



# Challenges In Growing Thermally Evaporated Double Half-Heusler $Mn_2CoFeGe_2$ Thin Films

T. Naaraayanan<sup>1</sup>, M. Saroja<sup>1</sup>, M. Venkatachalam<sup>1</sup>

<sup>1</sup>Department of Electronics, Research and Development Centre, Erode Arts and Science College (Autonomous), Erode-638009, India.

## Abstract

We present initial results from investigations into thin films of a novel material, a double half-Heusler  $Mn_2FeCoGe_2$  thin film deposited onto silicon (100) and glass substrates using thermal evaporation. The specimens were examined for their morphology and crystallographic structure using scanning electron microscopy and diffraction techniques. The composition was analyzed using X-ray spectroscopy. Our investigation further underscores the critical role of stoichiometry in achieving the desired properties of the material.

**Keywords:**  $Mn_2CoFeGe_2$ , Double Half-Heusler, Thin films

## 1. Introduction

Interest in Heusler alloy thin films is growing due to their unique properties, such as tunable anisotropy, large spin polarization, and a high Curie temperature ( $T_c$ ). These characteristics make them appealing for various spintronic applications, including high-density magnetic storage,<sup>1,2</sup> spin-transfer-torque magnetic random-access memories<sup>3</sup>, and magnonic devices<sup>4</sup>.

Identifying new materials with exceptional properties remains a central challenge in condensed matter physics. Many manganese-based (Mn-based) Heusler alloys exhibit strong ferromagnetic behavior and possess high magnetic moments, making them suitable for various applications, including magnetic materials, magnetic sensors, magnetic memory devices, and spintronic devices. Additionally, some Mn-based Heusler alloys demonstrate a significant magnetostrictive effect, meaning they undergo noticeable shape changes in response to an applied magnetic field. This property is particularly advantageous for use in actuators and energy-harvesting devices.

We believe the differences were due to the challenges encountered when growing thin films of Heusler compounds,<sup>5</sup> which have delicate properties. The primary issue is the structural quality. However, it appears that Heusler compounds maintain a stable  $m\bar{3}m$  point group structure across many different chemical elements and deposition techniques, such as ingot melting,<sup>6</sup> and sputtering<sup>7,8</sup>.

## 2. Experimental Technique.

Initially, a 1-gram  $Mn_2FeCoGe_2$  alloy was prepared using an arc furnace. High-purity elemental manganese (Mn), iron (Fe), cobalt (Co), and germanium (Ge) ( $\geq 99.99\%$ ) were combined in stoichiometric proportions and melted together in a high-purity argon atmosphere. To compensate for weight loss during melting, primarily due to the volatility of manganese, an additional 3% manganese was added. The homogeneity of the ingot was ensured by remelting it several times and flipping it between melts. The final weight loss after melting was less than 1%. To further improve compositional uniformity, the ingot was sealed in a quartz ampoule and annealed at 1000 °C for six hours in a tube furnace. The sample was then naturally cooled to room temperature before analysis.

To deposit the thin films, the  $\text{Mn}_2\text{CoFeGe}_2$  ingot was broken into smaller pieces and placed in a tungsten boat for thermal evaporation using Advanced Processing Technologies (APT). All films were deposited onto silicon (Si (100)) and glass substrates during deposition, and the chamber pressure was maintained at  $5 \times 10^{-6}$  Torr. Afterward, post-annealing was performed on all samples at temperatures ranging from  $100^\circ\text{C}$  to  $400^\circ\text{C}$  for 30 minutes in a vacuum environment with a pressure of  $5 \times 10^{-3}$  Torr.

Thin film samples were exposed to  $\text{Cu-K}\alpha$  radiation ( $1.54 \text{ \AA}$ ) to obtain X-ray diffraction (XRD) patterns at room temperature using a Rigaku Smart Lab system (Japan). The composition of the sample was analyzed using a JEOL JIB-4700 FIB-SEM in energy-dispersive X-ray spectroscopy (EDS) mode, with EDS data acquired via scanning electron microscopy (SEM).

### 3. Results and Discussion

#### 3.1 X-ray diffraction studies:

The structural characteristics of the films were examined using a high-resolution X-ray diffractometer with a  $\text{Cu-K}\alpha$  radiation source. The diffraction patterns were recorded in the  $2\theta$  range of  $5^\circ$  to  $80^\circ$ , as illustrated in Figure 1. We observed only two peaks in the  $\text{Mn}_2\text{CoFeGe}_2$  film, occurring at approximately  $44\text{--}45^\circ$  and  $67\text{--}68^\circ$ . The positions of these peaks align well with the expected positions of the fundamental peaks associated with the (220) and (400) lattice planes on a Si (100) substrate, consistent with the XA- or L21-type structure<sup>9</sup>. However, the thin films on the glass substrate did not exhibit any XRD peaks, likely due to the limitations of the thermal evaporation method.

