Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary Material:

Spin-gapless semiconducting characteristics and related band topology of quaternary Heusler alloy CoFeMnSn

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S1: Density of state at different U_{eff} :

We have observed a decrease in the majority density of states (DOS) at the Fermi level (E_F) as the U value increases. The table provided compares various U values to the corresponding majority DOS.

Table S1: Variation of DOS near E_F in the majority band for different values of U_{eff}

U_{eff} (eV)	Majority DOS at E_F (States/eV)
0	0.34
0.2	0.26
0.5	0.14
0.8	0.08

U_{eff} = 0 eV U_{eff} = 0.2 eV U_{eff} = 0.5 eV U_{eff} = 0.8 eV

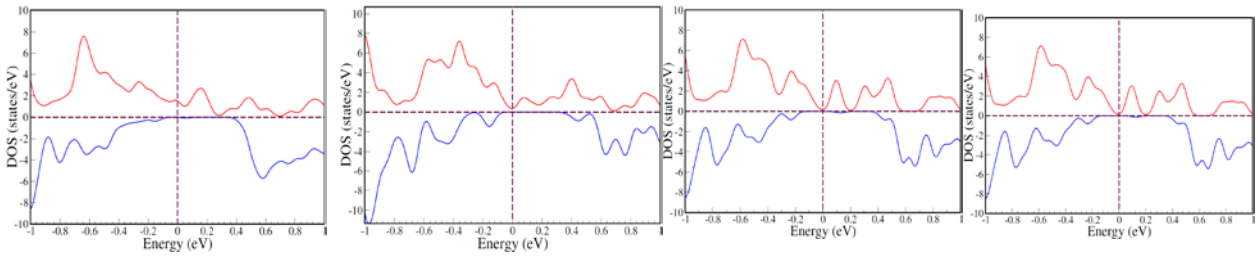

Figure S1: Density of states for various $U_{ef\,f}$, where majority DOS value decreases with increase of U_{eff} and a gap maintained.

S2: Density of state at different K points:

Our calculations of the DOS using various K-mesh sampling methods led us to conclude that, following 12 × 12 × 12 point sampling, the relaxation energy is nearly constant and the DOS are nearly unchanged at various Higher K-mesh.

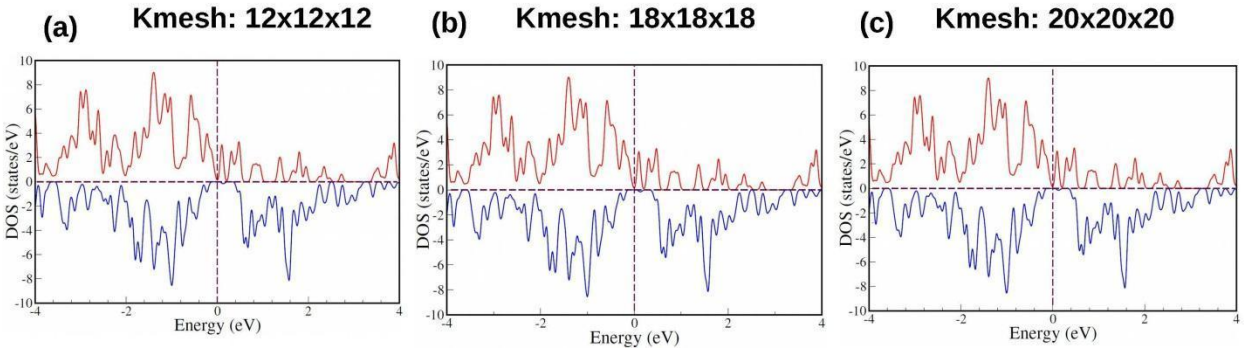

Figure S2: Density of states for various K-mesh

S3: The Bader charge analysis for P = 0 GPa and P = 50 GPa

Based on our Bader charge analysis, we have noted that Sn is redistributing its charge to Co, Fe, and itself, maintaining a total charge of 28 at both 0 and 50 GPa pressure. This observation supports our assertion that the Slater-Pauling (SP) rule is upheld. Additionally, this redistribution is evident in the charge density resulting from both spin up and spin down, aligning well with the magnetic moment we have calculated.

