Predictive Modelling of the Structure and Phase Stability of High-Entropy Materials: Case Study of $Al_xCrFeCoNi$

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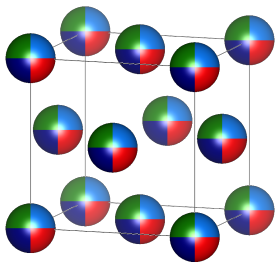
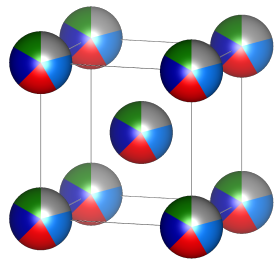
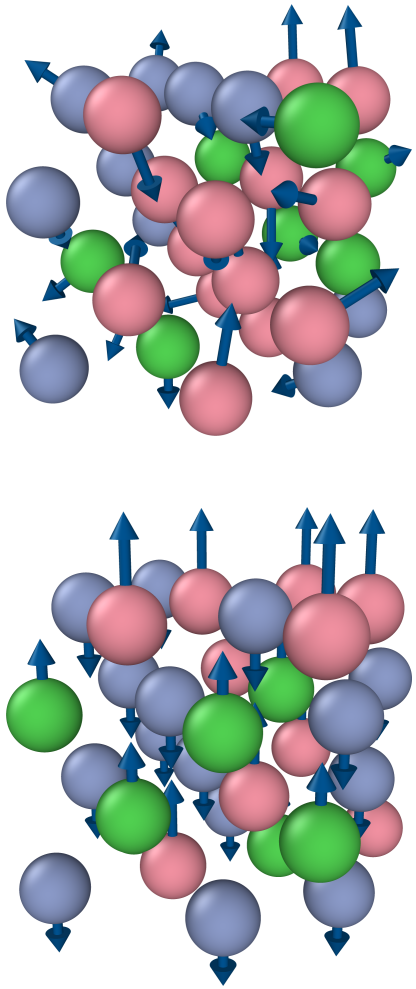




High-Entropy Materials: Modelling Challenges

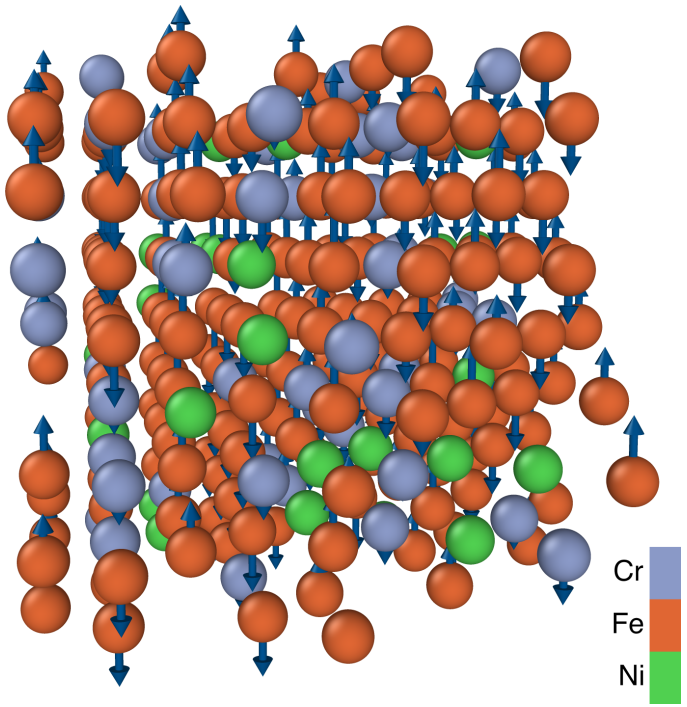
- Huge space of potential compositions, but most modelling results are specific to *one* particular composition.
- Even for one particular composition, need to inspect many possible atomic configurations.
- Magnetic elements: Fe, Mn, Ni, Co. Which magnetic state to model in DFT?