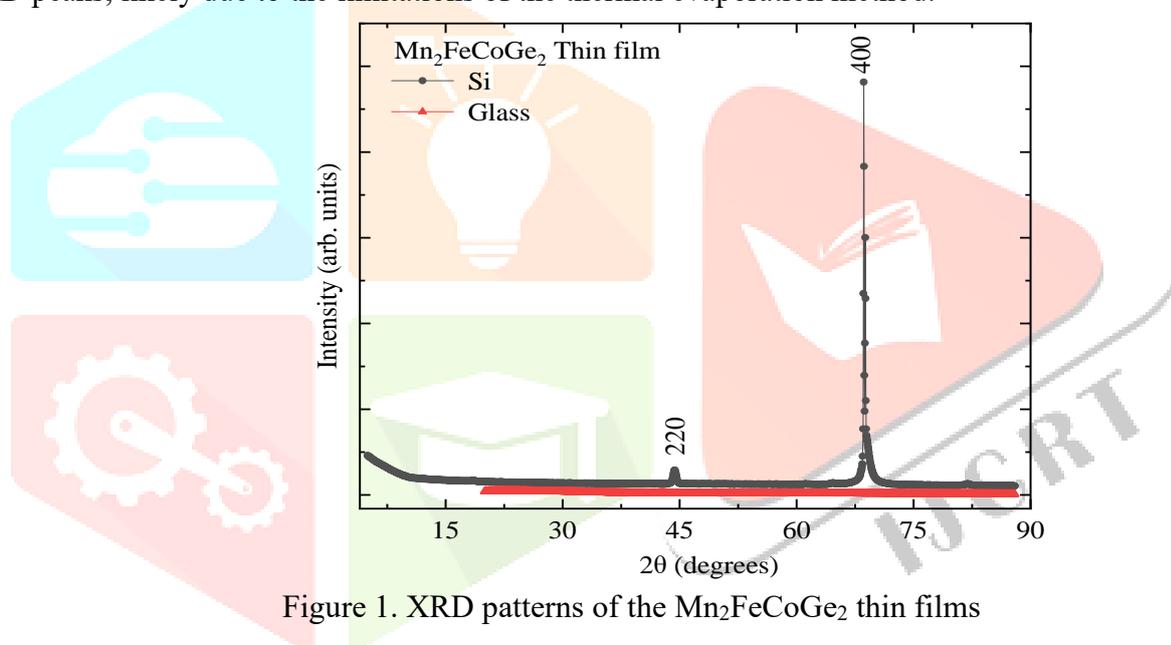


Figure 1. XRD patterns of the  $\text{Mn}_2\text{FeCoGe}_2$  thin films

#### 3.2 Morphological analysis

The surface microstructure was examined using FESEM images presented in Figure 2, while the EDS image is displayed in Figure 3. These figures illustrate the elemental distribution at the same scale of  $0.1 \mu\text{m}$  on the glass substrate, with Figure 4 showing the surface microstructure and Figure 5 an EDS image for the Si (100) substrate. The images indicate the elemental distribution. The film surface evolves into a continuous layer. The surface grains or granules may appear because the film was grown for an insufficient time, preventing the formation of a continuous surface, resulting in small grains being visible across the surface. As the deposition time increases, the merging of islands begins, creating a continuous surface structure.

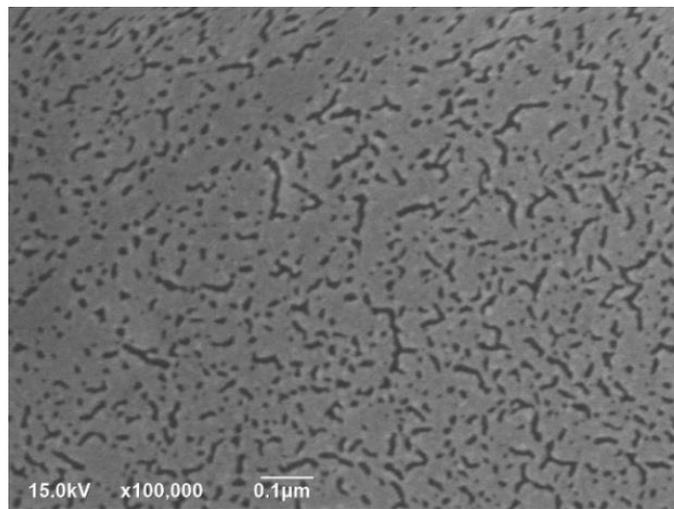


Figure 2. FESEM images of the Mn<sub>2</sub>FeCoGe<sub>2</sub> films on the Glass substrate

