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ABSTRACT

In this article, we studied the effect of annealing (600 °C for 1 h) and the applied magnetic field from 50 Oe to 20 kOe of Co_2FeSi glass-coated microwires with ordered $L2_1$ structure prepared by Taylor–Ulitovsky technique on the magnetic behavior. The as-prepared and annealed samples show a ferromagnetic behavior at the range of measuring temperature (5 to 400 K) and magnetic field (50 Oe to 20 kOe). M–H loops of as prepared sample do not show a squared shape. Meanwhile, perfectly squared hysteresis loops have detected for the annealed sample. In addition, annealed sample shows high magnetization M/M_{5K} ratio, coercivity, and anisotropy field, as-compared to the as-prepared one. The annealed sample shows considerable irreversibility when the magnetic behavior changes with temperature upon the applied magnetic field at 50 and 200 Oe. Such irreversibility does not found in the as-prepared sample measured at the same magnetic field due to mixed amorphous and crystalline structure. By increasing the external magnetic field higher than 200 Oe and up to 20 kOe a gradual changing in the magnetic behavior has been detected where the irreversibility disappeared at applying magnetic field about 1 kOe and the magnetic behavior is totally change by increasing the external magnetic field up to the maximum 20 kOe. The difference in the magnetic behavior of the annealed glass-coated Co_2FeSi glass-coated microwires indicates the effect of internal stresses induced by the presence of the glass-coating and the annealing-induced recrystallization.

I. INTRODUCTION

Co_2-based Heusler alloys, (Co_2FeSi) in particular, are considering promising smart materials for multifunction applications, especially in the spintronics, due to unique physical properties such as extraordinary electronic structure, high Curie point, near to 100% spin polarization at the Fermi level position, high saturation magnetization, very low Gilbert damping constant (α = 0.004) and unusual anomalous Hall Effect.[1–5] Up until now, Co_2FeSi Heusler alloys have been used to design superconductor (SC)-based devices,[6] ultra-high-performance giant magnetoresistance devices,[7] semiconductor-based spintronic devices[8] and magnetic tunnel junctions.[9]

Due to its unique combination of (electronic, electrical, mechanical, magnetic, and anticorrosive) properties, Co_2-based Half-Metallic Heusler alloys made using the Taylor–Ulitovsky technique provide significant advantages over conventional Co_2-based Heusler alloys.[10–13] Additionally, the tunable microstructure of the microwires prepared by the Taylor–Ulitovsky method has led to the fabrication of Heusler-based glass-coated microwires with a variety of microstructures, including amorphous, poly/monocrystalline, and granular structures. The growing interest in this method has increased significantly in last decades due to their technological applications, especially in sensing and biomedical applications.[14–19] In addition, the Taylor–Ulitovsky provide a simple, fast (up to a few hundred meters per minute) and low cost preparation technique to fabricate thin\thick Heusler-based glass-coated microwires alloys without the necessity of additional long thermal treatments.[14,15] One of the advantages of the Taylor–Ulitovsky technique is the ability to produce microwires with metallic nucleus with a wide range of diameters (from 200 nm to 100 μm) and several kilometers long.[19,20] Moreover, the glass coating add additional flexibility to prevent metallic core nuclei from the oxidation and enhance the mechanical and biocompatibility properties.[11,19,21,22] Therefore, Co_2FeSi Heusler alloy glass coated microwires are expected to be useful in a variety of applications.

In current study, we will focus on the magnetic behavior of as-prepared and annealed (600 °C for 1 h) of Co_2FeSi glass-coated microwires under a wide range to temperature (5 to 400 K) and magnetic field (50 Oe to 20 kOe). Different magnetic behavior has observed depending on the external applied magnetic field and the temperature. The annealed sample show gradual uniform magnetic dependence by increasing the applied magnetic field.

