(Supplementary Information)

Disordered Spin Gapless Semiconducting CoFeCrGa Heusler Alloy Thin Films on Si (100): Experiment and Theory

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S1. Surface and Morphological Analysis: XRR and AFM

The Fig. S1 (a) shows the XRR profile recorded for B-550 film sample of nominal thickness 50nm. The presence Kiessig fringes even up to $2\theta \sim 4$ assures the good surface quality and less intermixing at the interface [1]. The recorded curve was fitted using X-ray reflectivity (segmented V.1.2) software through simulation using the model which assumes the formation of a native oxide layer over the top surface of the CFCG film as shown in the inset of the Fig. S1 (a). The thickness, density, and roughness of CFCG and oxide layers evaluated through the fitted data is shown the inset of the bottom left of the Fig. S1 (a). The simulated curve gives the thickness of CFCG film ~ 47.59 nm and that of oxide layer as ~3.32 nm. The density of the CFCG film (~6.78 g/cc) is found to be nearly close to its bulk density value of 7.47 g/cc, which suggests that the grown film is non-porous and densely packed.

The topography of the CFCG film was investigated using AFM technique. The sample was scanned over an area of 500×500 nm^2 in tapping mode. The cantilever of the dimension 225×30×3 μm^3, embedded with a platinum coated tip of radius <10 nm was used in tapping mode. The Fig. S1 (b) shows one of the AFM images recorded on the 50 nm thin CFCG film. It can be seen that film is quite homogeneous and clearly exhibits a granular microstructure. The grains formed are of spherical shape with an average size of ~25 nm. The root mean square roughness (R_{rms}) of the film is ~1.80 nm which is equivalent to that obtained from XRR measurement (~1.64 nm)

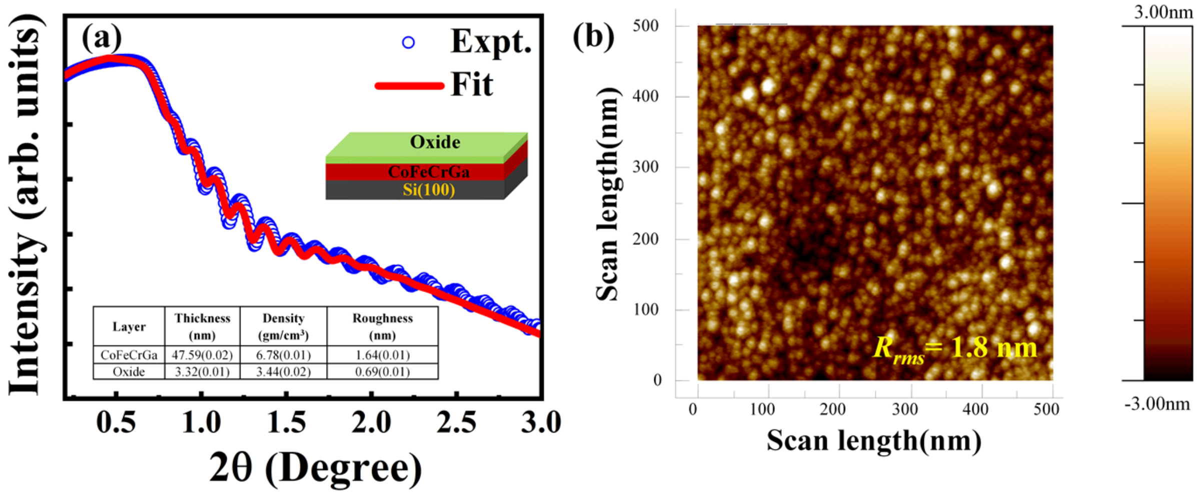

Fig. S1 (a) XRR spectra of the B-550 CFCG thin film. The simulation model and the values of the thickness, density, and roughness (along with the errors in the parenthesis) extracted through the fitted data are shown in the top right and bottom left of the main panel, respectively. **(b)** The topographical AFM image of B-550 sample with its root mean square roughness (R_{rms}).

S3. Compositional Analysis: EDAX

The EDAX measurement was performed over B-550 film to quantify and analyse the different elements present in the sample as shown in Fig. S2 (a-e). Figures S2 (a, b, c & d) show the elemental mapping of individual Co, Fe, Cr and Ga elements on ~ 250 μm^2 area of the sample which depicts that all the elements are uniformly distributed over the film surface and the film sample is slightly Gallium deficient. The EDS spectrum (counts/sec *vs* keV) for Co, Fe, Cr, Ga, and Si atoms is shown in Fig. S2 (e). The table in the inset shows the normalized composition of Co, Fe, Cr and Ga elements in terms of weight and atomic percentage which confirmed their stoichiometry to be $Co_{24.2}Fe_{29.1}Cr_{25.1}Ga_{21.6}$. The excess Fe and deficient Ga may be possibly causing the disorder in the film sample.

