The Relationship between Magnetism and Composition in Heusler Co_2CrAl-based Thin Films

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Introduction

A number of $L2_1$ phase alloys (composition X_2YZ) have exceptional magnetoelectric properties. Within this group, Co_2CrAl is predicated as a promising candidate for half-metallic ferromagnetism. However, synthesis of this stoichiometry is a challenge because of its susceptibility to secondary phase precipitation and structural defect formation, causing discrepancies in the theoretical and experimentally observed properties.

In addition, recent techniques for growing incongruent melting compounds have produced alloys that suffer from phase transformation via spinodal decomposition, prompting secondary phase formation that also depresses magnetic properties. Since this secondary phase is difficult to dissolve once it is formed, it is necessary to access a single-phase field from the start.

This project explores using the space of off-stoichiometry and elemental substitution to design an optimized composition that avoids the region of immiscibility and therefore realizes desirable magnetic behavior.

Objective and Methods

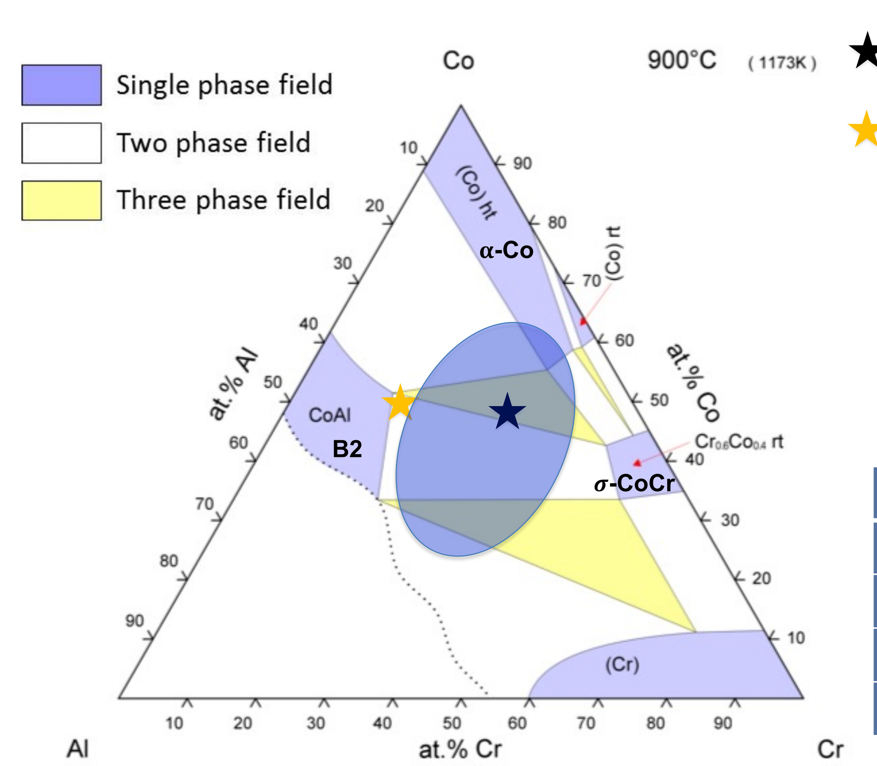
Objective.

This project will investigate the effects of phase formation and atomic disorder in full-Heusler systems on their unique magnetic phenomena. By exploring off-stoichiometry, the objective is to track changes in magnetism that result from changes in the order or composition of Heusler Co_2CrAl-based combinatorial thin films.

Methods.

1. ML model (non-linear dimensionality reduction and Gaussian Mixtures Modeling) for unsupervised classification of Extended X-ray Absorption Fine Structure (EXAFS) data.
2. Synchrotron XRD from Standard Linear Accelerator Center (SLAC) for characterizing disorder and changes in long-range order.
3. Magnetic hysteresis from Magneto-Optical Kerr Effect (NanoMOKE3) measurements at room temperature.