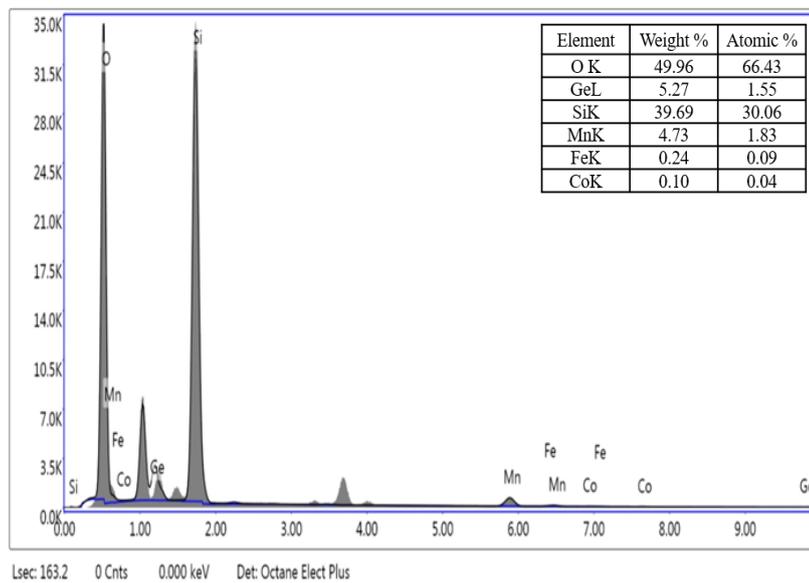


Figure 3. The EDS spectrum reveals the main components (Iron (Fe), cobalt (Co), manganese (Mn), and Germanium (Ge)) of the Mn<sub>2</sub>FeCoGe<sub>2</sub> films on the Glass substrate.

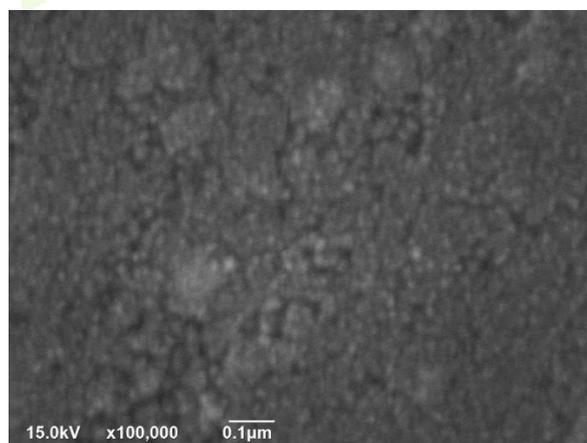


Figure 4. FESEM images of the Mn<sub>2</sub>FeCoGe<sub>2</sub> films on the Si (100) substrate

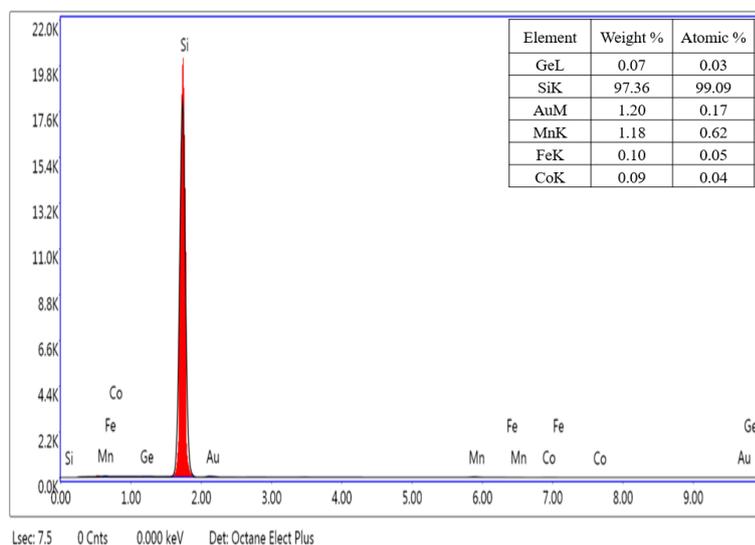


Figure 5. The EDS spectrum reveals the main components (Iron (Fe), cobalt (Co), manganese (Mn), and Germanium (Ge) of the  $Mn_2FeCoGe_2$  films on Si (100) substrate.