High-Entropy Materials: Modelling Solutions

A few options to model phase stability:

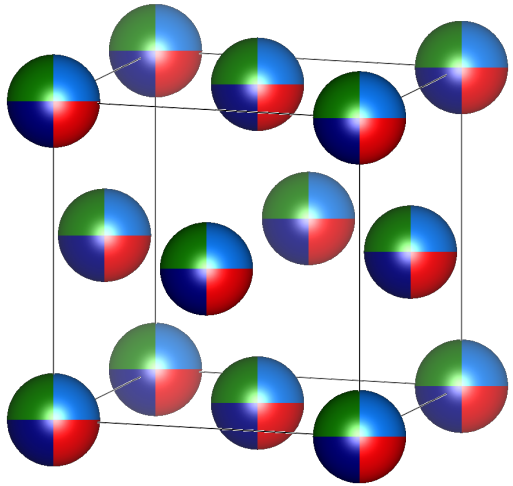
- 'Brute-force' DFT study: run DFT on many atomic configurations. *Hugely expensive*.
- Interatomic potentials, cluster expansions, MLIPs. *Cheaper, but still need DFT training data.*
- Effective medium theories: average over disorder in a (clever) physically meaningful way. *Cheap!*

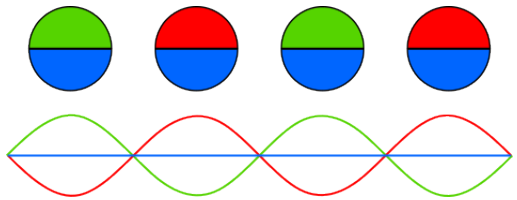


Our Description

Our approach uses the last option: an effective medium theory.

- Evaluate internal energy of fully disordered alloy within DFT using *Coherent Potential Approximation (CPA)*.
- Apply inhomogeneous chemical perturbation and assess energetic cost[1,2].
- Clever bit: do this using *concentration waves*, *i.e.* in **k**-space.





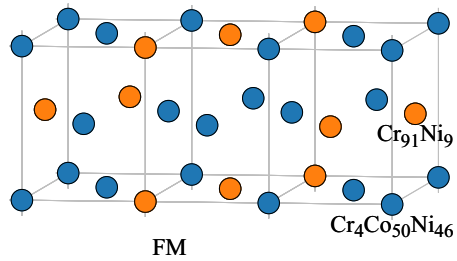
[1] C. D. Woodgate, J. B. Staunton, Phys. Rev. B **105** 115124 (2022)
[2] C. D. Woodgate, J. B. Staunton, Phys. Rev. Mater **7** 013801 (2023)

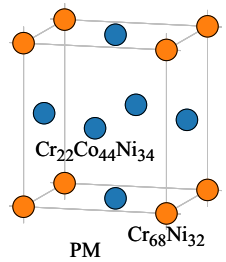
Inferring Orderings

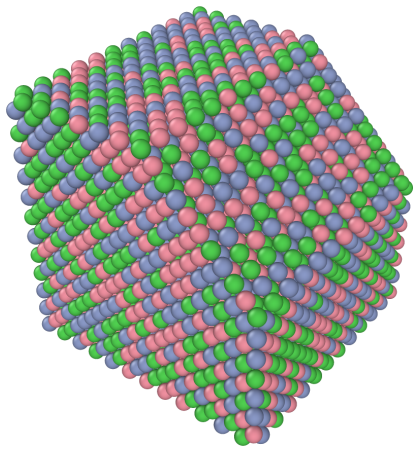
Two options:

- Option 1: infer orderings directly.
  - Perturbative analysis
    $\rightarrow$ Taylor expansion of Gibbs free energy.
- Option 2: lattice-based atomistic simulations.
  - Perturbative analysis
    $\rightarrow$ effective pair interactions.

$$H(\{\xi_{i\alpha}\}) = \frac{1}{2} \sum_{\substack{i\alpha \\ j\alpha'}} V_{i\alpha;j\alpha'}\, \xi_{i\alpha}\xi_{j\beta}$$





