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Anomalous magnetic behavior in half-metallic Heusler Co_2FeSi alloy glass-coated microwires with high Curie temperature

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In the current study, we have succeeded to fabricate Co_2FeSi Heusler alloy glass-covered microwires with magnetic core nucleus diameter d = 4.36 µm and total diameter D = 17.55 µm, with high Curie temperature (T_c > 1100 K) and well-defined magnetic anisotropy for high temperature spintronic devices application. The magnetic properties of as-prepared and annealed at different temperature (873 K, 973 K and 1073 K for 1 h) of Co_2FeSi Heusler alloy glass-covered microwires have been investigated. Strong dependence of the magnetic properties on the annealing conditions has been indicated. Anomalous magnetic behavior for annealed samples at 873 K and 973 K has been found and structural properties of such samples have been analyzed. Critical temperatures 155 K and 250 K have been detected for annealed samples at 873 K and 973 K, respectively, where the behavior of M-H loops and coercivity changed. Below the critical point the M-H curve shows "kink or wasp-waisted" magnetic behavior and complex magnetic reversal mechanism is supposed. The anomalous magnetic behavior is due to the martensitic phases induced by annealing conditions below the room temperature. This unusual magnetization behavior provides opportunities to understand the phenomena of different types of magnetic domain structures in the preferred crystallographically oriented Co_2FeSi Heusler alloy glass-coated micro-wires, essentially helpful for designing the devices based on magnetization switching.

1. Introduction

Spintronics is one of the most promising interdisciplinary research areas, allowing the development of next-generation nanodevices to reduce power consumption and enhance their processing and memory performance [1]. A new generation of materials with multifunction applications should be developed to meet the different kinds of demands, such as high spin polarization or high Curie temperature, T_c. One of the promising smart materials with suitable properties for spintronic and magneto-electronic applications are the Heusler compounds [2]. Among their advantages are the perfect lattice matching with major substrates, high T_c above the room temperature and intermetallic controllability for spin density of states at the Fermi energy level, where almost 100% of spin polarized near the Fermi level is reported [2–7].

Among half-metallic alloys, Co_2-based full-Heusler compounds are the most promising materials due to their electric structure based on ab initio calculations [8], exotic transport properties, high thermal stability, high Curie temperatures (T_c ≈ 1100 K) for the bulk sample, high magnetic moment (~ 6 μ_B/f.u.) and low Gilbert damping constant (α = 0.004) [9,10]. Additionally, Co_2-based Heusler compounds are of current interest, because of their large anomalous Hall effect due to the large Berry curvature linked with their band structure [11,12]. For these reasons, Co-based full-Heusler alloys are attracting the attention of the scientific community. Therefore, these kinds of alloys are widely investigated in different structures such as ribbons [13,14], nanoparticles [15–17], thin films [18–20], and nano/micro wires [21]. It is worth noting, that for the application purpose, the fabrication of Heusler alloys in nanoparticles and thin films forms are facing many challenges such as, the high cost of preparation techniques, inhomogeneity of the chemical composition,

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the easy oxidation from the perspective of proper atomic ordering and material chemistry [22]. In addition to the lattice mismatch between the alloy and the substrate, diffusion of substrate atoms into the film leads to the presence of atomic disorder and phase separations which are frequently encountered [23]. Moreover, the Heusler alloys prepared by arc melting or grown as thin films require long time and high temperature annealing treatments to initiate the necessary structural ordering [24]. To avoid these disadvantageous steps, alternative rapid quenching production of Heusler alloys has been recently carried out [25,26].

Rapid melt quenching process allows easy, cheap and fast fabrication of amorphous, nanocrystalline or metastable crystalline materials with planar (ribbons) or cylindrical (micro wires) geometries [27,28]. There are several fabrication methods involving rapid melt quenching allowing the preparation of magnetic wires in a wide range of diameters: the thickest wires with diameters, *d*, ranging between 60 and 320 µm can be fabricated by the "in-rotating water" method known since the 80 s [29,30]. The Taylor-Ulitovsky technique, was known since 60 s allows the preparation of thinner (typically 0.5–40 µm in diameter) cast metallic wires coated by flexible glass. Among the advantages of this technique, perhaps the main one is that it allows fast (up to a few hundred meters per minute) production of quite long (up to few kilometers) and thin microwires. However, the metallic nucleus diameter can be as large as 100 µm [27,31,32]. In addition to the enhanced mechanical properties and fast and inexpensive fabrication method, the existence of thin, flexible, insulating, and highly transparent glass coating, can be useful for biomedical applications due to its biocompatibility [31]. Therefore, the Co_2-based Heusler glass-covered microwires represent a promising smart metamaterial for a multi-function application for a new generation of spintronics. As far as we know, there is a lack of studies focusing on the fabrication, structural, mechanical and magnetic characterization of Co_2-based Heusler glass-covered micro wires. Thus, in current study we will focus on the structural and the magnetic properties of Co_2FeSi glass-covered microwires to illustrate its possible applications in advanced spintronics.

