Magnetism Matters: Modelling Atomic Arrangements in Multicomponent Alloys

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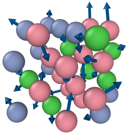

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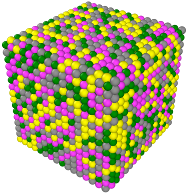

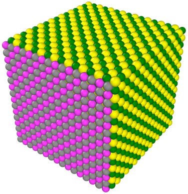

- At what temperature will order emerge? What is the nature of order? Short-range? Long-range? Materials properties?

Challenge for Modellers

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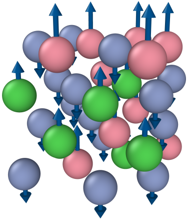

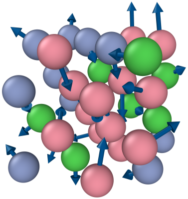

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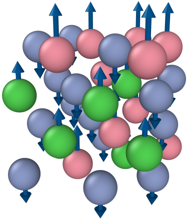
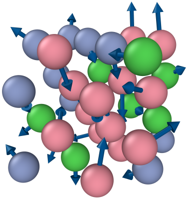
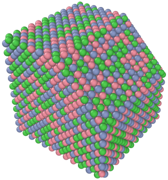


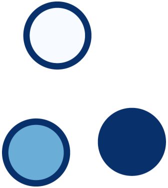


- Would like a *computationally efficient* modelling approach to assess phase stability.

Our Description

- On lattice.





[1]Woodgate, Staunton, Phys. Rev. B **105** 115124 (2022)
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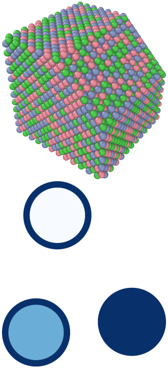

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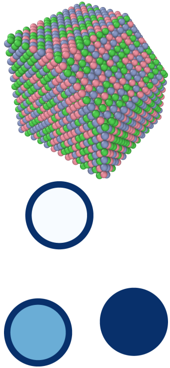

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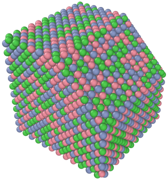
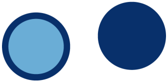


$$c_{i\alpha} \equiv \langle \xi_{i\alpha} \rangle$$

- Perturb high-T, homogeneous state $c_{i\alpha} = c_\alpha + \Delta c_{i\alpha}$ and see what favourable correlations are[1,2].



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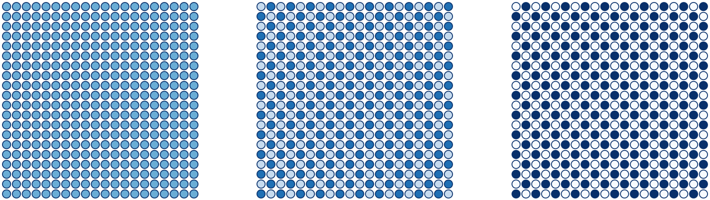
Concentration Waves

$$c_{i\alpha} = c_\alpha + \eta_\alpha \frac{1}{2} \left(e^{i\mathbf{q}\cdot\mathbf{R}_i} + e^{-i\mathbf{q}\cdot\mathbf{R}_i} \right), \quad \mathbf{q} = \left(\tfrac{1}{2}, \tfrac{1}{2}\right).$$



$\eta = (0, 0)$ $\eta = (0.25, -0.25)$ $\eta = (0.5, -0.5)$

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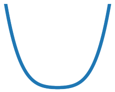
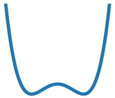
Energetics: First-Principles

- Evaluate cost of fluctuations *ab initio* via DFT, using KKR-CPA and a linear response theory[1][2][3].