## Conclusion

In this study, we fabricated  $Mn_2FeCoGe_2$  thin films on Si (100) and glass substrates using thermal evaporation technology. We encountered several challenges during the growth process and examined the XRD and EDS characteristics. Unfortunately, the results did not meet our expectations. We aim to report our findings to assist the research community in understanding the difficulties associated with growing such a complex  $Mn_2FeCoGe_2$  double half-Heusler alloy. There are still many Heusler compounds, including quaternary alloys, that remain to be explored.

## Declaration of Competing Interest

The authors declare that there is no conflict of interest regarding the publication of this article.

## Acknowledgments

The authors would like to express their gratitude to the Departments of Electronics, Research and Development Centre, Erode Arts and Science College (Autonomous) for providing all resources required for the completion of this work.

## References.

- (1) Bai, Z.; Shen, L.; Han, G.; Feng, Y. P. DATA STORAGE: REVIEW OF HEUSLER COMPOUNDS. *SPIN* **2012**, *02* (04), 1230006. <https://doi.org/10.1142/S201032471230006X>.
- (2) Ma, Q. L.; Zhang, X. M.; Miyazaki, T.; Mizukami, S. Artificially Engineered Heusler Ferrimagnetic Superlattice Exhibiting Perpendicular Magnetic Anisotropy. *Sci Rep* **2015**, *5* (1), 7863. <https://doi.org/10.1038/srep07863>.
- (3) Ikeda, S.; Hayakawa, J.; Lee, Y. M.; Matsukura, F.; Ohno, Y.; Hanyu, T.; Ohno, H. Magnetic Tunnel Junctions for Spintronic Memories and Beyond. *IEEE Trans. Electron Devices* **2007**, *54* (5), 991–1002. <https://doi.org/10.1109/TED.2007.894617>.
- (4) Chumak, A. V.; Vasyuchka, V. I.; Serga, A. A.; Hillebrands, B. Magnon Spintronics. *Nature Phys* **2015**, *11* (6), 453–461. <https://doi.org/10.1038/nphys3347>.
- (5) Guillemard, C.; Petit-Watelot, S.; Devolder, T.; Pasquier, L.; Boulet, P.; Migot, S.; Ghanbaja, J.; Bertran, F.; Andrieu, S. Issues in Growing Heusler Compounds in Thin Films for Spintronic Applications. *Journal of Applied Physics* **2020**, *128* (24). <https://doi.org/10.1063/5.0014241>.
- (6) Webster, P. J. Magnetic and Chemical Order in Heusler Alloys Containing Cobalt and Manganese. *Journal of Physics and Chemistry of Solids* **1971**, *32* (6), 1221–1231. [https://doi.org/10.1016/s0022-3697\(71\)80180-4](https://doi.org/10.1016/s0022-3697(71)80180-4).
- (7) Sakuraba, Y.; Kokado, S.; Hirayama, Y.; Furubayashi, T.; Sukegawa, H.; Li, S.; Takahashi, Y. K.; Hono, K. Quantitative Analysis of Anisotropic Magnetoresistance in  $Co_2MnZ$  and  $Co_2FeZ$  Epitaxial

Thin Films: A Facile Way to Investigate Spin-Polarization in Half-Metallic Heusler Compounds. *Applied Physics Letters* **2014**, *104* (17). <https://doi.org/10.1063/1.4874851>.

- (8) Ortiz, G.; García-García, A.; Biziere, N.; Boust, F.; Bobo, J. F.; Snoeck, E. Growth, Structural, and Magnetic Characterization of Epitaxial Co<sub>2</sub>MnSi Films Deposited on MgO and Cr Seed Layers. *Journal of Applied Physics* **2013**, *113* (4). <https://doi.org/10.1063/1.4789801>.
- (9) Tajiri, H.; Kumara, L. S. R.; Sakuraba, Y.; Chen, Z.; Wang, J.; Zhou, W.; Varun, K.; Ueda, K.; Yamada, S.; Hamaya, K.; Hono, K. Structural Insight Using Anomalous XRD into Mn<sub>2</sub>CoAl Heusler Alloy Films Grown by Magnetron Sputtering, IBAS, and MBE Techniques. *Acta Materialia* **2022**, *235*, 118063. <https://doi.org/10.1016/j.actamat.2022.118063>.