In our previous work we have been successful in fabricating a long Co_2FeSi microwire (~1 km) with the metallic nucleus diameter of about 4 µm, characterized by well oriented monocrystalline structure (*B2* phase, with the lattice parameter a = 5.615 Å) and T_c higher than 1000 K [21]. In the given contribution, we present the structural and magnetic characterization of glass-covered Heusler Co_2FeSi alloy micro wires prepared by Taylor-Utilovsky technique. The effect of temperature and annealing conditions has been investigated. The magnetic behavior of Co_2FeSi strongly depends on the temperature, magnetic field, and the annealing conditions. We illustrate that the annealing conditions induce different magnetic phases with anomalous magnetic behavior that does not exist in the as-prepared samples. We show that it is possible to produce up to kilometers of monocrystalline Co_2FeSi Heusler-based micro wires in few minutes using this simple production technique and tune the magnetic properties of microwire by annealing conditions that may enhance the suitability of Co_2FeSi for spintronic applications.

2. Materials and methods

The ternary metal alloy with nominal composition of $Co_{50}Fe_{25}Si_{25}$ was prepared by arc melting from high purity metals (Co, Fe and Si) > 99.9% in argon atmosphere to prevent oxide formation during melting. The alloys were re-melted several times to improve homogeneity. Magnetic glass-coated microwires were fabricated by the Taylor-Ulitovsky technique, which consists on drawing and casting directly from the melted $Co_{50}Fe_{25}Si_{25}$ alloy [26,32–37]. The diameter of inner metallic nucleus of microwire sample is around 4.36 µm, while the diameter of external Pyrex coating is around 17.55 µm. After preparation, the Co_2FeSi microwire was subsequently thermally annealed at different temperatures 873 K, 973 K and 1073 K for (1 h) in a protective helium atmosphere.

The chemical composition of the metallic nucleus evaluated using Energy dispersive X-ray, EDX, spectroscopy was quite homogeneous indicating almost the same composition in different parts of the microwire. From the EDX data (not shown), we determined that the chemical composition of the metallic nucleus is $Co_{50}Fe_{25}Si_{25}$. The composition of the metallic nucleus is in accordance with the stoichiometric one Co_2FeSi.

Structure and phase composition have been studied using a BRUKER (D8 Advance, Bruker AXS GmbH, Karlsruhe, Germany) X-ray diffractometer. Cu K_α (λ = 1.54 Å) radiation was used in all the patterns. The magnetization curves were measured using PPMS (Physical Property Magnetic System, Quantum Design Inc., San Diego, CA) vibrating-sample magnetometer at temperatures, T, between 5 and 400 K. A magnetic field, H, from 50 Oe to 50 kOe was directed along the sample axis.

3. Results and discussion

Fig. 1 indicates the magnetic hysteresis loops of as prepared Co_2FeSi microwires measured in applied magnetic field between ± 50 kOe and low scale magnetic field, in Fig. 1a and b, respectively, at

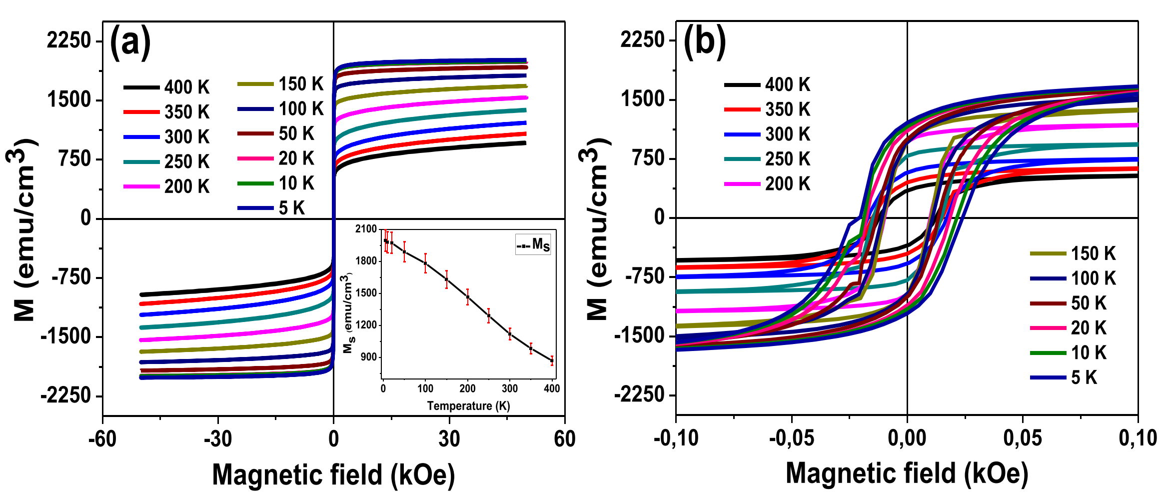

Fig. 1. Hysteresis loops, measured in magnetic field applied parallel to the axis of micro wires in the temperature range from 5 to 400 K for as prepared Co_2FeSi micro wires measured magnetic field (a) 50 kOe and (b) hysteresis loops with low scale magnetic field. The inset of (a) illustrates saturation magnetization values with temperature.