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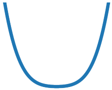
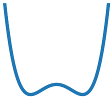
- Mean-field free energy:

$$G = \beta^{-1} \sum_{i\alpha} c_{i\alpha} \log c_{i\alpha} - \sum_{i\alpha}{}' \nu_{i\alpha} c_{i\alpha} + \langle \Omega_{\text{el}} \rangle_0 [\{c_{i\alpha}\}]$$





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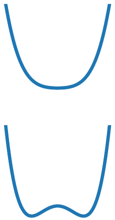

- Important quantity:

$$S^{(2)}_{i\alpha; j\alpha'} \equiv \frac{\partial^2 \langle \Omega_{\text{el}} \rangle_0}{\partial c_{i\alpha} \partial c_{j\alpha'}} \rightsquigarrow \Psi^{-1}_{\alpha\alpha'}(\mathbf{q})$$

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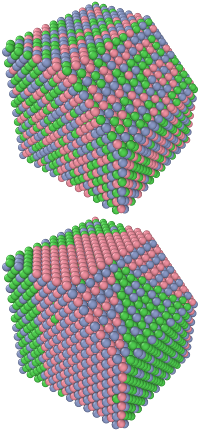
Energetics: Interatomic Potential

- Bragg-Williams Hamiltonian for atomistic modelling:

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



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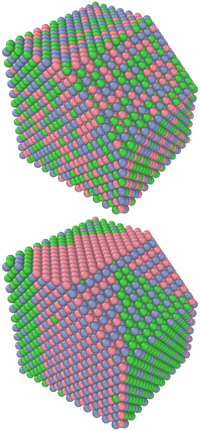

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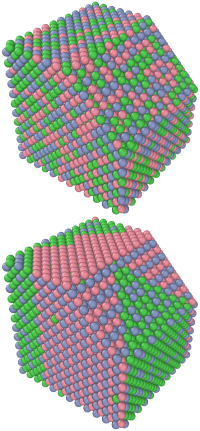
- If H as above, $V_{i\alpha; j\alpha'} = -S^{(2)}_{i\alpha; j\alpha'}$ *exactly*.
- **Generate physically-motivated configurations for subsequent study.**



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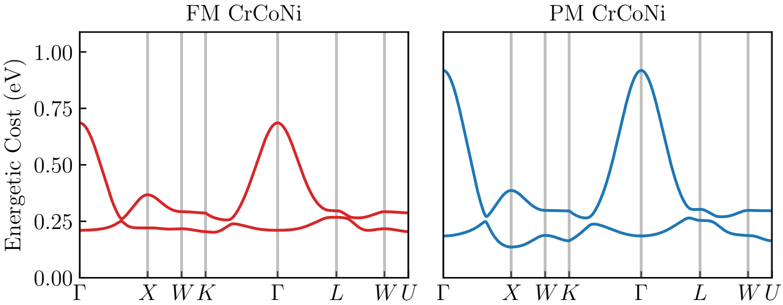
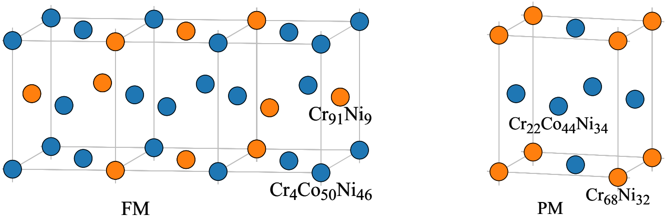
CrCoNi: Linear Response



- Shape of modes *and* location of minimum altered.

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

CrCoNi: Inferred Orderings



- Different predicted chemical orderings based on magnetic state! Can we observe this experimentally in some systems?

