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

Influence of Mn Doping on Magnetic and Structural Properties of Co_2FeSi Heusler Alloy

L. Galdun[a,b], T. Ryba[a], J. Kováč[c], V.M. Prida[b], B. Hernando[b], Z. Vargová[d] and R. Varga[a,*]

[a]Institute of Physics, Faculty of Sciences, P.J. Safarik University, Park Angelinum 9, 041 54 Košice, Slovakia
[b]Departamento De Física, Facultad de Ciencias, Universidad de Oviedo, Calvo Sotelo s/n, 33007 Oviedo, Spain
[c]Institute of Experimental Physics, SAS, Watsonova 47, 040 01 Košice, Slovakia
[d]Institute of Chemistry, Faculty of Sciences, P.J. Safarik University, Moyzesova 11, 040 01 Košice, Slovakia

We have studied the effect of Mn doping on structural and magnetic properties of Co_2FeSi Heusler alloy. Co_2FeSi, $Co_2Mn_{0.25}Fe_{0.75}Si$ and $Co_2Mn_{0.5}Fe_{0.5}Si$ alloys were prepared by melt spinning technique which offers fast and simple production of large amount of materials in a single production step. The rapid quenching method provides an opportunity to prepare Heusler alloys with highly ordered $L2_1$ structure confirmed by X-ray analysis in all the samples. Magnetic measurements revealed high Curie temperatures (> 1000 K) and well defined easy magnetization plane parallel with the ribbon plane. Those attributes predispose given samples for applications in spintronics.

DOI: 10.12693/APhysPolA.131.866

PACS/topics: 71.20.Be, 72.20.Lp

1. Introduction

Half-metallic Heusler alloys have been proposed as ideal candidates for spintronic devices due to the theoretical predictions to exhibit 100% spin polarization. This property is a result of a band gap in one of the spin channels in the density of states at the Fermi level (E_F) [1]. Co_2-based full-Heusler alloys have a special interest thanks to their large minority band gap, high magnetic moment, and the Curie temperature (T_C) [2].

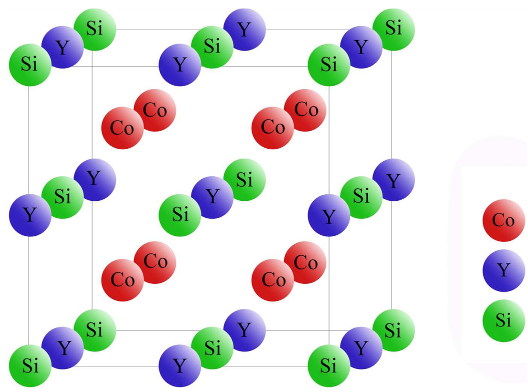

Fig. 1. Schematic representation of the $L2_1$ structure adopted by the $Co_2Mn_yFe_xSi$ Heusler alloys.

One of the most important feature for the applications in spintronics for the half-metallic ferromagnets is the size and stability of the band gap in one of the spin channels at E_F. Small gaps and gaps with E_F located close to the band edges may easily be destroyed [3]. Co_2FeSi is one of the most promising materials for spintronic applications due to the highest magnetic moment (5.97 ± 0.05 μ_B at 5 K) per unit cell and the highest T_C (1100 K) among Co_2-based Heusler alloys [4]. Calculations also show that the structural disorder (especially *A*2 and *DO*3) or any small change in lattice parameters may destroy the half-metallicity by shifting the E_F outside of the band gap [3, 5]. Thus, the highly ordered $L2_1$ phase is required to maximize the values of spin polarization. Therefore, the Heusler alloys prepared by arc melting or grown thin films need an additional long thermal treatment (weeks) at high temperatures [5–7]. This disadvantageous post production can be eliminated by using rapid quenching methods like melt spinning, which offers the benefit of easy preparation of large amount of the Heusler alloys with correct $L2_1$ crystalline structure [8] (Fig. 1).

On the other hand, it was shown that the E_F is near to the bottom of the conduction band in the case of Co_2FeSi. It was shown earlier [3] that the substitution of Fe by Mn atoms may shift the E_F closer to the middle of the band gap and therefore stabilize the half-metallicity.