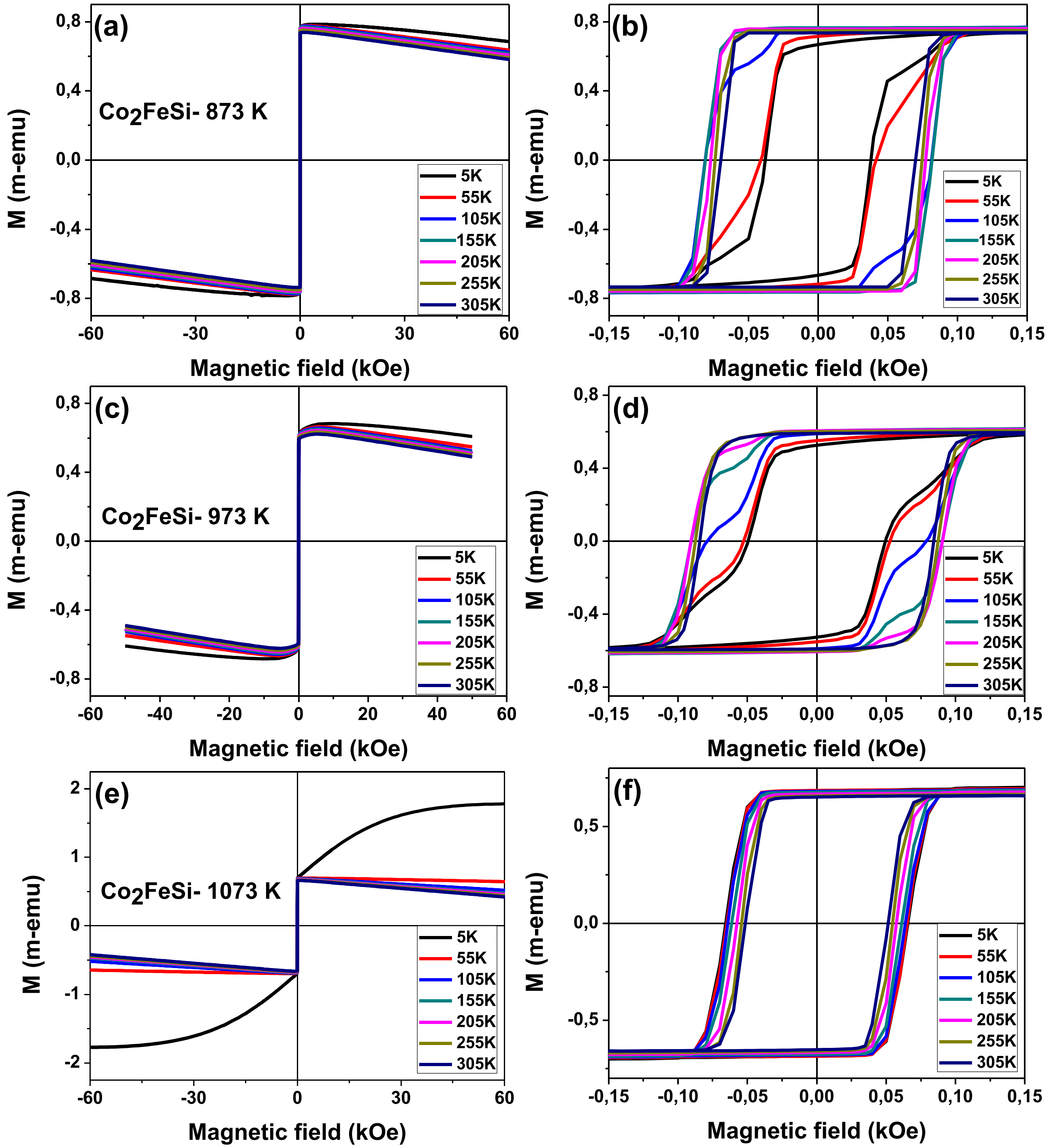

Fig. 2. Hysteresis loops, measured in magnetic field applied parallel to the axis of microwires in the temperature range from 5 to 305 K for annealed Co_2FeSi micro wires at different temperature for one hour (a, b) at 873 K, (c, d) at 973 K and (e, f) at 1073 K.

temperature range between 5 and 400 K. As plotted in Fig. 1a and b, all hysteresis loops show ferromagnetic behavior over the entire measuring temperature range, where the magnetic parameters (saturation magnetization, M_s, magnetic remanence, M_r, and coercivity, H_c) increase with decreasing the temperature. The maximum values of the magnetic parameters are detected at 5 K and the lowest values are observed at 400 K. Also, all loops show regular M-H loops shape such as has been observed in similar Co_2FeSi alloys deposited by different techniques and in different forms [9,19,21,22,38]. For annealed Co_2FeSi microwires at different temperatures many differences have been observed. First, the M-H hoops appear wider and squared for all Co_2FeSi annealed samples at different temperature as shown in Fig. 2. Secondly, H_c within the measuring temperature range for annealed samples shows irregular magnetic behavior compared to as prepared one. Finally, irregular magnetic loops shapes have been observed at low temperature for Co_2FeSi annealed samples at 873 K and 973 K. This indicates changes in the micro magnetic structures of annealed samples compared to the as prepared sample. All these observations will be discussed in next paragraphs.

