Figures S1-S4 show the experimental X-ray diffraction data and the calculated pattern, which was used to confirm the crystal structure for each sample. The calculated and experimental XRD pattern for CoFeMnSi has been previously published in Alijani *et al.*, *Physical Review B* **84** 224416 (2011).

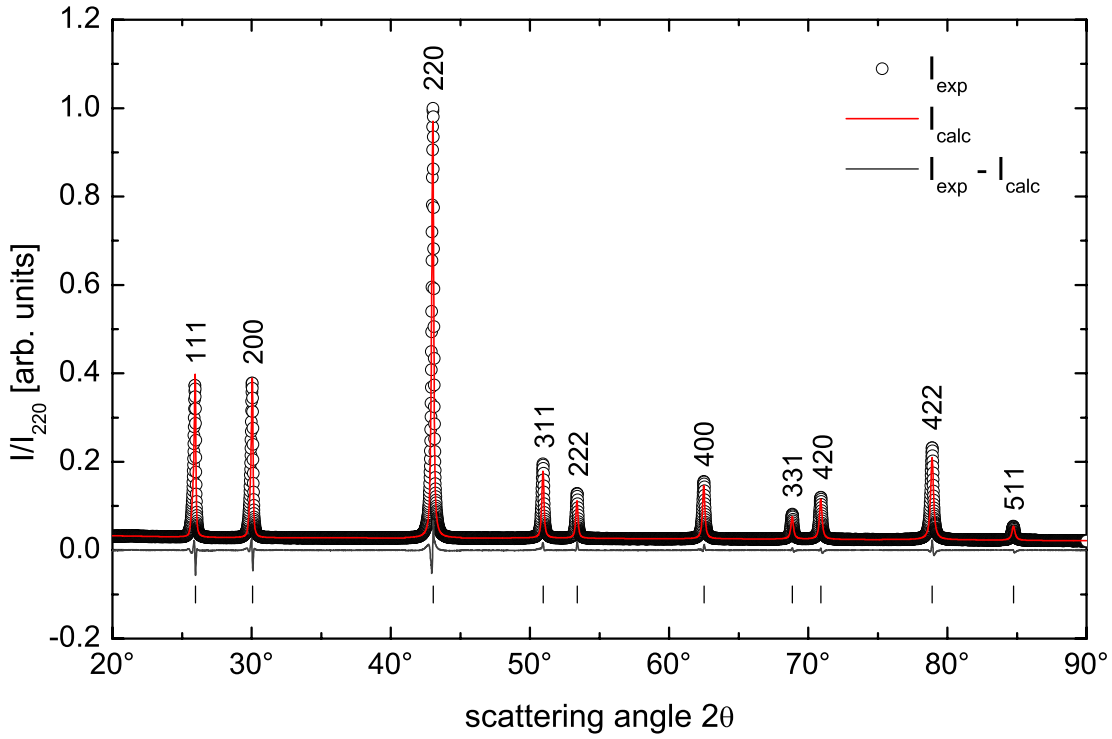

Figure S1. Observed and calculated X-ray diffraction pattern for NiMnSb. This compound exhibits the $C1_b$ structure.