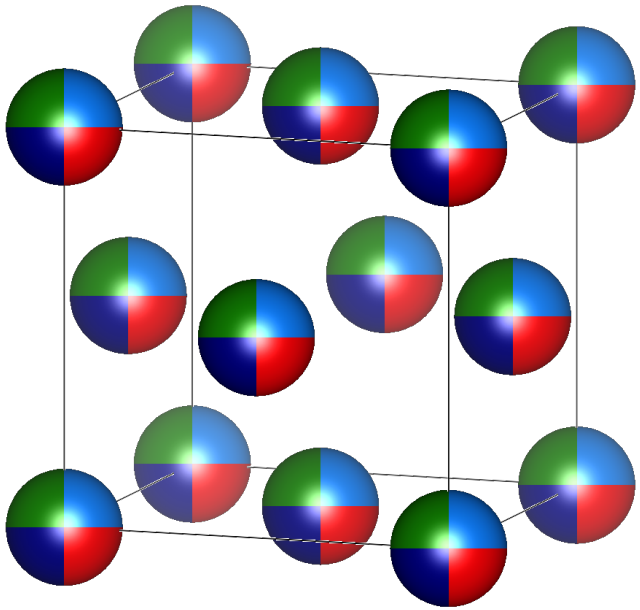


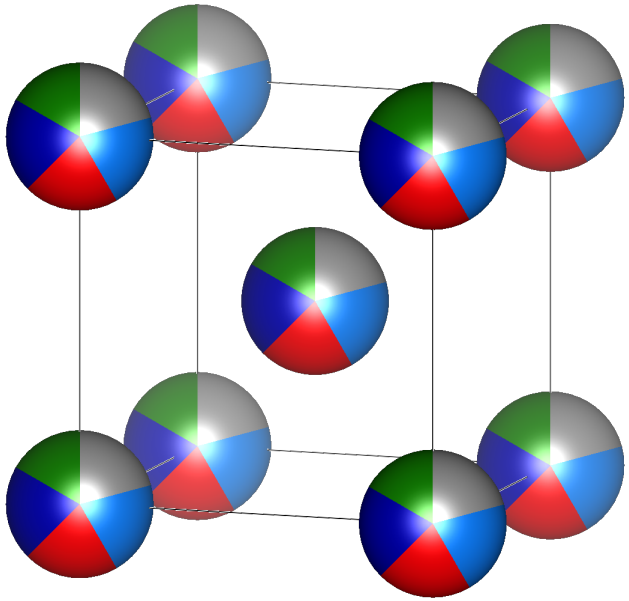
[3] Woodgate, Hedlund, Lewis, Staunton, Phys. Rev. Mater **7** 053801 (2023)

Successful Applications

- CrMnFeCoNi and derivatives.
  - C. D. Woodgate, J. B. Staunton, Phys. Rev. B **105** 115124 (2022).
- VNbMoTaW and derivatives.
  - C. D. Woodgate, J. B. Staunton, Phys. Rev. Mater. **7** 013801 (2023).
- Influcence of Magnetism on Atomic Ordering.
  - C. D. Woodgate, D. Hedlund, L. H. Lewis, J. B. Staunton, Phys. Rev. Mater. **7**, 053801 (2023).
- Influence of Ti additions: Ti_xVNbMoTaW
  - C. D. Woodgate, J. B. Staunton, J Appl. Phys. **135** 135106 (2024).
- Al_xCrFeCoNi 'Superalloy'.
  - C. D. Woodgate, G. A. Marchant, L. B. Pártay, J. B. Staunton, arXiv:2404.01373.