Here, we report on the influence of Mn doping on magnetic and structural properties of rapidly quenched Co_2FeSi Heusler alloy. We show that melt spinning technique is found to be an effective method in the production of the Heusler alloys.

2. Experimental

As-cast ingots with a nominal composition of $Co_{50}Fe_{25}Si_{25}$, $Co_{50}Mn_{6.25}Fe_{18.75}Si_{25}$ and $Co_{50}Mn_{12.5}Fe_{12.5}Si_{25}$ were produced by arc-melting in Ar atmosphere, using highly pure elements. Further,

*corresponding author; e-mail: rastislav.varga@upjs.sk

ribbons 2.3–2.4 mm width and 5–13 mm length were prepared by melt spinning technique in He atmosphere. The thickness of the ribbons is about 60 μm.

Morphological characteristics and chemical composition of the ribbons were performed by scanning electron microscope (SEM) with energy-dispersive X-ray spectroscopy (EDX) option. Structure and crystalline phase was determined by X-ray diffraction (XRD) using Seifert XRD 3000 T/T diffractometer with Mo K_α radiation ($\lambda = 0.7107$ Å) at room temperature (RT) and evaluated by the software FullProf. Volume hysteresis loops along the parallel and perpendicular with respect to the ribbon axis were measured using vibrating sample magnetometer VSM Versalab (QD) at RT. Thermomagnetic measurements were performed by VSM (home-made) at the magnetic field 80 kA m^{-1}.

3. Result and discussion

SEM micrographs of the fractured cross-section of ribbons Co_2FeSi (Fig. 2a) $Co_2Mn_{0.25}Fe_{0.75}Si$ (Fig. 2b) and $Co_2Mn_{0.5}Fe_{0.5}Si$ (Fig. 2c) show smooth fracture surface with any columnar microstructure. SEM/EDX chemical analysis revealed an averaged composition of $Co_{48.7}Fe_{25.9}Si_{25.4}$, $Co_{47.4}Mn_{8.4}Fe_{20.1}Si_{24.1}$ and $Co_{51.3}Mn_{12.6}Fe_{12.6}Si_{23.5}$. The samples are homogeneous and the real compositions are in good agreement with nominal ones. Ribbons formed by melt spinning technique have an increased macroscopic chemical homogeneity, similar as it has been reported in Ni–Mn–Co–In system [9].

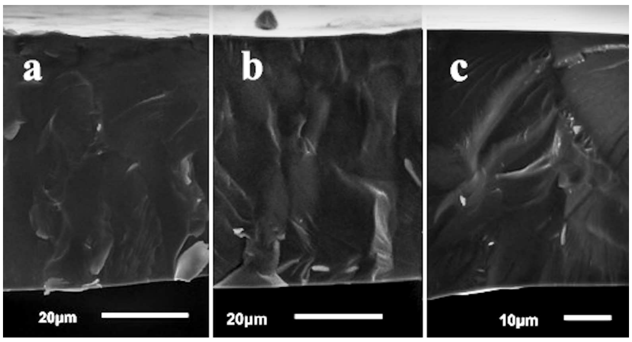

Fig. 2. SEM micrograph of (a) Co_2FeSi, (b) $Co_2Mn_{0.25}Fe_{0.75}Si$, (c) $Co_2Mn_{0.5}Fe_{0.5}Si$ rapidly quenched ribbons.

The X-ray diffraction patterns of rapidly quenched Co_2FeSi, $Co_2Mn_{0.25}Fe_{0.75}Si$ and $Co_2Mn_{0.5}Fe_{0.5}Si$ ribbons observed at room temperature show a Heusler type $L2_1$ crystalline structure (Fig. 3). It was found that the main characteristic super-lattice reflections [(111), (200), (311)] peaks and fundamental (220), (400), (422), etc., peaks represent high ordered $L2_1$ cubic structure (space group: *Fm-3m*). The lattice parameter of Co_2FeSi was found to be 5.640(7) Å, of $Co_2Mn_{0.25}Fe_{0.75}Si$ to 5.652(2) Å and of $Co_2Mn_{0.5}Fe_{0.5}Si$ to 5.650(5) Å, which are in agreement with similar composition [3, 10].