Motivation for choosing compositional series



★ Co_2CrAl

★ Alternative single-phase $Co_2Cr_{0.4}Al_{1.6}$ [1]

Figure 1. Isothermal section for Co-Cr-Al system at 900°C. [2]

Magnetron sputtering used to deposit the target compositional series on a SiO_2/Si substrate with the following parameters:

Deposition temperature (°C)	25
Deposition time (hrs)	1.15
Vacuum during deposition (mtorr)	7.5
Post-annealing temperature (°C)	400
Annealing time (hrs)	10

Results

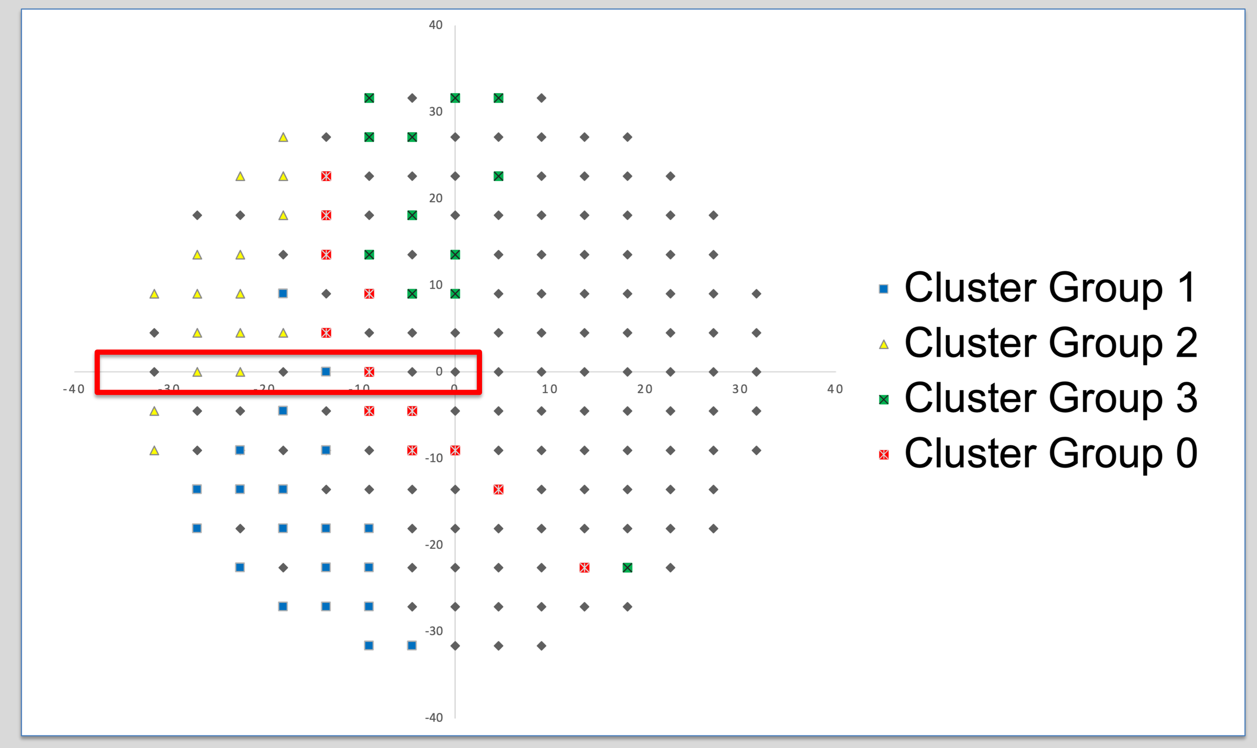


Figure 2. Cluster groups informed by ML model.