II. MATERIAL AND METHODS

Starting from melting high purities of Co (99.99%), Fe (99.99%) and Si (99.99%) with aspect ratio (2:1:1) using conventional arc furnace in argon atmosphere to prevent oxide formation during melting process. The melting process repeated for five time to obtain high homogeny alloy. Then we checked the chemical composition using the EDX/SEM, as reported in our previous work.[12,15,23] After confirming the chemical composition, we proceeded to fabricate the glass-coated microwires by using Taylor–Ulitovsky method. For more details about the method of glass-coated microwire fabrication was reported and described elsewhere.[14,15,23–25] The inner metallic nucleus diameter, d, of Co_2FeSi glass-coated microwires has final geometric parameters of 4.4 μm, while the total diameter of the exterior Pyrex coating, D, is around 17.6 μm. After fabrication, the Co_2FeSi microwire was subsequently annealed at 600 °C for 1 h in a protective helium atmosphere. The structural analysis and EDX/SEM evaluation of the metallic nucleus' chemical composition were previously examined and described elsewhere.[11–13] The magnetic properties were studied by using PPMS (Physical Property Magnetic System, Quantum Design Inc., San Diego, CA) at temperatures, T, between 5 and 400 K and wide range of applied magnetic field (H = 50 Oe to 20 kOe). The results are provided in terms of the normalized magnetization, M/M_{5K}, where M_{5K} is the magnetic moment measured at 5 K with a magnetic field equal to 20 kOe. The microwire bunch was employed for magnetic measurements revealing relative variations of magnetization.

III. RESULTS AND DISCUSSION

Figure 1 shows the axial hysteresis loops of Co_2FeSi as-prepared and annealed glass-coated microwires, measured by the PPMS with an applied magnetic field of 20 kOe parallel to the microwires' axis and a temperature range of 300 to 5 K. Every loop was adjusted to the

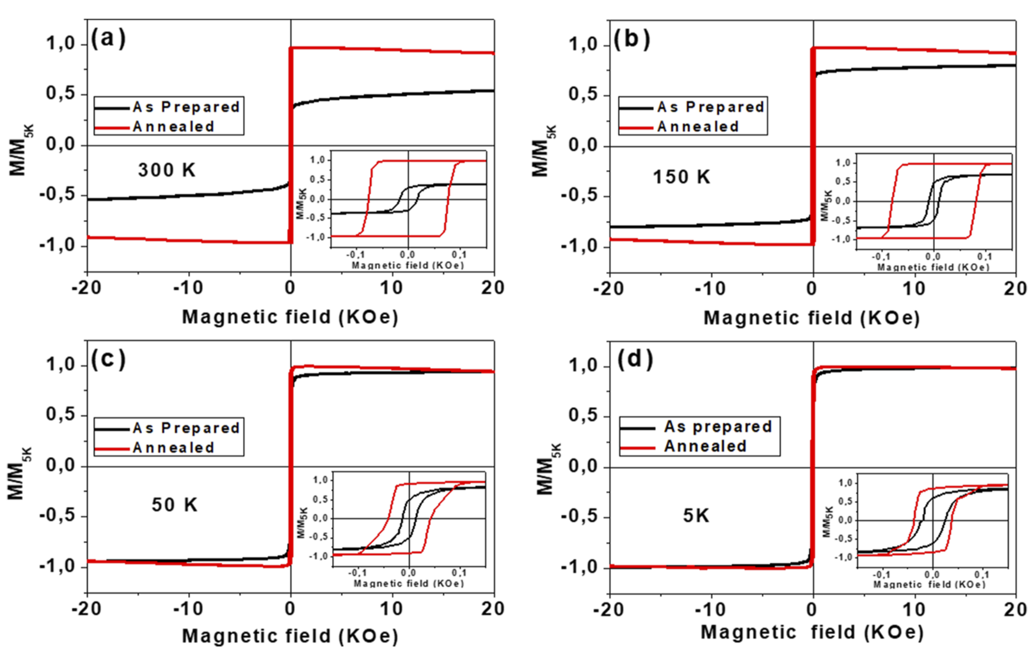

FIG. 1. Magnetization curves of as-prepared and annealed Co_2FeSi glass-coated microwires measured at maximum field 20 kOe measured at 300 K, (a) 150 K, (b) 50 K, (c) and 5 K, (d) M/M_{5K} (T) magnetic curves dependencies measured at low field are presented in the inset.

maximum magnetic moment measured at 5 K. As shown in Fig. 1, all annealed samples show perfectly squared hysteresis loops with higher coercivity and M/M_{5K} ratio as-compared to the as-prepared sample. The M/M_{5K} ratio show a notable increase by decreasing the temperature from 300 to 5 K. The coercivity increases as the temperature decreases from 300 K, reaching a maximum at 150 K, and then starts to drop by decreasing the temperature to 5 K. This is an intriguing finding in magnetic M–H loops of annealed material. In contrast, the M/M_{5K} ratio for the annealed sample just slightly changes with temperature, while the as-prepared sample shows a significant variation.