Fig. 2 shows M-H loops of annealed Co_2FeSi microwires with different annealing temperature and the temperature variation from

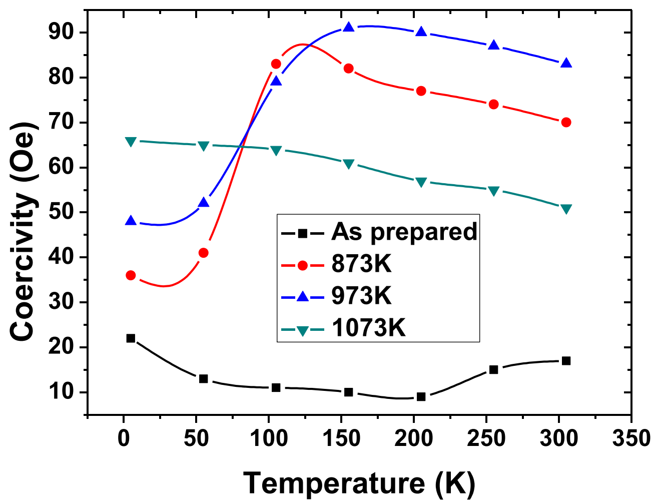

Fig. 3. Temperature dependence of the coercivity for Co_2FeSi micro wire for as prepared and annealed with different temperatures samples (lines for eye guide).

305 K to 5 K. All the annealed Co_2FeSi samples show ferromagnetic behavior below/at/above the room temperature. Above the room temperature the loops show regular squared loops with slight decrease of H_c, M_s and M_r with increasing the temperature i.e., the annealed samples follow the same tendency of the magnetic parameters of as prepared Co_2FeSi samples with the temperature.

For Co_2FeSi microwires annealed at 873 K and 973 K a drastic change of the magnetic behavior has been observed. Although all annealed microwire samples show ferromagnetic behavior for all range of the measuring temperature as illustrated in Fig. 2a–d, anomalous magnetic features have been detected for these specific annealed samples below the room temperature. A critical temperature has been detected where the distortion in the magnetic hysteresis loops starts to appear at 105 K and 255 K for annealed Co_2FeSi microwires at 873 K and 973 K, respectively. Above these critical temperatures the magnetic hysteresis loops show regular shape with one step magnetic behavior and below these critical temperatures the loops show "kink or wasp-waisted" magnetic behavior with multi-step magnetic switching. Actually, this kind of magnetic behavior has been observed and reported for various magnetic structure forms such as magnetic nanoparticles, nanostructured thin films and amorphous magnetic microwires, as reported elsewhere [33,39–44]. To our knowledge, this magnetic behavior is first time observed in Co_2FeSi alloy glass-covered microwires. Many reasons have been reported and discussed to describe such magnetic behavior. Among several origins of multi-step hysteresis loops, the effect of the oxidation of the soft part of the alloys due to the strong magnetic coupling between the two different magnetic phases, in our case Co (hard) phase and Fe (soft) phase, with different coercive field or reordering of ferromagnetic spins below the critical temperature under the influence of the applied magnetic field were discussed [42]. These reorientations of spins are responsible for the kink in the hysteresis loops, due to the pinning of domain walls motion in the direction of the spin order [45]. In addition, this type of anomalous magnetic behavior may appear because of a mixture of the grain boundaries and combination of soft/hard magnetic phases having different magnetic features [46]. Similarly, the multi-step hysteresis loops have been observed in magnetic microwires with mixed amorphous-nanocrystalline structure [33,46]. In this case, one of the magnetic phases presents soft magnetic behavior, while the second phase is magnetically harder. Moreover, the alternative reason for the multi-step magnetic behavior for the magnetic glass-covered microwires is related to the magnetostatic interaction in the linear microwires array consisting of the superposition effect of an external magnetic field and the stray field from the magnetic microwires linear array, as reported and explained elsewhere [32,47]. In the single microwire the magnetostatic interaction can be originated by the fluctuations of the metallic nucleus diameter, internal stresses, stresses arising during cutting of the samples, etc. We think that the strength of the magnetostatic interaction is affected by the difference between the two magnetic fields, as increases with the increase of the difference between the stray field and switching field and vice versa. For the effective superposition the single microwire behave like two magnetic microwires as a result of the large difference between the two fields below the critical temperature and resulting in the multistep magnetic behavior.

The more interesting part in the current results is the fact that the multi-step behavior of Co_2FeSi microwire does not appear in as-prepared samples. It only appears in the annealed samples at 873 K and 973 K and then disappears for annealed sample at 1073 K. Therefore, we believe that the annealing temperature induces two magnetic phases that have a different magnetic behavior and it is strongly related to the critical temperature where the multi-step magnetic behavior has been occurred. Consequently, below the critical temperature the coercive field of each magnetic phase taken separately may be different. After switching of one of the phases, superposition of the demagnetizing field of the already re-magnetized another phase and of the external magnetic field is insufficient to switch the magnetization of second phase with the larger moment. Moreover, the coexistence of 50% Co (hard) and 25% Fe (soft) phases in the annealed micro wires samples are the main reasons for the "kink or wasp-waisted" hysteresis loops and multi-step magnetic behavior.