Case Study: $Al_xCrFeCoNi$

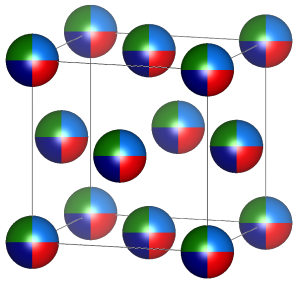


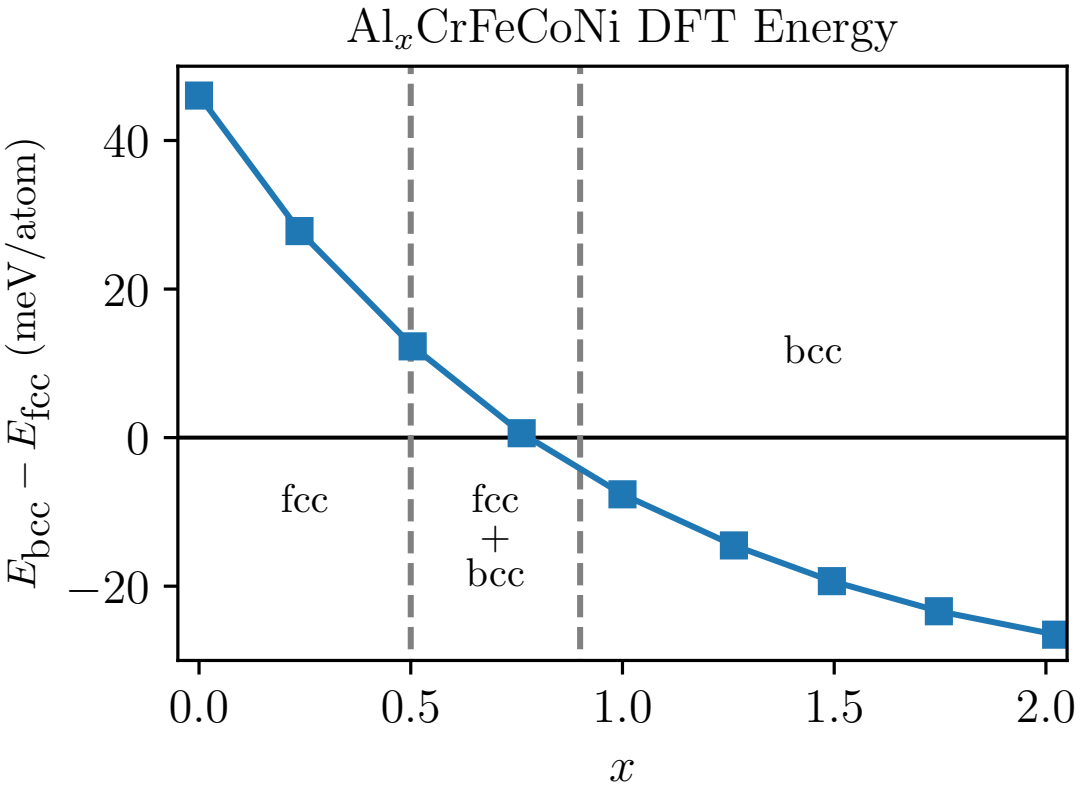


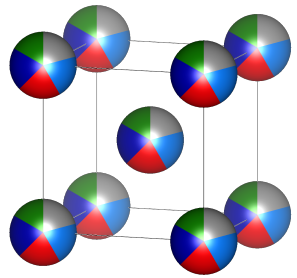
[3] Woodgate, Marchant, Pártay, Staunton, arXiv:2404.13173