Figure 2 summarized the temperature dependence of magnetization, M/M_{5K}, of the as-prepared sample. In Fig. 1 is illustrated that the samples exhibit ferromagnetic behavior over the whole measurement temperature range of 5 to 400 K. Additionally, when the temperature decreases, the magnetization rises to its maximum at 5 K. A more substantial magnetization dependence was found for field cooling (FC) and field heating (FH) protocols when an applied magnetic field is 50 Oe. Additionally, there is a discrepancy between FC and FH magnetization curves. This mismatching can be attributed to the variation of the internal stresses in glass-coated microwires stress with temperature and the applied external magnetic field. Additionally, the magnetic field dependence does not exhibit a consistent trend with temperature; this is because disordered crystalline phases coexist with ordered ones in the same sample. As reported recently by the XRD analysis, as-prepared Co_2FeSi glass-coated microwires show a mixed structure (amorphous + crystalline [with $L2_1$ (ordered) and B2 (disordered)].[10–12] An increase of average grain size (from 18 to 30 nm) and content of crystalline phase (from 50 to almost 100%) was observed upon annealing of Co_2FeSi glass-coated microwires.[11,12] Softer magnetic properties and the non-squared hysteresis loops shape (illustrated in Fig. 1) can be attributed to the contribution of an amorphous precursor and non-uniform magnetic behavior with variation the magnetic field and temperature (seen in Fig. 2). Such substantial dependence of the hysteresis loops on the average grain size and

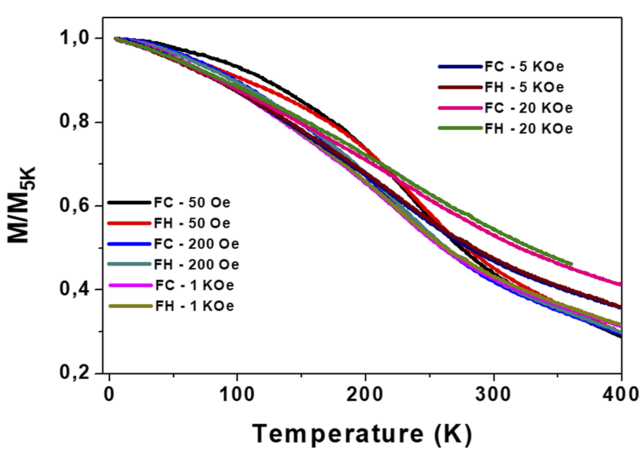

FIG. 2. Measured temperature dependence of magnetization for as-prepared Co_2FeSi glass-coated microwires with 50 Oe to 20 kOe of applied external magnetic field.

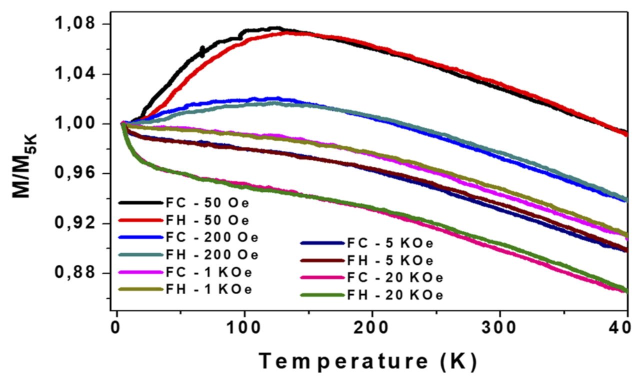

FIG. 3. Temperature dependence of magnetization measured for annealed Co_2FeSi glass-coated microwires with applied external magnetic field 50 Oe to 20 kOe.

the crystalline phase content is reported for nanocrystalline materials and, in particular, for magnetic microwires with nanocrystalline structure.[26–28]

