (2006).
- [10] V.S. Larin, A.V. Torcunov, A. Zhukov, J. González, M. Vasquez, L. Panina, J. Magn. Magn. Mater. 249, 39 (2002).
- [11] K. Srinivas, M.M. Raja, S.V. Kamat, J. Alloys Comp. 619, 177 (2015).
- [12] J. Dubowik, I. Gościańska, K. Załęski, Y.V. Kudryavtsev, Y.P. Lee, *IEEE Trans. Magn.* 65, 2534 (2009).