$Al_xCrFeCoNi$: Underlying Lattice

Hutsepot

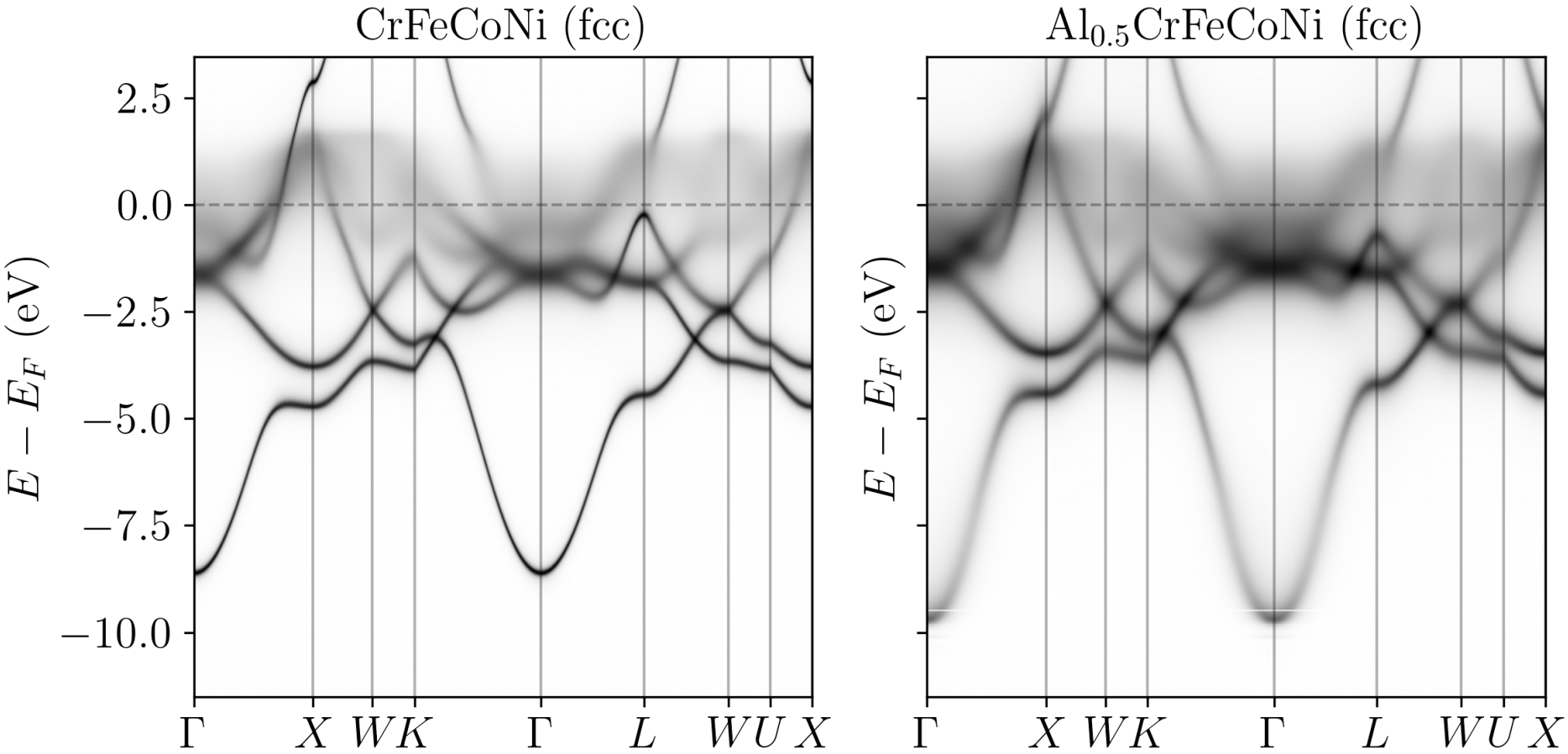






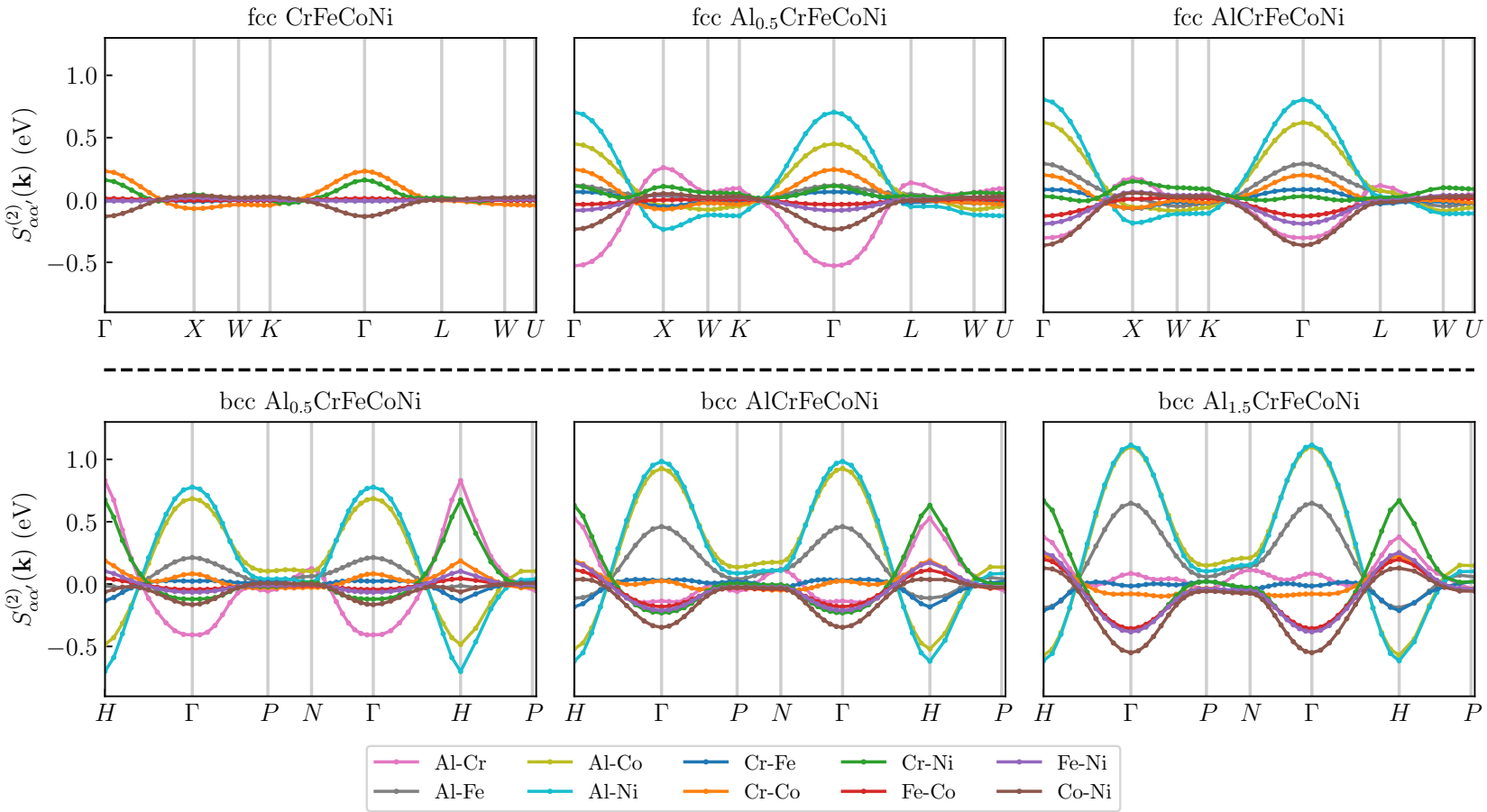
[3]Woodgate, Marchant, Pártay, Staunton, arXiv:2404.13173

$Al_xCrFeCoNi$: Electronic Structure



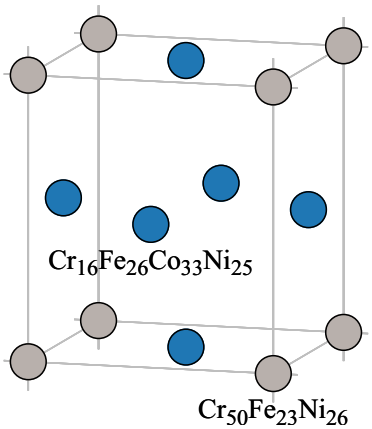
[3] Woodgate, Marchant, Pártay, Staunton, arXiv:2404.13173

