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Utilising Unsupervised Machine Learning on Correlated EDS and 4DSTEM Data for Investigating the Structural Ordering Within Co_2FeSi Thin Films

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Co-based full Heusler alloys, Co_2FeSi (CFS) in particular, have been regarded as promising candidates for spintronics applications because of their full-spin polarization ability and high magnetic moment [1,2]. However, the structural ordering within these materials influences these desired magnetic properties immensely. In this study, the structural ordering of phases within CFS thin films was investigated using correlated EDS and 4DSTEM approaches. Unsupervised machine learning (ML) methods, including decomposition and clustering approaches [3] were utilized to provide insight into the structural and compositional variations within the films. Two known orderings, B2 ($Pm\bar{3}m$) and $L2_1$ ($Fm\bar{3}m$), were expected from previous XRD measurements, however, probabilistic fuzzy clustering-based ML applied to the combined compositional (EDS) and structural (4DSTEM) information indicated an unexpected additional phase. Figure 1 shows clustering outputs of a CFS thin film grown on Si(111). The cluster in Figure 1-a indicates the unexpected phase, evident by the highlighted reflection in its diffraction pattern, and the cluster in Figure 1-b indicates the B2 phase of this system, evident by the highlighted (002) reflection.

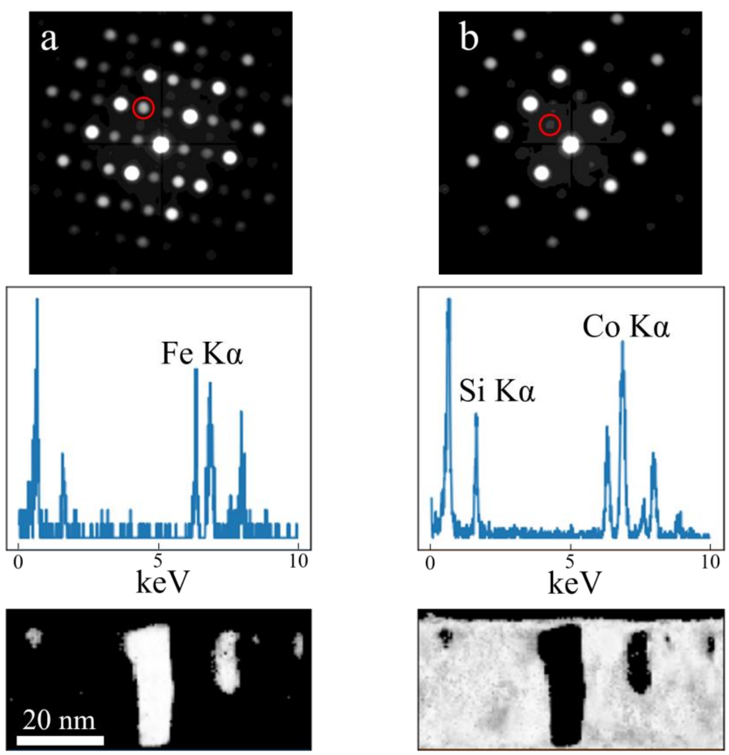

Figure 1. Clustering outputs of a CFS thin film. Top to bottom, the decomposed diffraction patterns in [110], accompanying EDS spectra and membership maps.

Each cluster output has its own decomposed diffraction pattern and EDS spectrum as well as a membership map which indicates the location of the cluster in the scan. Memberships in the figure show how these phases are complementary in the films. The unexpected phase has a different stacking of {111} lattice planes leading to different stoichiometry to both B2 and $L2_1$ phases, which is supported by the EDS cluster output belonging to this phase. Figure 2 demonstrates a representation of the crystal structure of this unexpected phase, modeled on these structural and compositional findings. DFT calculations based on this model support the stability of this structural ordering in Co-deficient samples and indicate that this minority superstructure could be the reason behind the variations in the magnetocrystalline anisotropy recorded in these thin films.

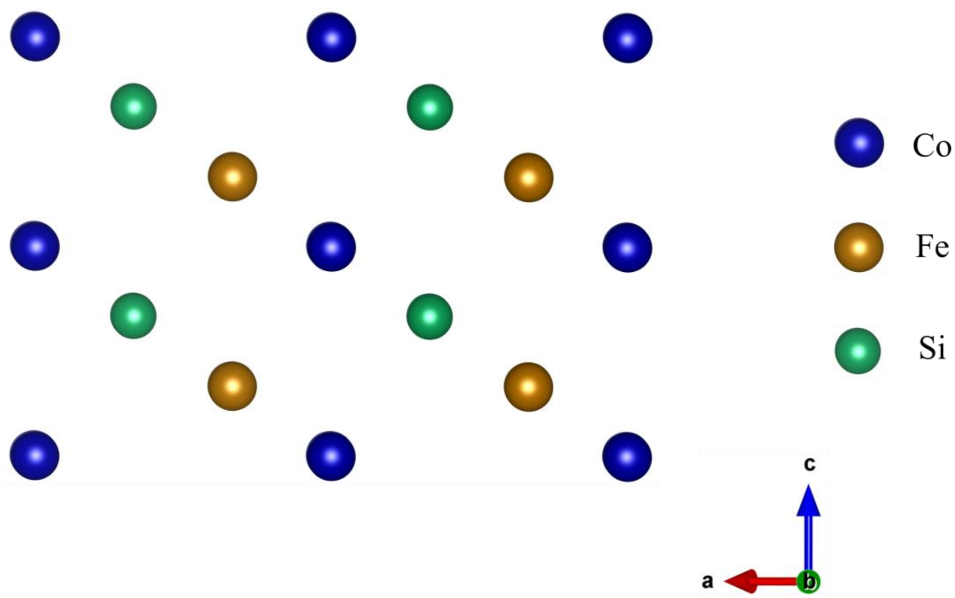

Figure 2. The modeled crystal structure of the unexpected phase.

Correlated signals allow an unprecedented amount of information to be extracted by exploring the covariance of the two orthogonal datasets (EDS and electron diffraction data in this case) and treating it as a single dataset in the ML analysis. This method provides a fast and reliable understanding of the complex microstructure of these materials [4].

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