While analyzing the magnetic hysteresis loops of as-prepared and annealed Co_2FeSi samples, an anomalous H_c tendency with temperature has been observed, as plotted in Fig. 3. All annealed samples show higher coercive field H_c as compare to the as- prepared one. The as-prepared sample shows quite soft magnetic behavior with slight change of H_c with temperature as the difference between the value of coercivity at 5 K (H_c = 21 Oe) and 305 K (H_c = 18 Oe) is about 3 Oe. Meanwhile, for samples annealed at 873 K and 973 K, an anomalous tendency in the coercivity has been detected: H_c starts to increase with decreasing the temperature and reaches the maximum value at 105 K and 155 K for the samples annealed at 873 K and 973 K, respectively. Then, H_c starts to decrease with decrease in the temperature and reaches the lowest value at 5 K (see Fig. 3). This anomalous behavior of the H_c disappears for sample annealed at 1073 K as indicated in the Fig. 3. It is noteworthy that, the anomalous behavior of H_c for annealed samples at 873 K and 973 K is well matched with the critical temperature where the multi-steps magnetic behavior has been observed (see Fig. 4a and b): regular ferromagnetic behavior has been observed for temperature higher than the critical temperature and multi-step magnetic behavior has been detected for hysteresis loops measured below the critical temperature. As known for crystalline and polycrystalline ferromagnetic materials the decreasing of the temperature increases the magnetic anisotropy field i.e., increases H_c. The changing in the H_c tendency at low temperature is strongly related to the change in the micromagnetic structure which is induced by the transformation in the magnetic phases as previously explained at Fig. 2.

It is important to understand that the thermal stability of the ferromagnetic materials is extremely important concerning its potential use in spintronic devices in order to operate at below or above the room temperature. Therefore, we have performed the temperature dependence of magnetization (M vs. T) i.e., field cooling, FC, and field heating, FH, at different applied magnetic field (50 Oe to 1 kOe) and temperature range from 5 K to 400 K.

Fig. 5 represents the temperature dependence of the magnetization of Co_2FeSi micro wires. As indicated in Fig. 5a the as-prepared sample shows a strong ferromagnetic behavior for all range of