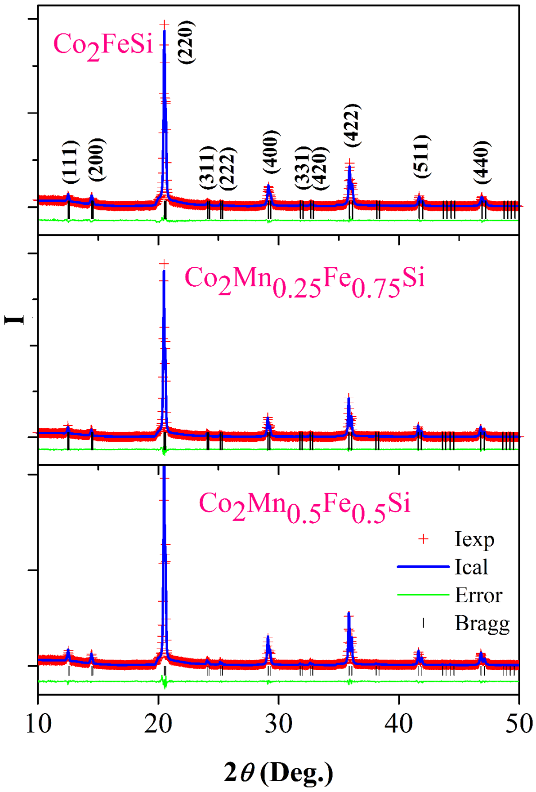

Fig. 3. X-ray diffraction patterns of rapidly quenched Co_2FeSi, $Co_2Mn_{0.25}Fe_{0.75}Si$ and $Co_2Mn_{0.5}Fe_{0.5}Si$ ribbons.

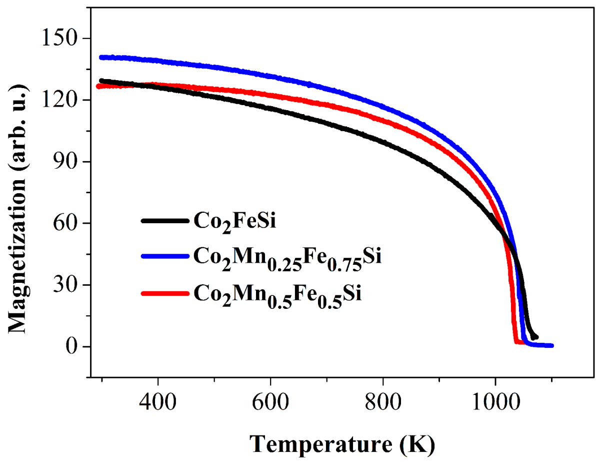

Fig. 4. Temperature dependence of saturation magnetization of rapidly quenched Co_2FeSi, $Co_2Mn_{0.25}Fe_{0.75}Si$ and $Co_2Mn_{0.5}Fe_{0.5}Si$ ribbons.

The temperature dependence of saturation magnetization of the alloys (Fig. 4) shows the ordinary ferromagnetic behavior — magnetization decreases with increase of temperature. The Curie temperature (T_c) of Co_2FeSi Heusler ribbon was found to be about 1070 K and it corresponds well with the value from the literature [10]. In the case of $Co_2Mn_{0.25}Fe_{0.75}Si$ and $Co_2Mn_{0.5}Fe_{0.5}Si$ the slight change in T_c ($\approx$ 1050 K and $\approx$ 1036 K, respectively) is consistent with the partial substitution (25 and 50%) of Fe by Mn atoms and the values are in a good agreement with the reported material $Co_2Mn_{0.5}Fe_{0.5}Si$ on nanoscale [11]. It should be noted that the T_c of both materials are similar and much higher than RT, which makes them attractive candidates for applications in spintronics.

In order to observe the magnetic anisotropy of the Co_2FeSi $Co_2Mn_{0.25}Fe_{0.75}Si$ and $Co_2Mn_{0.5}Fe_{0.5}Si$ melt spun ribbons, the magnetic hysteresis loops were measured in applied field parallel and perpendicular with respect to the ribbons plane.

From Fig. 5 is evident that the easy magnetization plane is parallel to the plane of the ribbons while the magnetization hard plane is perpendicular to the ribbon's plane. The coercitive field for Co_2FeSi is 3.44 Oe in parallel and 77.91 Oe in perpendicular direction. In the case of $Co_2Mn_{0.25}Fe_{0.75}Si$ and $Co_2Mn_{0.5}Fe_{0.5}Si$ are the coercitive fields 2.56 and 4.49 Oe for the parallel and 94.44 and 72.82 Oe for the perpendicular direction.