The M/M_{5K} vs (T) dependence of Co_2FeSi glass-coated microwires annealed for 1 h at 600 °C is completely different from that of the as-prepared sample. Several anomalous features have been observed in the temperature and magnetic field dependencies. The magnetic phase transition with considerable irreversibility has been demonstrated with a blocking temperature of 150 K in the M/M_{5K} vs (T) dependence with applied low magnetic field, i.e. 50 and 200 Oe. Such behavior does not appear in as-prepared sample when the same magnetic field is applied. This difference (see Figs. 2 and 3) can be explained by the mixed amorphous/crystalline structure of as-prepared sample, as recently reported elsewhere.[11,12] The irreversible magnetic behavior vanished when the applied magnetic field is above 200 Oe. In this case the M/M_{5K} vs (T) curves exhibit similar to the as-prepared Co_2FeSi glass-coated microwire behavior. The most intriguing aspect is that the annealed Co_2FeSi glass-coated microwire becomes extremely sensitive to temperature and the applied magnetic field (see Fig. 3). The anomalous magnetic behavior of annealed Co_2FeSi is due to the annealing-induced recrystallization process, which is accompanied by an increase in crystalline phase content, atomic order and a decrease in internal stresses. Additionally, this process induces two different magnetic phases (the martensitic phase), each of, which has a different magnetic response.[11,12]

IV. CONCLUSION

In conclusion, we illustrate the effect of annealing on hysteresis loops and on the magnetic field and temperature effect on the magnetic properties of Co_2FeSi glass-coated microwires with nanocrystalline structure where $L2_1$ is the dominant structure order. Annealing leads to a change in the magnetic properties of Co_2FeSi glass coated microwires, such as the appearance of a perfectly squared hysteresis loop shape with M/M_{5K} close to unity. In addition, an improvement in the magnetic response from non-uniform for as-prepared sample to a uniform response to an applied magnetic field for an annealed sample is observed. At low field, the annealed sample, shows a martensitic phase transition with a large irreversible magnetic behavior, while this magnetic transition did not appear in the as-prepared sample. The result obtained confirms the strong effect of annealing on the magnetic properties and their temperature and magnetic field dependence of Co_2FeSi glass-coated microwires.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

M. Salaheldeen: Data curation (equal); Formal analysis (equal); Investigation (equal); Validation (equal); Writing – original draft (equal). **M. Ipatov**: Data curation (equal); Formal analysis (equal); Methodology (equal); Validation (equal). **V. Zhukova**: Data curation (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Validation (equal). **A. García-Gomez**: Data curation (equal); Formal analysis (equal); Investigation (equal). **J. Gonzalez**: Funding acquisition (equal); Project administration (equal); Resources (equal). **A. Zhukov**: Conceptualization (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Project administration (equal); Resources (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