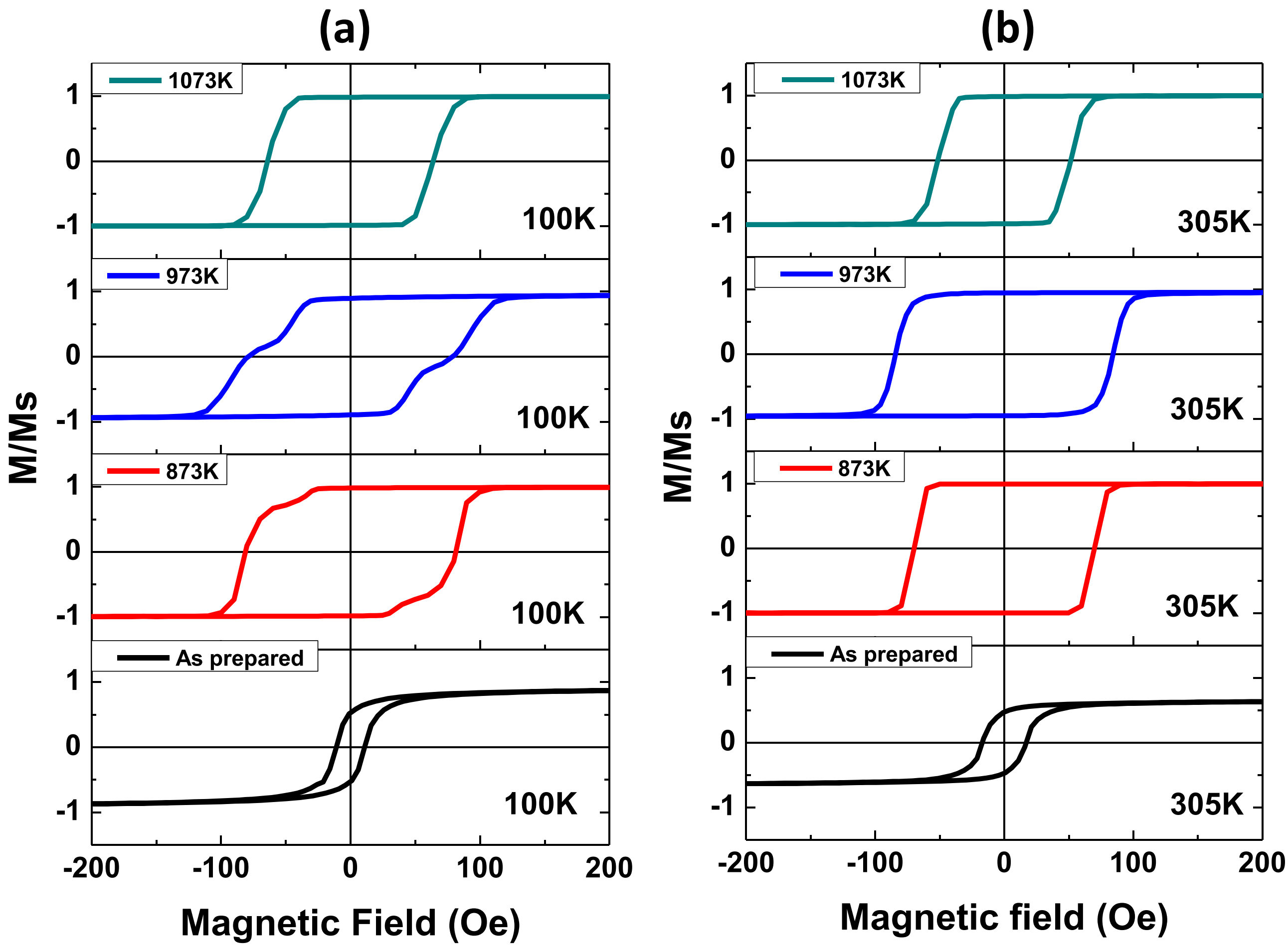

Fig. 4. Hysteresis loops, measured in magnetic field applied parallel to the axis of micro wires (a) at 100 K and (b) at 305 K for as prepared and annealed for one hour at 873 K, 973 K and 1073 K Co_2FeSi microwires.

temperature. By decreasing/increasing the temperature the magnetization increase/decrease reaches maximum/minimum values at 5 K/400 K for each applied magnetic field i.e., typical behavior of ferromagnetic materials with temperature. This behavior of the M vs. T explains the regular magnetic behavior of the H_c and M-H loops at different temperature for as prepared Co_2FeSi micro wire sample. In addition, from the M vs T curve we were not be able to directly measure T_c of the as prepared sample because it is expected to be (> 1100 K) as reported elsewhere [38], and this temperature is out of our PPMS temperature range (the maximum T is 400 K). Meanwhile, for annealed samples at 873 K and 973 K magnetic phase transition has been observed, as evidenced by a sharp drop of the magnetic moment when we applied 50 Oe and 200 Oe external magnetic fields during FC and FH as seen in Fig. 5b and c. The more interesting of this sharp drop is its disappearance when applied high magnetic field of 1 kOe for both annealed Co_2FeSi samples at 873 K and 973 K. These magnetic phases which appear in the annealed samples at 873 K and 973 K related to the anomalous magnetic behavior have been described in Figs. 2–4. The blocking temperature, T_B, is found at 150 K and 205 K for the annealed samples at 873 K and 973 K, respectively. Therefore, there is an excellent match with T_B and the critical temperature as we supposed above for annealed samples at 873 K and 973 K. The as-prepared sample and sample annealed at 1073 K do not show any kind of irreversibility behavior (see Fig. 5a and d). Thus, Co_2FeSi Heusler alloys are sensitive to the annealing conditions which affect the lattice structure as well as the local atomic environment and also their stoichiometric composition [22]. In general, the anomalies in M vs T for magnetic Heusler alloys have been explained by considering three main phases transition temperatures, Curie Temperature of martensitic (T_{CM}) phase, temperature of martensitic transition, T_M, and Curie temperature of the austenitic cubic phase at high temperature [48–51]. In the present case the temperature of the martensitic transition is more favorable, since for samples annealed at 873 K and 973 K T_c > 1100 K i.e., T_c > > T_M. Moreover, the range of the T_M is matched with the critical temperature that has been described where the “kink or wasp-waisted” hysteresis loops and multi-step magnetic behavior appear. We believe that the annealing temperature at 873 K and 973 K induces a recrystallization process accompanied by the atomic order and the reduction of internal stresses, moreover, induces two different magnetic phases. The martensitic phase with different magnetic response is responsible for the anomalous magnetic behavior of annealed Co_2FeSi.

In order to study the evolution of the structure of our samples upon annealing, X-ray powder diffraction measurements where made as can be seen in Fig. 6. These measurements allowed us to bring some light in to the anomalous behavior observed in the hysteresis loops. All the spectra present a broadened peak centered around 2θ ≈ 22.5º that corresponds to the amorphous glass coating, as we expected due to the Taylor-Ulitovsky method. Along with the amorphous pattern, the as-prepared sample presents another peak at 2θ ≈ 45.6º.