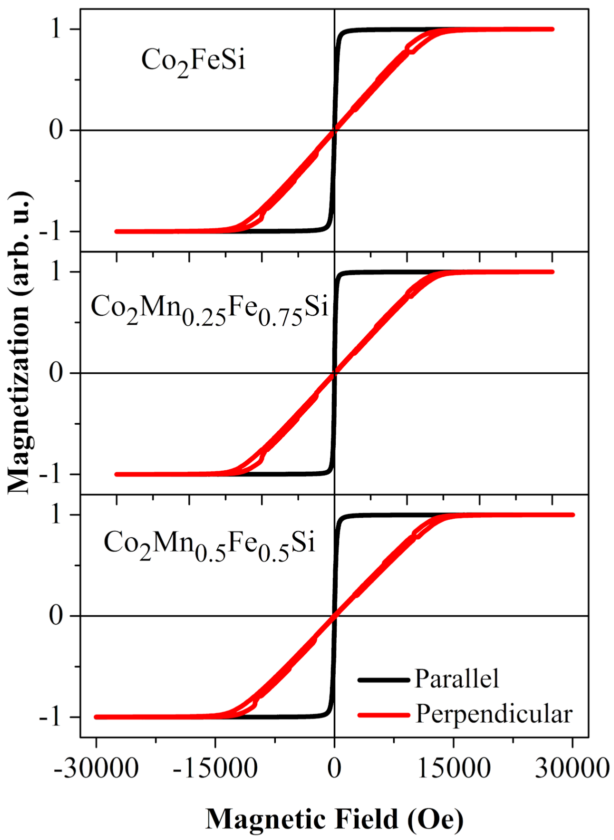

Fig. 5. Magnetic hysteresis loops of rapidly quenched Co_2FeSi, $Co_2Mn_{0.25}Fe_{0.75}Si$ and $Co_2Mn_{0.5}Fe_{0.5}Si$ ribbons.

4. Conclusions

In this work we report on the influence of Mn doping on structural and magnetic properties of Co_2FeSi Heusler alloy. We show that with melt spinning technique it is possible to prepare Heusler alloys with correct $L2_1$ crystalline structure which is essential for high spin polarization values. Co_2FeSi, $Co_2Mn_{0.25}Fe_{0.75}Si$ and $Co_2Mn_{0.5}Fe_{0.5}Si$ melt spun ribbons show soft magnetic behavior with the easy magnetization plane parallel to the ribbons plane. The given attributes predispose those materials for spintronic applications.

Acknowledgments

This work has been financially supported by NanoCEXmat ITMS 26220120035, VEGA 1/0164/16, 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

[1] 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] B. Balke, H.C. Kandpal, G.H. Fecher, C. Felser, *J. Magn. Magn. Mater.* **310**, 1823 (2007).

[4] L. Makinistian, M.M. Faiz, R.P. Panguluri, B. Balke, S. Wurmehl, C. Felser, E.A. Albanesi, A.G. Petukhov, B. Nadgorny, *Phys. Rev. B* **87**, 220402 (2013).

[5] *Spintronics: From Materials to Devices*, Eds. C. Felser, G.H. Fecher, Springer Science & Business Media, 2013.

[6] T. Ryba, Z. Vargova, J. Kovac, P. Diko, V. Kavečanský, S. Piovarči, C. Garcia, R. Varga, *IEEE Trans. Magn.* **50**, 2600103 (2015).

[7] H. Tuzcuoglu, M.S. Gabor, T. Petrisor Jr., C. Tiusan, F. Zighem, S.M. Chérif, P. Moch, *J. Appl. Phys.* **115**, 043918 (2014).

[8] 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).

[9] W. Cai, Y. Feng, J.H. Sui, Z.Y. Gao, G.F. Dong, *Scr. Mater.* **58**, 830 (2008).

[10] K. Srinivas, M.M. Raja, S.V. Kamat, *J. Alloys Comp.* **619**, 177 (2015).

[11] K. Kabra, N. Lakshmi, K. Venugopalan, *AIP Conf. Proc.* **1536**, 1031 (2013).