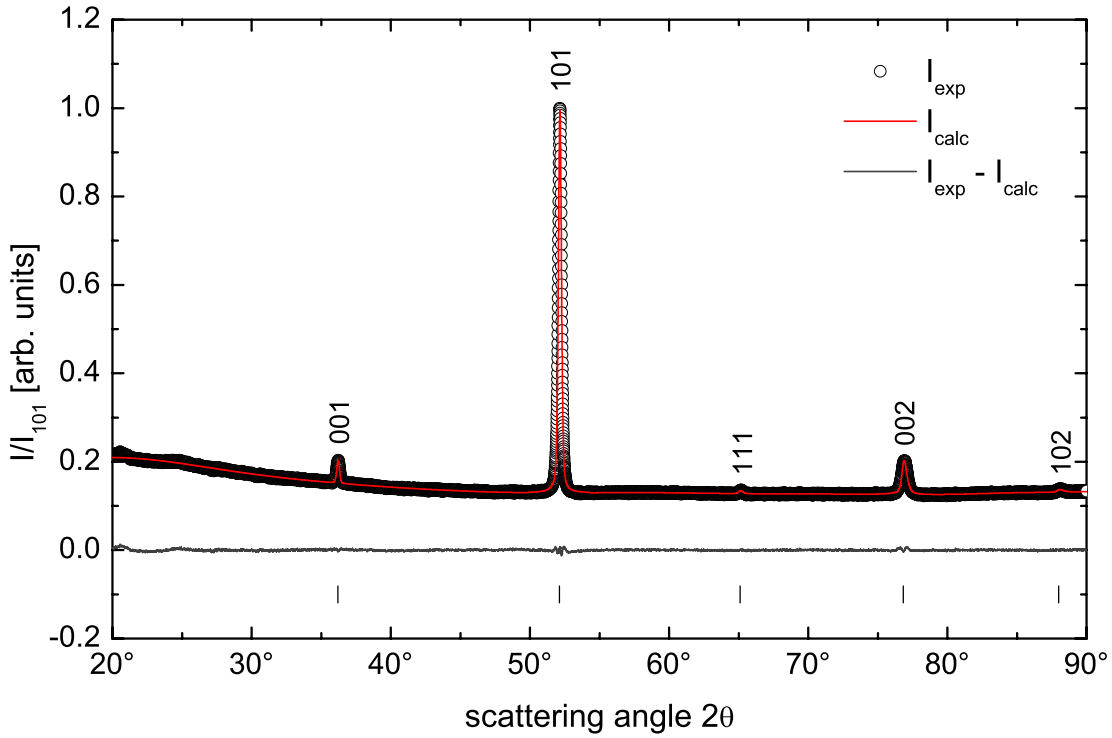

Figure S2. Observed and calculated X-ray diffraction pattern for Co_2MnAl. This compound has the B2 structure.

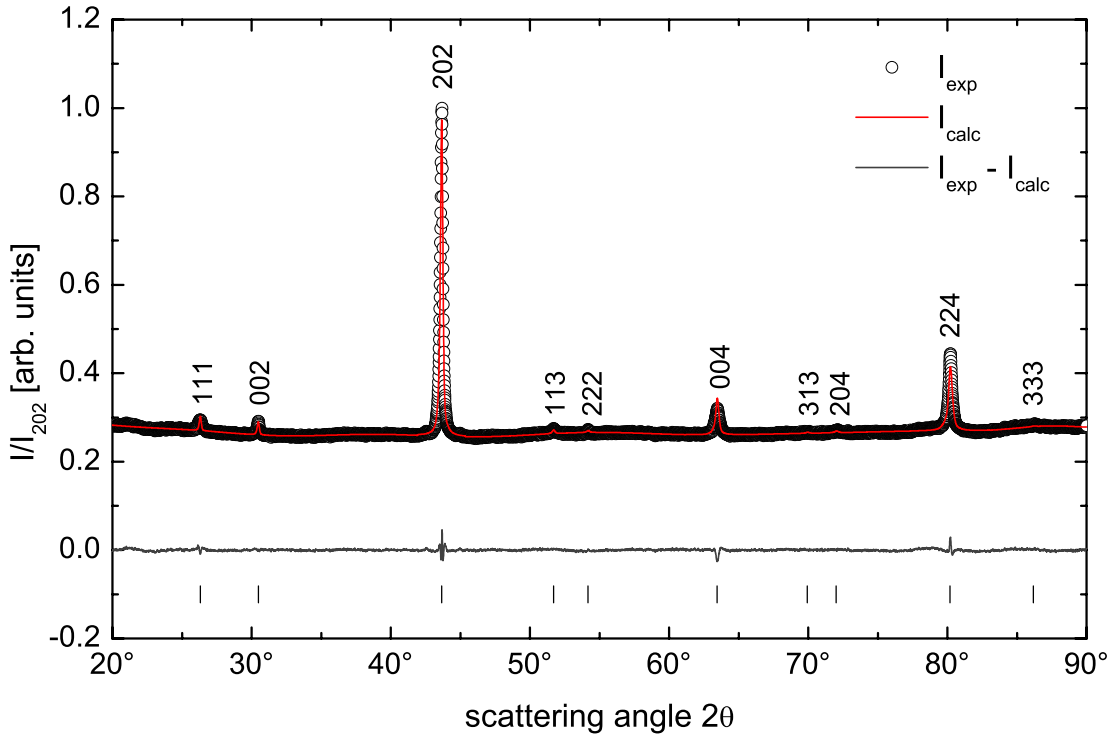

Figure S3. Observed and calculated X-ray diffraction pattern for Mn_2CoAl. This compound exhibits the inverse X_a structure.