Phase identification of the studied samples by a single and broad crystalline peak observed in as-prepared and annealed samples (see Fig. 6) seems to be problematic. Several phases (cubic Fe_3C, $Fe_{1.34}Si_{0.66}$, Co_2FeSi as well as $Fe_3Co_3Si_2$) for the XRD peak at 2θ

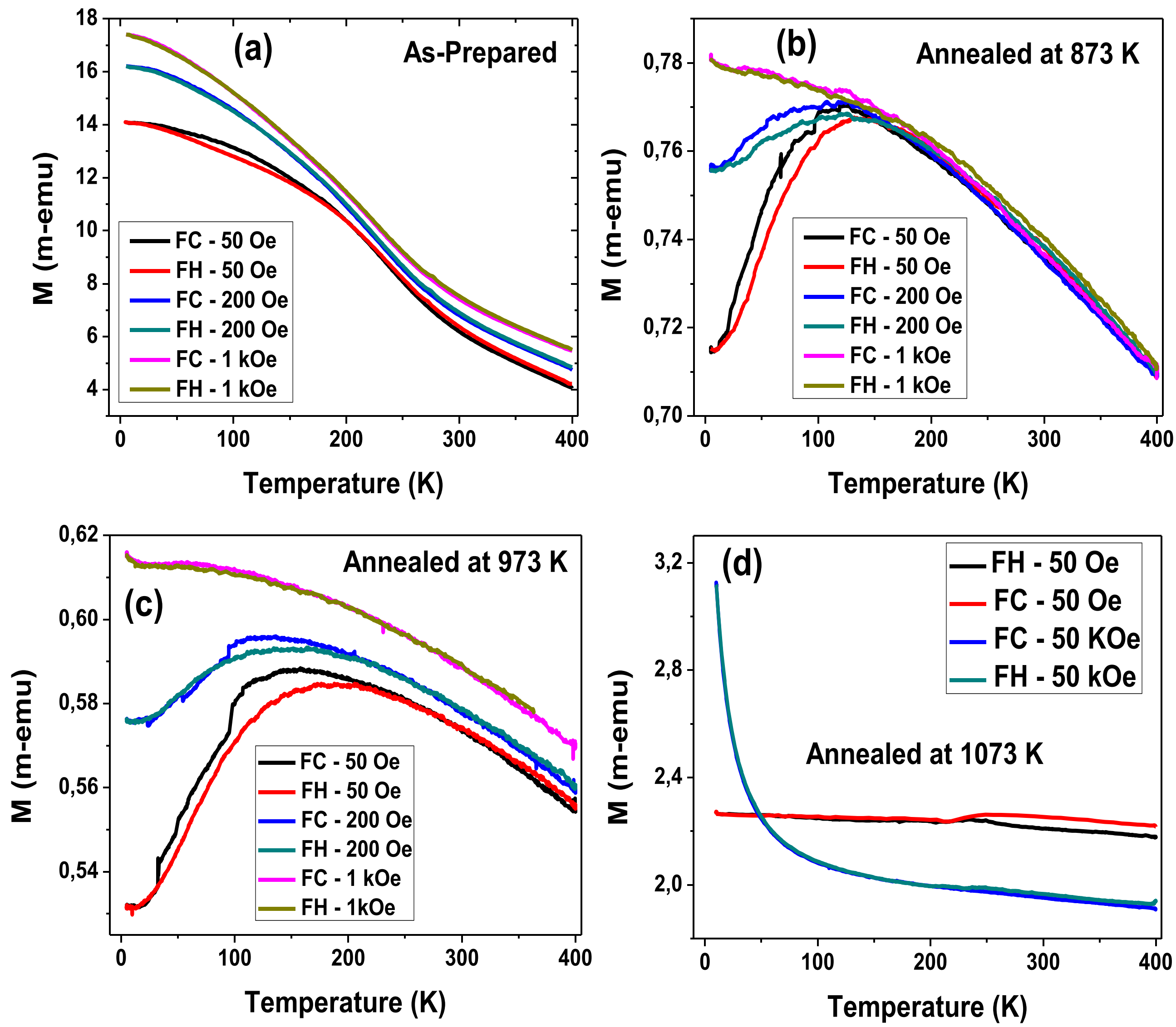

Fig. 5. Temperature dependence of magnetization measured for (a) As-prepared Co_2FeSi, (b) annealed at 873 K, (c) annealed at 973 K and (d) annealed at 1073 K with different applied magnetic field.

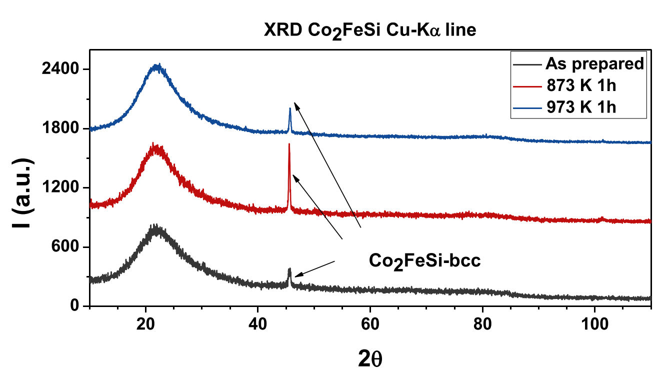

Fig. 6. X-ray powder diffraction patterns of the sample. The XRD spectra were taken from the as prepared sample and after 1 h of thermal annealing at 873 K and 973 K, respectively.

