Magnetism Matters: Modelling Atomic Arrangements in Multicomponent Alloys

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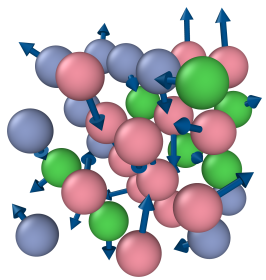
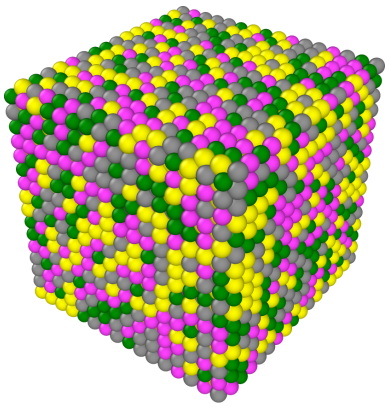
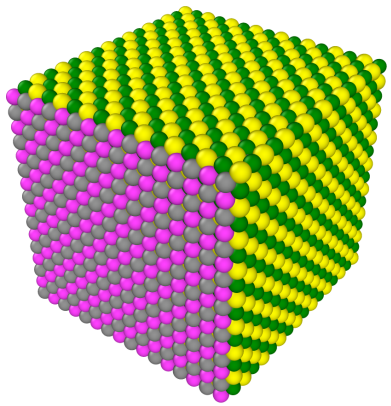


Multicomponent Alloys

- Steels, *e.g.* $Fe_{70}Cr_{20}Ni_{10}$.
- High Entropy Alloys (HEAs), *e.g.* CrMnFeCoNi, CrCoNi, NbMoTaW.
- HEAs: solid solution stabilised by large "entropy of mixing"

$$TS = -k_B T \sum_{\alpha} c_\alpha \log c_\alpha. \tag{1}$$

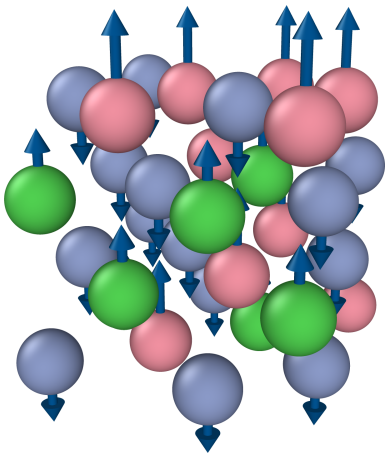


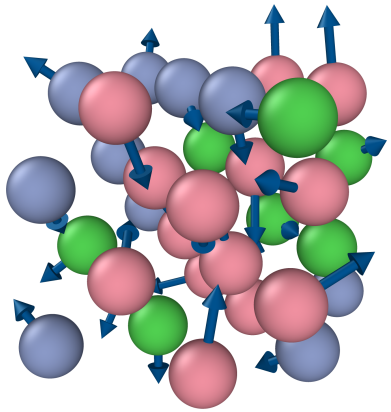


- At what temperature will order emerge? What is the nature of order? Short-range? Long-range? Materials properties?

Challenge for Modellers

- Space of possible atomic configurations is *vast*. Challenges conventional, supercell-based techniques.
- For alloys containing magnetic elements, *e.g.* CrCoNi, how should magnetism be treated?



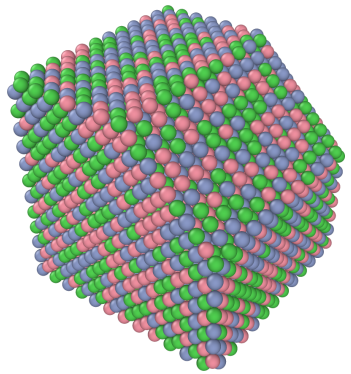


- Would like a *computationally efficient* modelling approach to assess phase stability.

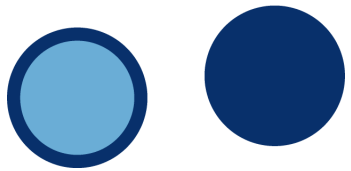
Our Description

- On lattice.
- Specify configuration by $\{\xi_{i\alpha}\}$.
- Interested in the *average* value of these, *i.e.* partial occupancies:

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



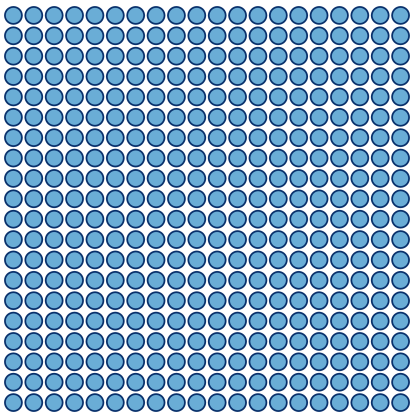
- Perturb high-T, homogeneous state $c_{i\alpha} = c_\alpha + \Delta c_{i\alpha}$ and see what favourable correlations are[1][2].