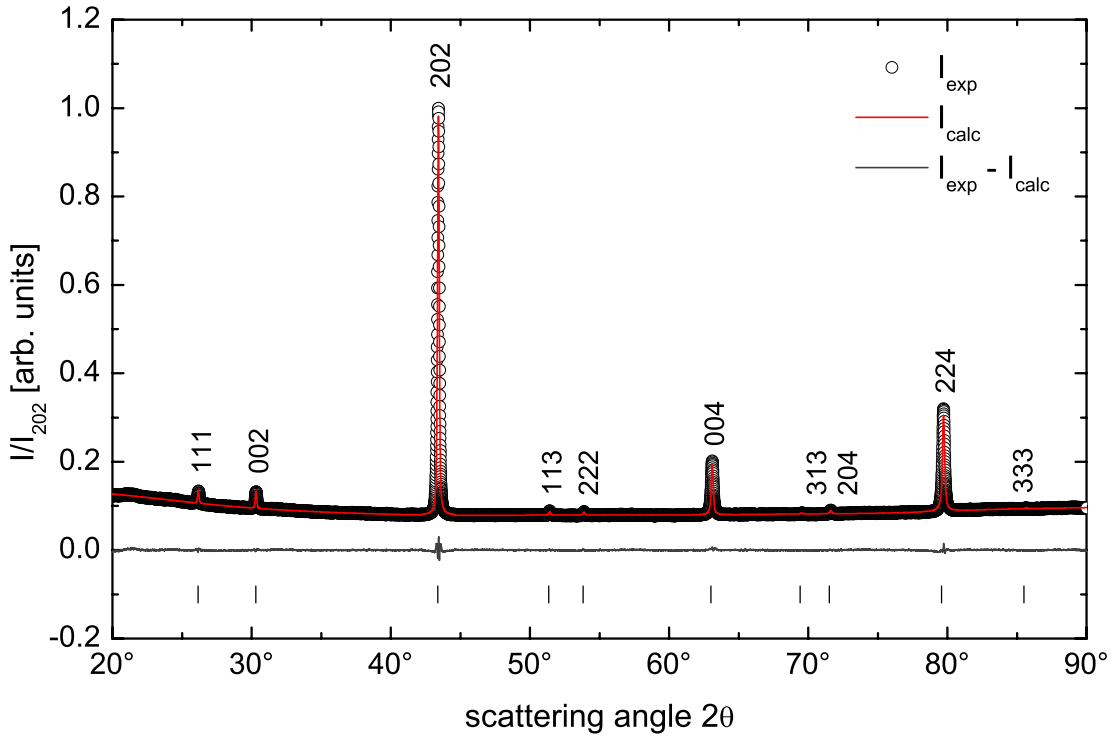

Figure S4. Observed and calculated X-ray diffraction pattern for Mn_2VAl. This compound exhibits the *L2₁* structure.

Muffin Tin Radii (R_{MT}) used in calculations

Heusler Compounds

Compound	X,Y	Z
XX'YZ (CoFeMnSi)	2.28 a_{0B}	2.14 a_{0B}
X_2YZ(Co_2MnAl, Mn_2VAl, Mn_2CoAl)	2.28 a_{0B}	2.14 a_{0B}
XYZ (NiMnSb)	2.41 a_{0B}	2.26 a_{0B}

Reference Materials

Material or Compound	Mn	O
Mn	2.28 a_{0B}	-
MnO, MnO_2	2.22 a_{0B}	1.97 a_{0B}

a_{0B} (Bohr radius) = 0.5291772083 Å