≈ 45.6° can be assumed. However, several previous studies of Co_2FeSi alloys have reported the presence of a Co_2FeSi cubic phase [20,21].

According to theoretical calculations obtained from the materialsproyect.org [52], the Co_2FeSi ferromagnetic phase presents bcc (Fm-3m) structure. Additionally, quite broad XRD of as-prepared sample can be interpreted as the superposition of wide halo typical for amorphous phase and crystalline peak of the bcc (Fm-3m) phase: accordingly, the coexistence of amorphous phase and the crystalline bcc (Fm-3m) phase in the metallic nucleus can be assumed. After 1 h of annealing at 873 K we observe an increase of the intensity of the crystalline peak corresponding to bcc structure.

As discussed elsewhere [43,46], the average grain size, D_g, of the corresponding crystalline phase can be evaluated using the Debye-Scherrer formula. Consequently, we obtained D_g of about 17.8 nm in the as-prepared samples, while some increase in D_g up to 31.6 nm is observed in annealed microwire at 873 K (see Table 1). This increase appears due to the crystallization of the amorphous phase into the bcc phase as the atoms relocate in the new lattice increasing the ferromagnetic behavior and destroying the multistep hysteresis loop. The rapid crystallization at these temperatures has also been reported as a target temperature in the fabrication of Co_2FeSi Heusler alloys for spintronics as shown in the work of Demidov et al., [53]. It is worth noting, that mixed amorphous-nanocrystalline structure, relatively soft magnetic properties and rectangular hysteresis loops were reported for several as-prepared glass-coated microwires, i.e., in $Fe_{72.3}Cu_1Nb_{3.1}Si_{14.5}B_{9.1}$, $Fe_{64.7}Pt_{33.3}B_2$ and $Fe_{50}Pt_{40}Si_{10}$ glass-coated microwires [46,54,55]. Similarly, to studied

Table 1
Average grain size (nm) of as-prepared and annealed Co_2FeSi microwire.

Sample	Grain size D_g (nm)
As prepared	17.8
Annealed at 873 K (1 h)	31.6
Annealed at 973 K (1 h)	26.3

Co_2FeSi microwires, slight D_g increase was observed in $Fe_{64.7}Pt_{33.3}B_2$ and $Fe_{50}Pt_{40}Si_{10}$ microwires with mixed amorphous-nanocrystalline structure upon annealing. As discussed elsewhere, the crystallization process of glass-coated microwires can present different features and even different structure of the precipitates related to strong internals stresses induced by the glass coating [56–58].

On the other hand, the multi-step hysteresis loops observed in annealed samples can also be attributed to the development of the recrystallization process and mixed amorphous-nanocrystalline structure, similarly to that observed previously upon devitrification of amorphous microwires [33,46].

Although a more extended analysis could be done in order to identify the rest of the peaks of the spectra with their corresponding phases, with this data we are able to obtain an explanation for the disappearance of the multistep hysteresis loops. This is due to the appearance of several phases with ferromagnetic ordering that overcome the amorphous magnetic structure and give rise to strong magnetic bi-stability phenomena.

4. Conclusion

In the current study we introduce a cheap and rapid process allowing to fabricate Co_2FeSi glass-coated micro wires by the Taylor-Ulitovsky technique. Magnetic behavior of Co_2FeSi alloy glass-coated micro wires as-prepared and with different annealing conditions has been described. The magnetic behavior of the as-prepared Co_2FeSi microwire shows similar magnetic properties as compared with Co_2FeSi alloys prepared by different techniques such as physical vapor deposition and chemical deposition techniques. The different conditions of annealing have been tested and investigated in the current study. A strong dependence of the magnetization and the structural properties on the annealing conditions has been confirmed. The annealed sample at 873 K and 973 K shows anomalous magnetic behavior below the room temperature combined with changes in the magnetic and structure phases compared to the as-prepared samples. We supposed a critical temperature, which well matched with T_B and T_M for the annealed sample at 873 K and 973 K, where the anomalous magnetic behavior starts to appear. Additionally, we illustrated that the Co_2FeSi glass-coated micro wires are sensitive to the annealing conditions which affect the lattice structure and the magnetic behavior. We guess that these observations will open the approach to use Co_2FeSi glass-coated micro wires with unusual magnetization behavior to design devices based on the magnetization switching.

Data Availability

Data will be made available on request.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Mohamed Salaheldeen, Alfonso Garcia-Gomez, Paula Corte-Leon, Mihail Ipatov, Valentina Zhukova, Julian Maria Gonzalez, Arcady Zhukov reports financial support was provided by Government of Spain, Ministerio de Ciencia, Innovación y Universidades. Mohamed Salaheldeen, Alfonso Garcia-Gomez, Paula Corte-Leon, Mihail Ipatov, Valentina Zhukova, Julian Maria Gonzalez, Arcady Zhukov reports was provided by European Commission. Mohamed Salaheldeen, Alfonso Garcia-Gomez, Paula Corte-Leon, Mihail Ipatov, Valentina Zhukova, Julian Maria Gonzalez, Arcady Zhukov reports was provided by Government of the Basque Country.

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