Magnetic Trends

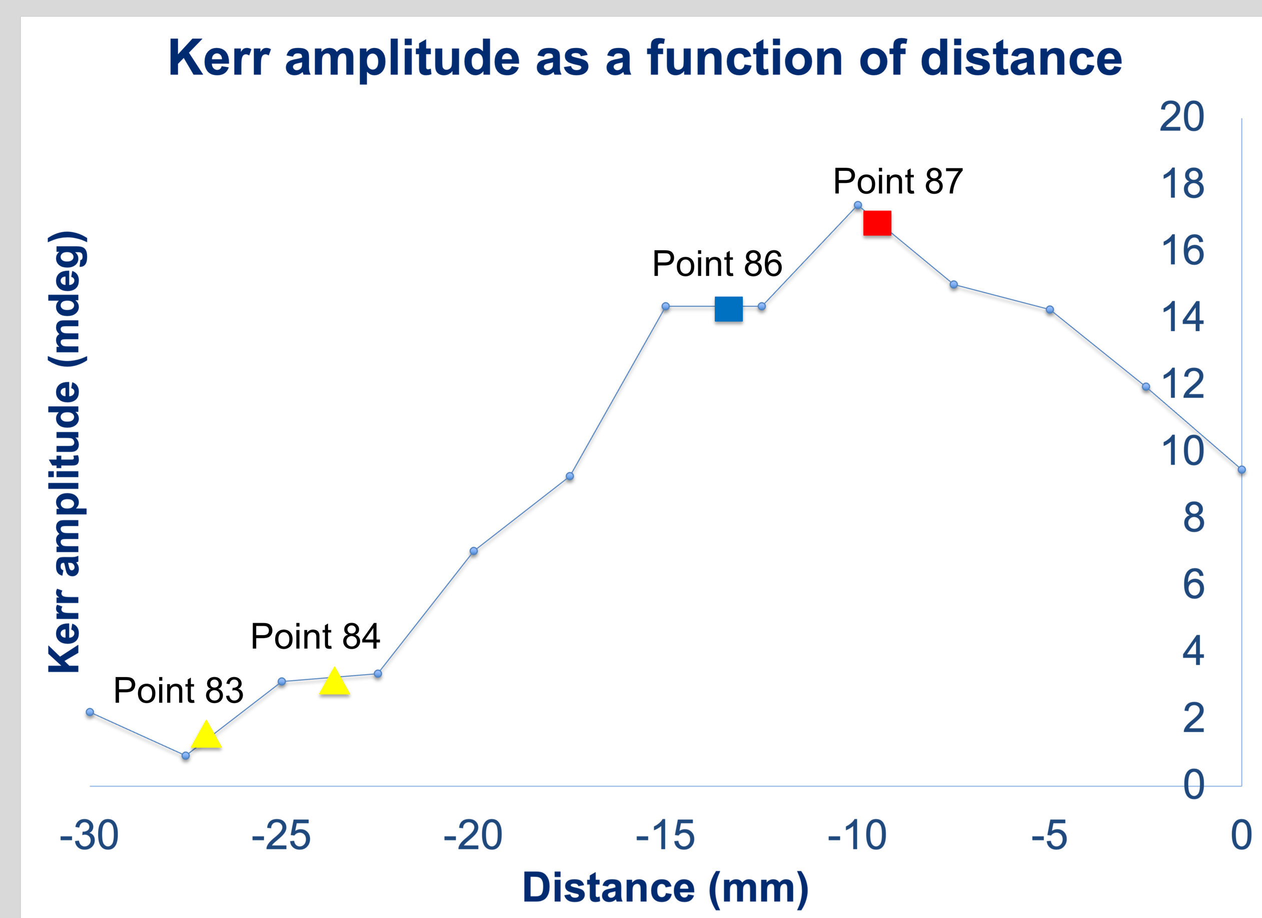


Figure 3. Kerr signal obtained from a line scan across the –x axis.