P=0 Gpa

Table S3.1: The Bader charge analysis for P = 0 GPa

Atom	X	Y	Z	Charge	At. vol.	Up-spin (charge)	Down-spin (charge)
Mn	2.982	2.982	2.982	6.595	11.522	4.767	1.827
Fe	1.491	1.491	1.491	8.261	12.848	4.370	3.890
Co	4.473	4.473	4.473	9.258	12.903	4.979	4.279
Sn	0.000	0.000	0.000	3.886	15.777	1.885	2.001

P= 50 Gpa

Table S3.2: The Bader charge analysis for P = 50 GPa

Atom	X	Y	Z	Charge	At. vol.	Up-spin (charge)	Down-spin (charge)

Mn	2.803	2.803	2.803	6.618	9.632	4.684	1.934
Fe	1.401	1.401	1.401	8.280	10.841	4.460	3.820
Co	4.205	4.205	4.205	9.490	0.974	5.364	4.126
Sn	0.000	0.000	0.000	3.610	12.615	1.615	1.995

S4: Effect of Large U on Fe site

In the large U limit on the Fe site, it is found that the total magnetic moment deviates from what is observed experimentally and breaks the SP rule. In the table below, we have listed the change of magnetic moments for all four atoms at U_{Fe}=1 & 3 eV. The SGS is robust for $U_{Fe} \leq 0.5$ eV, but for large U value the system deviates from SGS behavior as displayed in electronic band dispersion in Fig S4.

Table S4: Magnetic moment of individual atoms

Atom	Magnetic moment (U=1eV) in μ_B	Magnetic moment (U=3eV) in μ_B
Mn	2.90	2.62
Fe	0.81	1.80
Co	0.64	0.32
Sn	-0.13	-0.18
Total	4.22 μ_B/f.u.	4.56 μ_B/f.u.

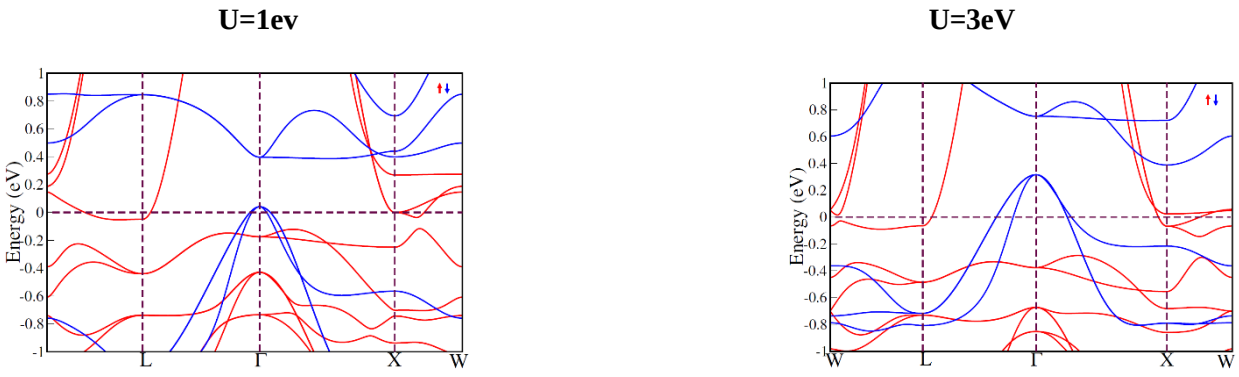

Figure S4: Band structure for large U (1 eV and 3 eV) on Fe site

S5: Effect of Large U on Co site

The SGS state is preserved upto U_{Co} ≤0.8 eV, and for higher U_{Co} values, the total magnetic moment deviates from the experimental value as well as Slater-Pauling value. As seen by the electronic band dispersion in Fig. S5, the system deviates from SGS behaviour for large U values.