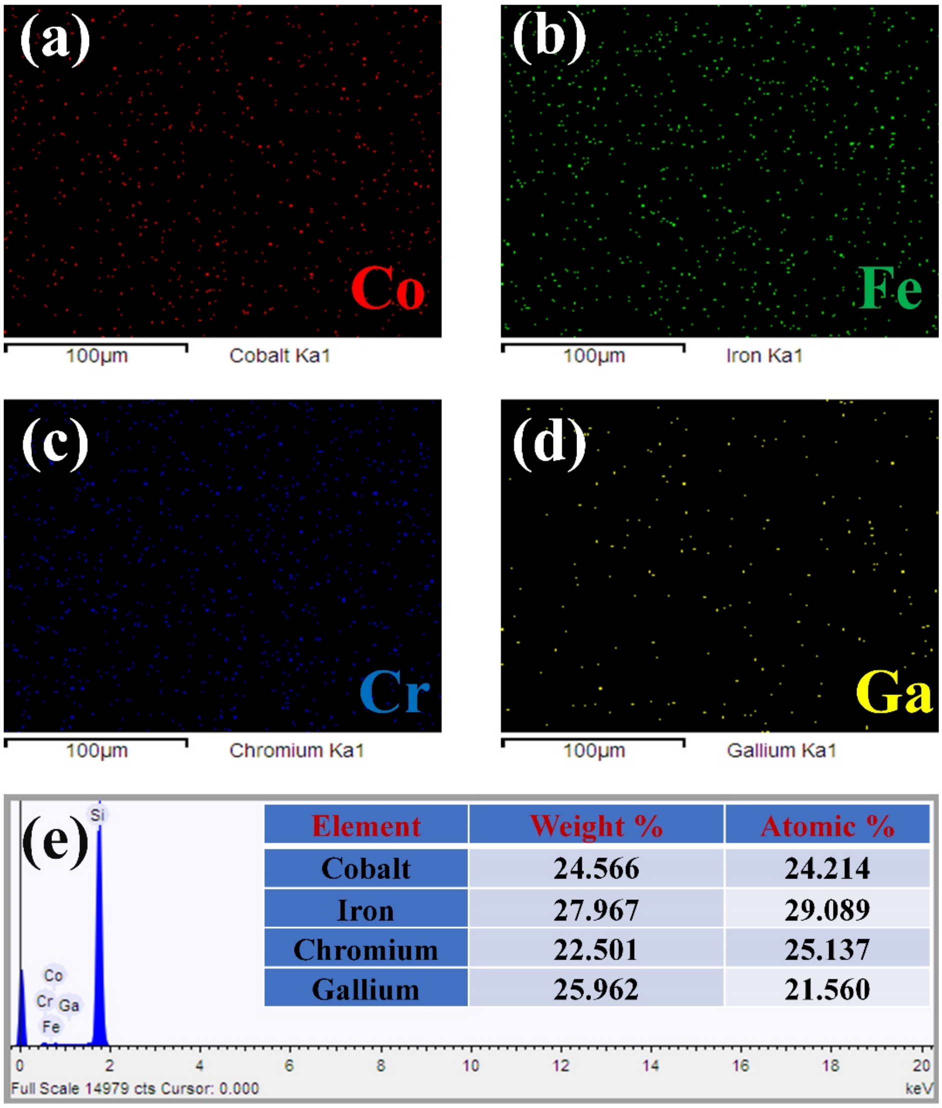

FIG S2. Elemental mapping of **(a)** Cobalt, **(b)** Iron, **(c)** Chromium and **(d)** Gallium on~$250\mu m^2$ area of the sample. **(e)** EDS spectrum (counts/sec *vs* keV) for Co, Fe, Cr, Ga, and Si atoms with table in the inset showing the normalized weight % and atomic% of CFCG film after removing the contribution of Si (100) substrate.

REFERENCES

1 N. K. Gupta, V. Barwal, S. Hait, L. Pandey, V. Mishra, L. Saravanan, A. Kumar, N. Sharma, N. Kumar, S. Husain and S. Chaudhary, *Thin Solid Films*, 2022, **756**, 139355.