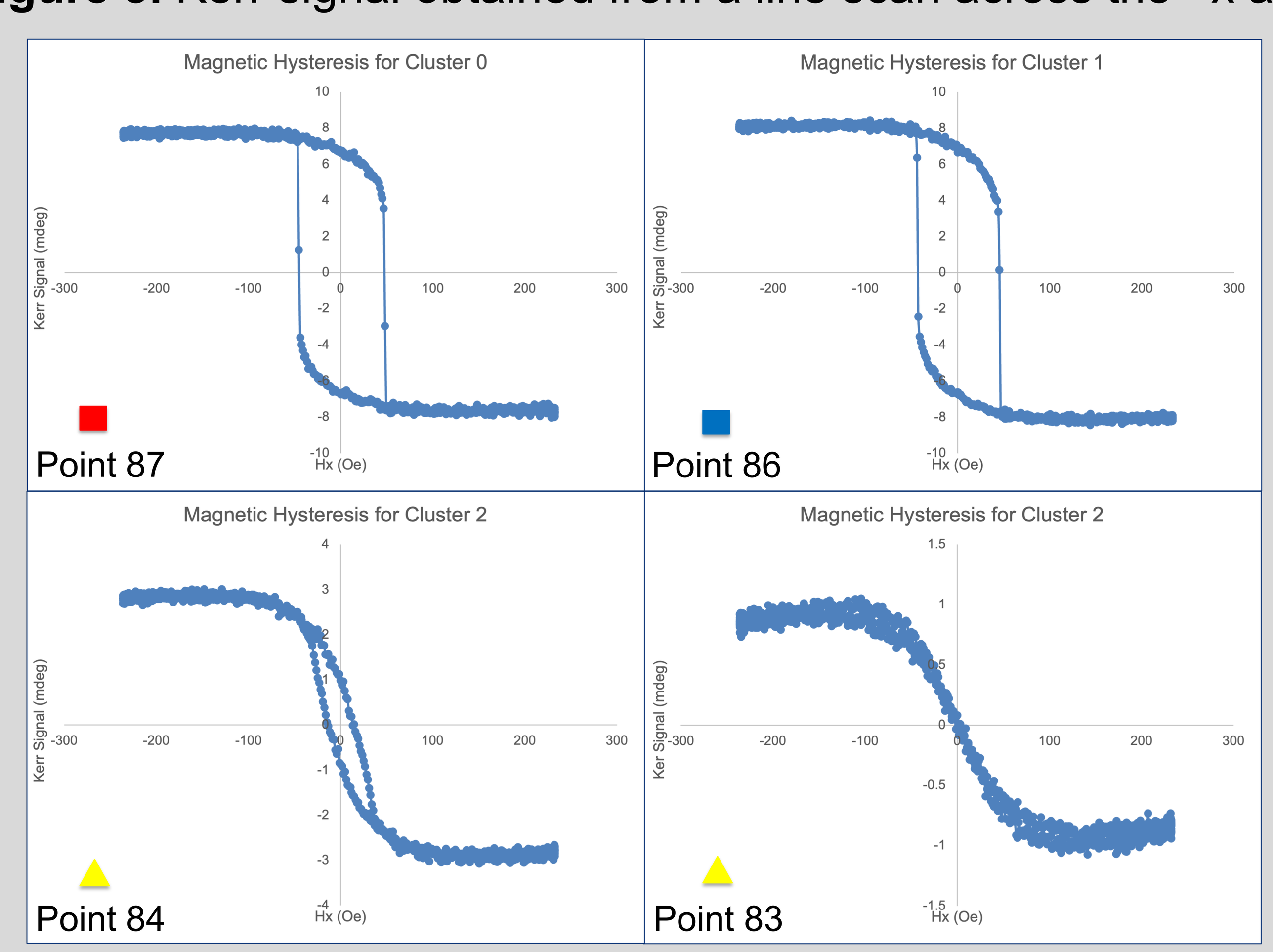


Figure 4. Evidence for different magnetic regimes in cluster groups.

* Magnetic field applied perpendicular to the flat of the wafer.