$Al_xCrFeCoNi$: Perturbative Analysis



[3] Woodgate, Marchant, Pártay, Staunton, arXiv:2404.13173

$Al_xCrFeCoNi$: Inferred Orderings



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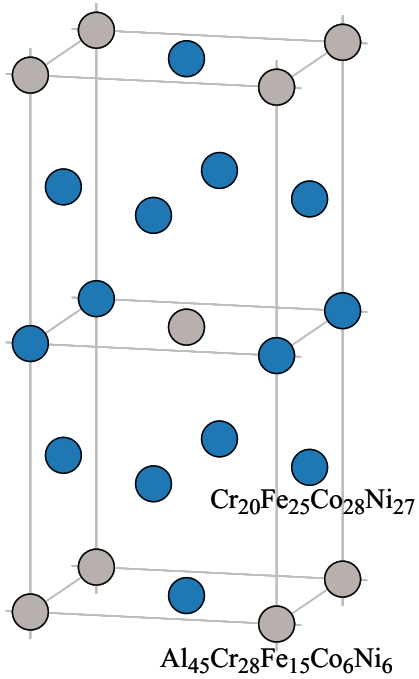
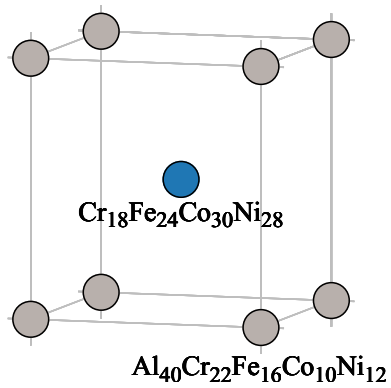
$L1_2$