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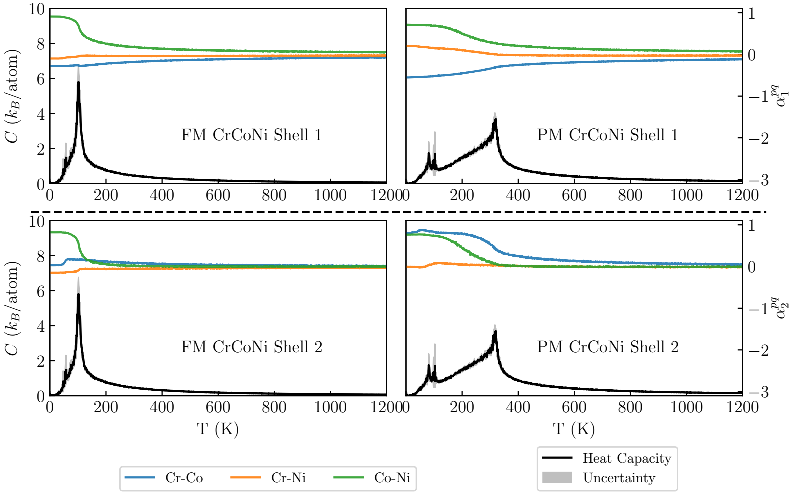
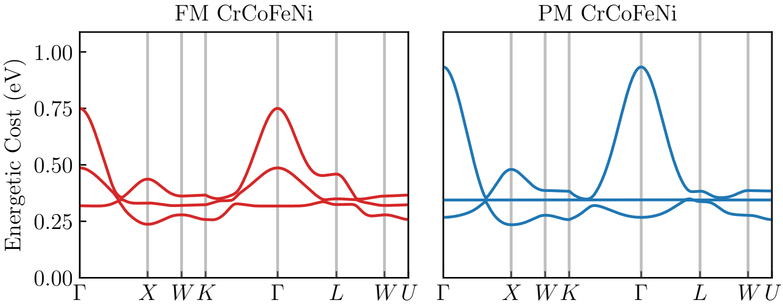
CrCoNi: Atomistic Modelling



CrFeCoNi: Linear Response



- Correlations involving Fe significantly strengthened.

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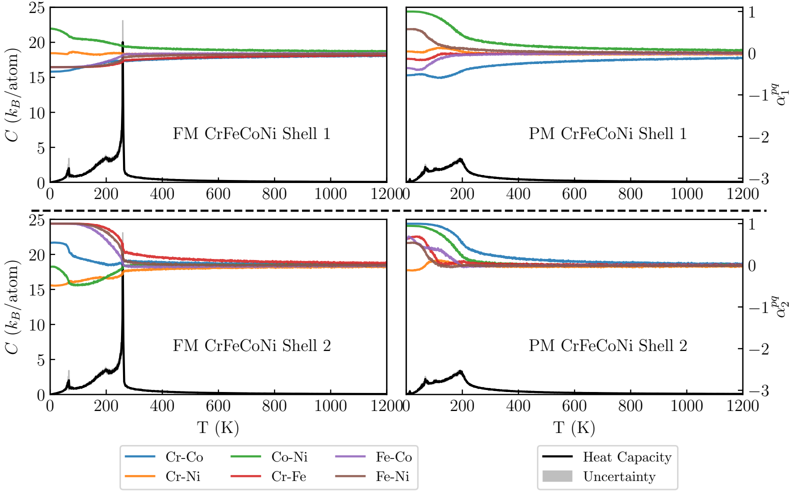
CrFeCoNi: Atomistic Modelling



Next Steps and Future Work

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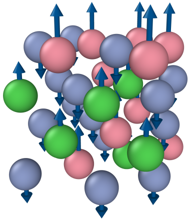
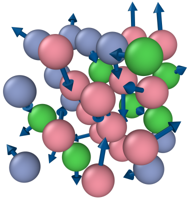
- Feed into more sophisticated techniques, *e.g.* use rapidly-generated configurations in training sets for machine-learned interatomic potentials[5].





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Take-Home Messages

When Modelling Alloys, Magnetism is *Important*

Nature of the magnetic state in an alloy can alter strength of interactions/correlations between elements.

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Can some multicomponent alloys be processed in an applied magnetic field to tune atomic ordering?

Interface with other techniques

Can use computationally efficient approach to generate configurations for subsequent studies.

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- Julie B. Staunton

Northeastern University, USA

- Laura H. Lewis

Our paper:
Woodgate, Hedlund, Lewis, Staunton, Phys. Rev. Mater. **7** 053801 (2023)