Composition Results

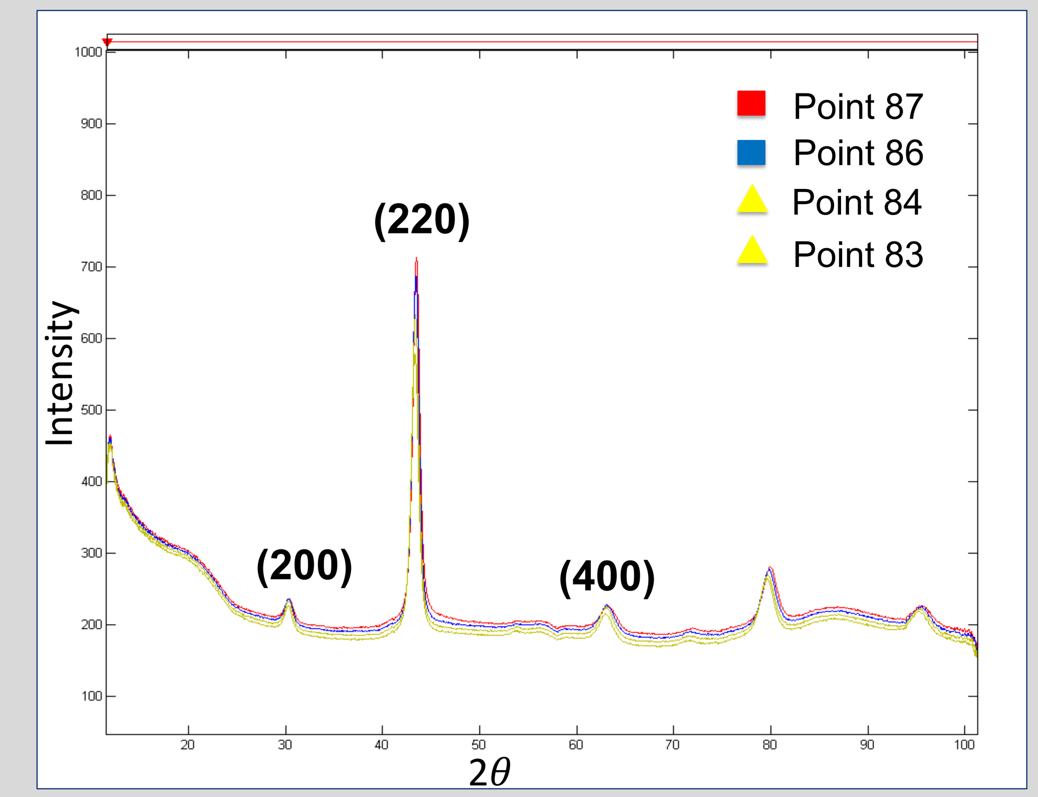


Figure 5. SLAC XRD spectra for region of interest.

There are no strong indicators of spinodal decomposition or secondary phase formation. Partial or short-range $L2_1$ ordering or B2 ordering, where the Co sublattice orders and Cr, Al sublattices disorder, are possible.

Changes in the relative intensities of the principal (220) peak may be linked to texture. Further analysis using EXAFS is needed.