$x = 0.5$
$D0_{22}$

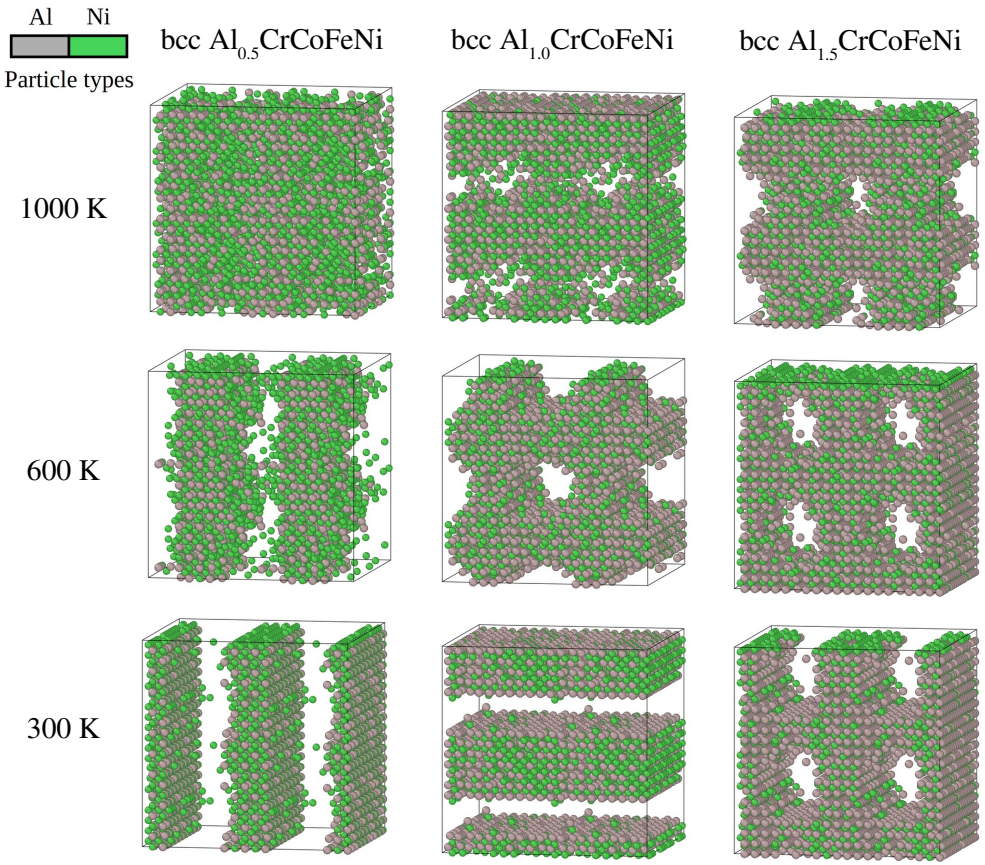


$x = 1$
B2

[3] Woodgate, Marchant, Pártay, Staunton, arXiv:2404.13173

$Al_xCrFeCoNi$: Atomistic Modelling

(Cell duplicated $2 \times 2 \times 2$ times for clarity.)



[3] Woodgate, Marchant, Pártay, Staunton, arXiv:2404.13173

Take-Home Messages

Coherent Potential Approximation (CPA)

CPA provides a *powerful* tool for modelling the electronic structure of disordered, high-entropy systems for minimal computational cost.

Perturbative Analysis

Analysis of energetic cost of chemical fluctuations applied to homogeneous CPA medium can tell us about atomic ordering tendencies.

Superb Agreement with Experiment

For < 1000 core-hours, can predict phase behaviour across range of compositions and temperatures.

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Our paper: arXiv:2404.13173

People

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- George A. Marchant
- Julie B. Staunton

Department of Chemistry, University of Warwick, UK

- Livia B. Pártay

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