Table S5: Magnetic moment of individual atoms

Atom	Magnetic moment (U=1eV) in μ_B	Magnetic moment (U=3eV) in μ_B
Mn	2.96	2.80
Fe	0.44	0.38
Co	0.89	1.33
Sn	-0.12	-0.13
Total	4.17 μ_B/f.u.	4.38 μ_B/f.u.

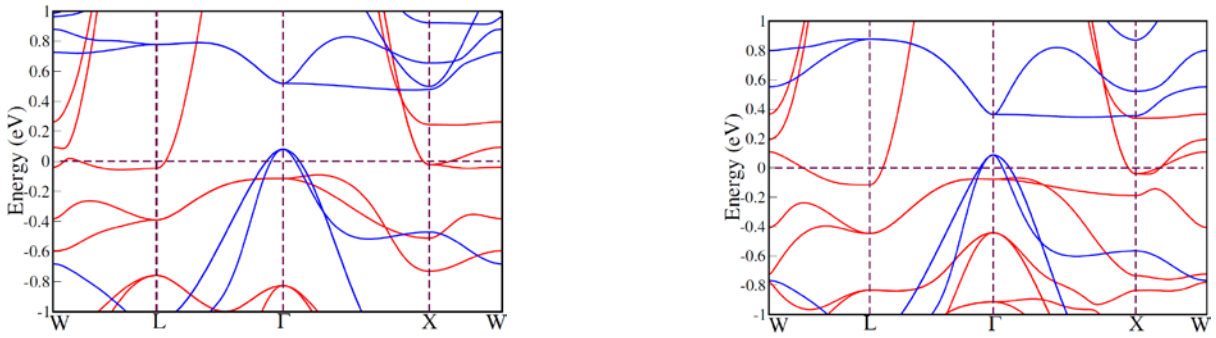

Figure S5: Band structure for large U (1 eV and 3 eV) on Co site

S6: Effect of Large U on Mn site

The SGS state is preserved upto $U_{Mn} \leq 1.3$ eV and for higher U_{Mn} values the total magnetic moment deviates from the experimental value and breaks the Slater-Pauling rule. The SGS state is completely vanished at 3 eV.

Table S6: Magnetic moment of individual atoms

Atom	Magnetic moment (U=1eV) in μ_B	Magnetic moment (U=3eV) in μ_B
Mn	3.10	3.56
Fe	0.64	0.35
Co	0.42	0.40
Sn	-0.10	-0.12
Total	4.07 μ_B/f.u.	4.19 μ_B/f.u.

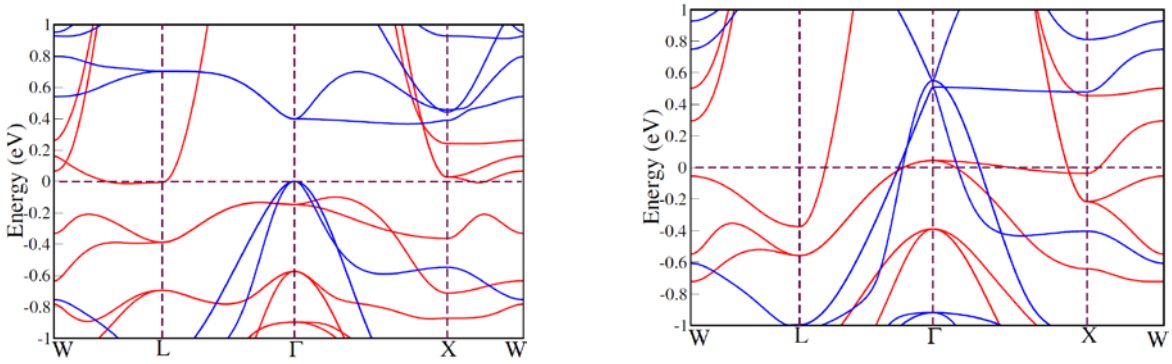

Figure S6: Band structure for large U (1 eV and 3 eV) on Mn site