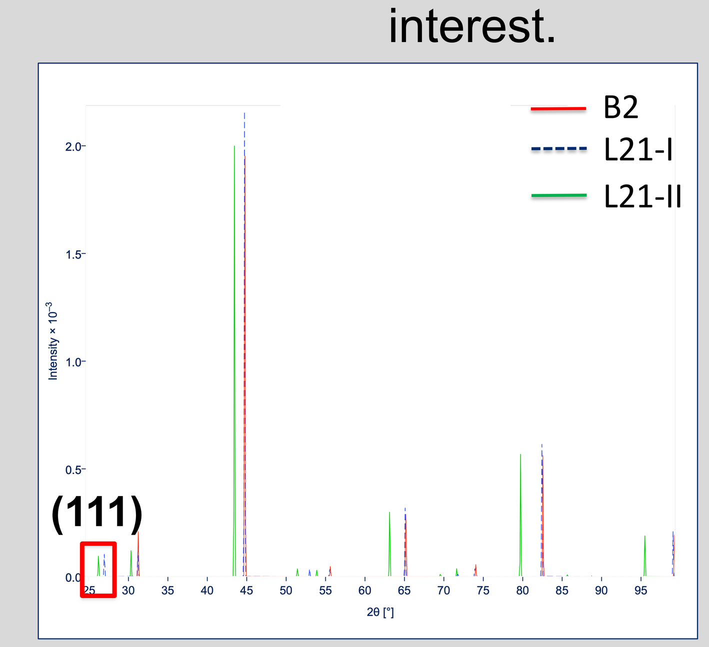


Figure 6. Theoretical XRD Spectra for $L2_1$ and B2 ordering [3].

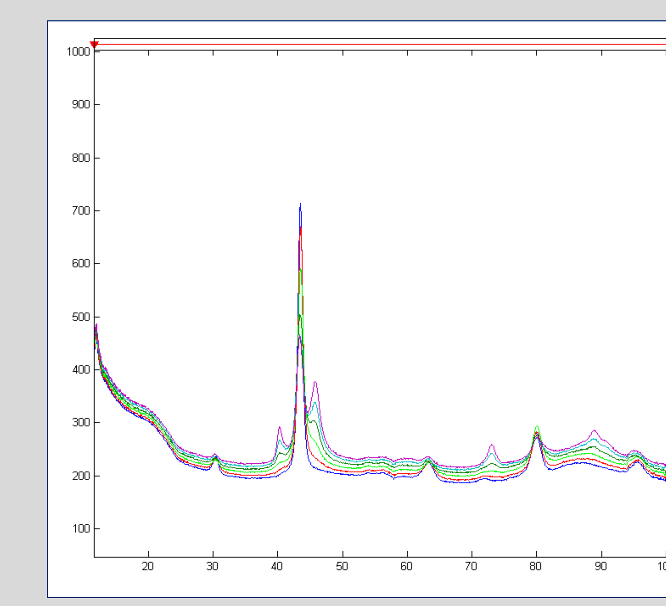


Figure 7. SLAC XRD spectra showing tetragonal and hexagonal phase precipitation.

Point 87, which exhibits the most robust magnetism, is the closest to the 2:1:1 stoichiometry.

Point	Co-At.%	Cr-At.%	Al-At.%
83	37.442	22.823	39.735
84	39.765	23.298	36.937
86	44.862	23.422	31.716
87	47.69	23.48	28.83

Table 1. XRF data for region of interest.

Conclusions and Future Plans

Located off-stoichiometric regions that avoid long-range order issues.
Measured different magnetic regimes relevant to changes in composition or structure and order consistent with the cluster groups from the ML model.
Achieved improved magnetism closest to the 2:1:1 stoichiometry where there are no strong indicators of secondary phase precipitation due to spinodal decomposition.
Future work: downselecting compositions with the most potential for exhibiting the least amount of B2 / most $L2_1$ for VSM, as well as introducing Fe to enhance magnetism.

References and Acknowledgements

[1] Omar, A., Boerrnert, F., Haft, M., Hampel, S., Loeser, W., Buechner, B., & Wurmehl, S. (2018). Crystal growth of off-stoichiometric Co2Cr1-xAl1+ x Heusler compounds: Avoiding the solid state miscibility gap. *Journal of Crystal Growth*, *498*, 103-108.
[2] E.S. Moskvitina, V.N. Kuznetsov, L.S. Guzei, Refinement of the Co–Cr–Al PhaseDiagram, Vestn. Mosk. Univ. Seriya 2 Khimiya (Moscow Univ. Chem. Bull.), vol. 33, 1992, pp. 373–374.
[3] Bergerhoff, G. & Brown, I.D. in „Crystallographic Databases“, F.H. Allen et al. (Hrsg.) Chester, International Union of Crystallography, (1987).

Thank you to Emily Holcombe, Dr. Mitra Taheri, and Dr. Orla Wilson for their continuous guidance and support.