REFERENCES

[1] I. Galanakis, P. H. Dederichs, and N. Papanikolaou, Phys. Rev. B **66**(17), 174429 (2002).
[2] B. K. Hazra, S. N. Kaul, S. Srinath, and M. M. Raja, J. Phys. D: Appl. Phys. **52**(32), 325002 (2019).
[3] S. Wurmehl, G. H. Fecher, H. C. Kandpal, V. Ksenofontov, C. Felser, H. J. Morais, and J. Morais, Phys. Rev. B **72**(18), 184434 (2005).
[4] P. Li, J. Koo, W. Ning, J. Li, L. Miao, L. Min, Y. Zhu, Y. Wang, N. Alem, C.-X. Liu, Z. Mao, and B. Yan, Nat. Commun. **11**(1), 1 (2020).
[5] I. Belopolski, K. Manna, D. S. Sanchez, G. Chang, B. Ernst, J. Yin, S. S. Zhang, T. Cochran, N. Shumiya, H. Zheng, B. Singh, G. Bian, D. Multer, M. Litskevich, X. Zhou, S.-M. Huang, B. Wang, T.-R. Chang, S.-Y. Xu, A. Bansil, C. Felser, H. Lin, and M. Z. Hasan, Science **365**(6459), 1278 (2019).
[6] Y. Fujita, T. Sasaki, and Y. Sakuraba, Thin Solid Films **745**, 139084 (2022).
[7] B. Büker, J. W. Jung, T. Sasaki, Y. Sakuraba, Y. Miura, T. Nakatani, A. Hütten, and K. Hono, Phys. Rev. B **103**(14), L140405 (2021).
[8] M. Yamada, F. Kuroda, M. Tsukahara, S. Yamada, T. Fukushima, K. Sawano, T. Oguchi and K. Hamaya *et al.*, NPG Asia Mater. **12**, 47 (2020).
[9] I. Shigeta, T. Kubota, Y. Sakuraba, C. G. Molenaar, J. N. Beukers, S. Kimura, A. A. Golubov, A. Brinkman, S. Awaji, K. Takanashi, and M. Hiroi, Appl. Phys. Lett. **112**(7), 072402 (2018).
[10] M. Salaheldeen, A. Garcia-Gomez, M. Ipatov, P. Corte-Leon, V. Zhukova, J. M. Blanco, and A. Zhukov, Chemosensors **10**(6), 225 (2022).
[11] M. Salaheldeen, A. Garcia-Gomez, P. Corte-Leon, M. Ipatov, V. Zhukova, J. Gonzalez, and A. Zhukov, J. Alloys Compound **923**, 166379 (2022).
[12] M. Salaheldeen, A. Garcia, P. Corte-Leon, M. Ipatov, V. Zhukova, and A. Zhukov, J. Mater. Research Technol. **20**, 4161 (2022).
[13] M. Salaheldeen, A. Talaat, M. Ipatov, V. Zhukova, and A. Zhukov, Processes **10**, 2248 (2022).
[14] A. Zhukov, P. Corte-Leon, L. Gonzalez-Legarreta, M. Ipatov, J. M. Blanco, A. Gonzalez, and V. Zhukova, J. Phys. D: Appl. Phys. **55**, 253003 (2022).
[15] A. Zhukov, M. Ipatov, A. Talaat, J. Blanco, B. Hernando, L. Gonzalez-Legarreta, J. Suñol, and V. Zhukova, Crystals **7**, 41 (2017).
[16] D. Kozejova, L. Fecova, P. Klein, R. Sabol, R. Hudak, I. Sulla, D. Mudronova, J. Galik, and R. Varga, J. Magn. Magn. Mater. **470**, 2 (2019).
[17] V. Zhukova, P. Corte-Leon, M. Ipatov, J. M. Blanco, L. Gonzalez-Legarreta, and A. Zhukov, Sensors **19**, 4767 (2019).
[18] V. Zhukova, P. Corte-Leon, J. M. Blanco, M. Ipatov, J. Gonzalez, and A. Zhukov, Chemosensors **9**(5), 100 (2021).
[19] A. Talaat, J. Alonso, V. Zhukova, E. Garaio, J. A. García, H. Srikanth, M. H. Phan, and A. Zhukov, Sci. Rep. **6**, 39300 (2016).
[20] H. Chiriac, N. Lupu, G. Stoian, G. Ababei, S. Corodeanu, and T.-A. Óvári, Crystals **7**, 48 (2017).
[21] O. Mitxelena-Iribarren, J. Campisi, I. Martínez de Apellániz, S. Lizarbe-Sancha, S. Arana, V. Zhukova, M. Mujika, and A. Zhukov, Mater. Sci. Eng. C **106**, 110261 (2020).
[22] V. Zhukova, A. F. Cobeño, A. Zhukov, A. R. de Arellano Lopez, S. López-Pombero, J. M. Blanco, V. Larin, and J. Gonzalez, J. Magn. Magn. Mater. **249**(P1-II), 19 (2002).
[23] S. Baranov, V. Larin, and A. Torcunov, Crystals **7**, 136 (2017).
[24] H. Chiriac and T. A. Óvári, Prog. Mater. Sci. **40**, 333–407 (1996).
[25] V. Zhukova, P. Corte-Leon, J. M. Blanco, M. Ipatov, L. Gonzalez-Legarreta, A. Gonzalez, and A. Zhukov, Chemosensors **10**(1), 26 (2022).
[26] G. Herzer, IEEE Trans. Magn. **26**, 1397 (1990).
[27] T. Kulik, J. Non. Cryst. Solids. **287**, 145 (2001).
[28] A. P. Zhukov, A. Talaat, M. Ipatov, J. M. Blanco, L. Gonzalez-Legarreta, B. Hernando, and V. Zhukova, IEEE Trans. Magn. **50**(6), 2501905 (2014).