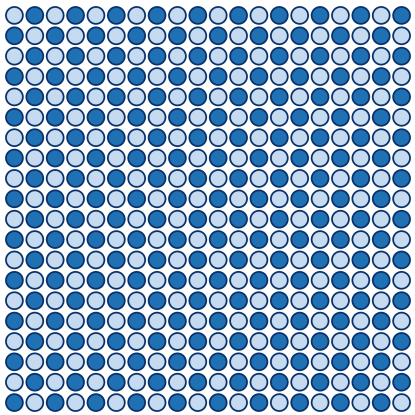
[1] Woodgate, Staunton, Phys. Rev. B **105** 115124 (2022)
[2] Woodgate, Staunton, Phys. Rev. Mater. **7** 013801 (2023)

Concentration Waves

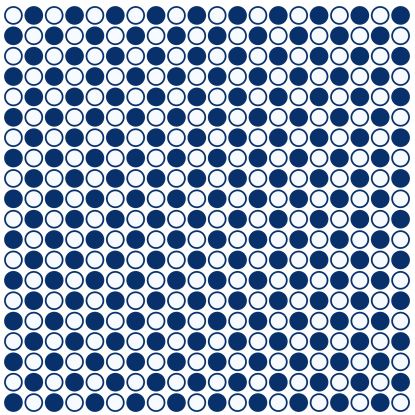
$$c_{i\alpha} = c_\alpha + \eta_\alpha \frac{1}{2}\left(e^{i\mathbf{q}\cdot\mathbf{R}_i} + e^{-i\mathbf{q}\cdot\mathbf{R}_i}\right), \quad \mathbf{q} = \left(\tfrac{1}{2}, \tfrac{1}{2}\right).$$



$\eta = (0, 0)$



$\eta = (0.25, -0.25)$



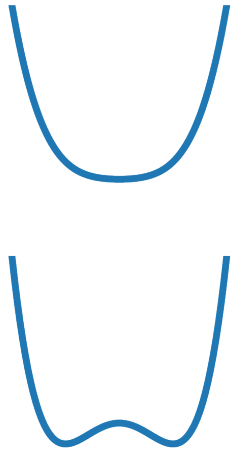
$\eta = (0.5, -0.5)$

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

Energetics: First-Principles

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

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



- Important quantity:

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

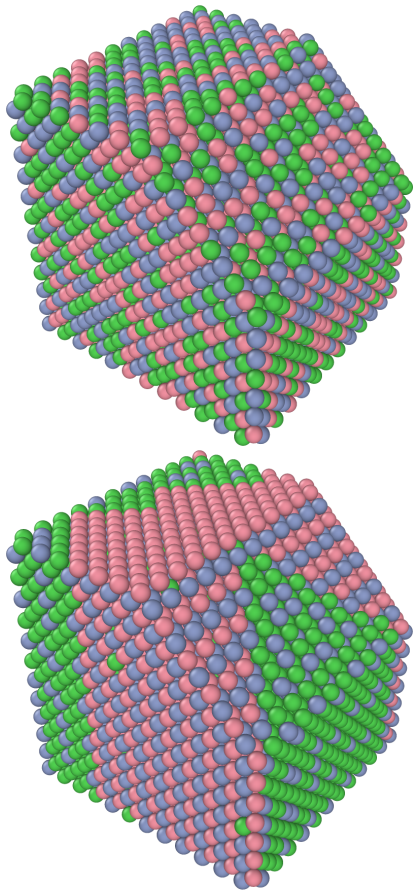
[1] Woodgate, Staunton, Phys. Rev. B **105** 115124 (2022)
[2] Woodgate, Staunton, Phys. Rev. Mater. **7** 013801 (2023)
[3] Khan, Staunton, Stocks, Phys. Rev. B **93** 054206 (2016)

Energetics: Interatomic Potential

- Bragg-Williams Hamiltonian for atomistic modelling:

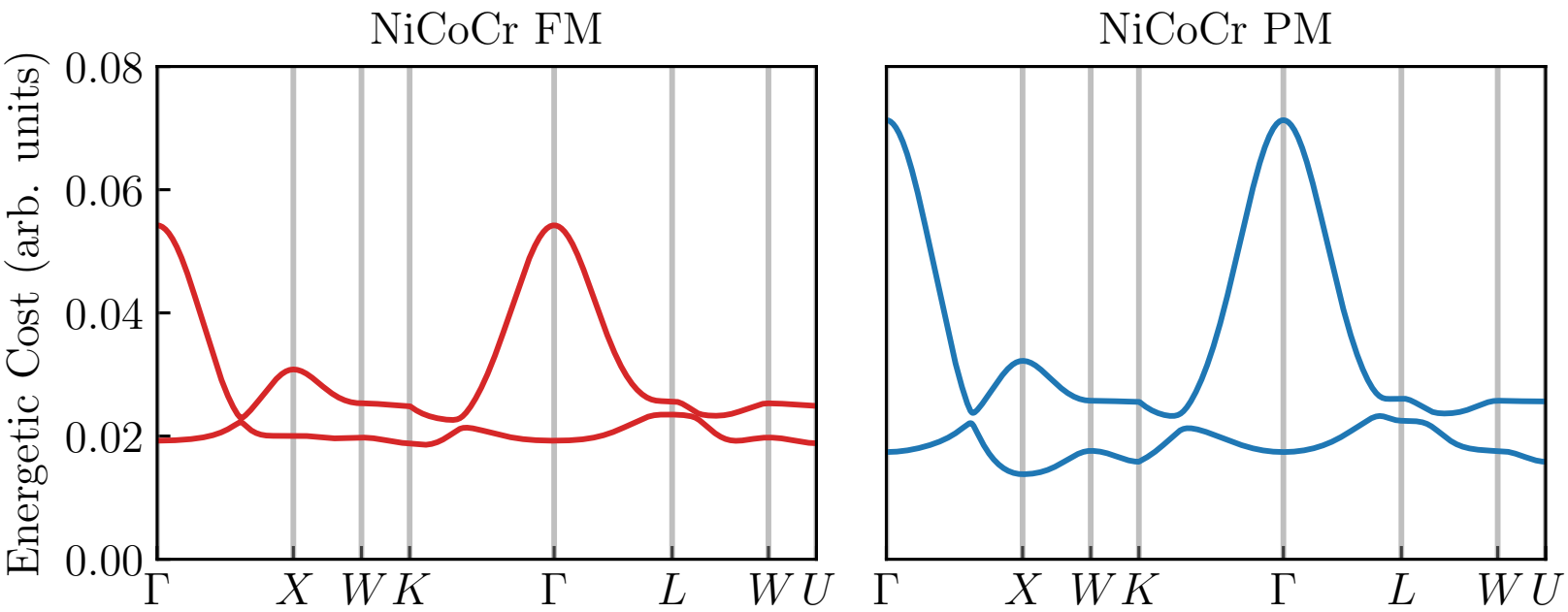
$$H(\{\xi_{i\alpha}\}) = \frac{1}{2} \sum_{\substack{i\alpha \\ j\alpha'}} V_{i\alpha;j\alpha'}\, \xi_{i\alpha} \xi_{j\beta}$$

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



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

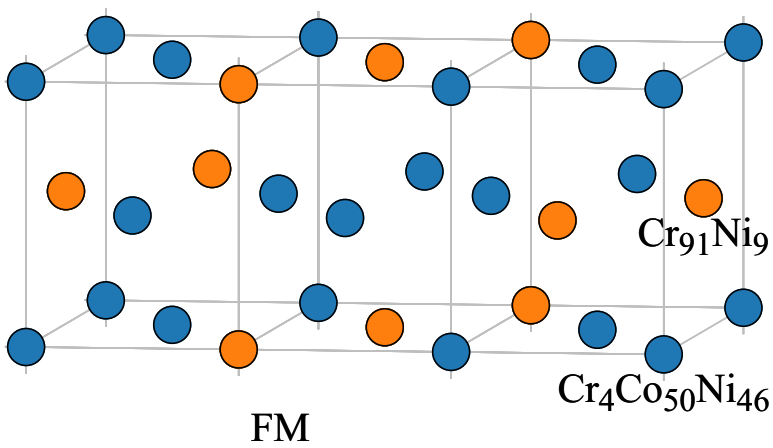
CrCoNi: Linear Response

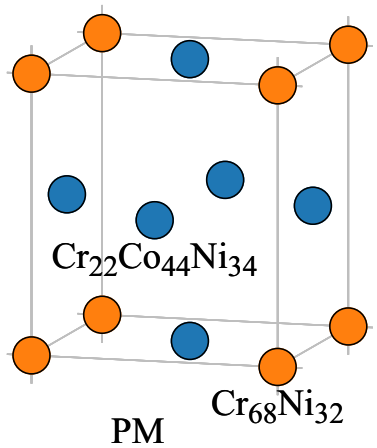


- Shape of modes *and* location of minimum altered.

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

CrCoNi: Inferred Orderings

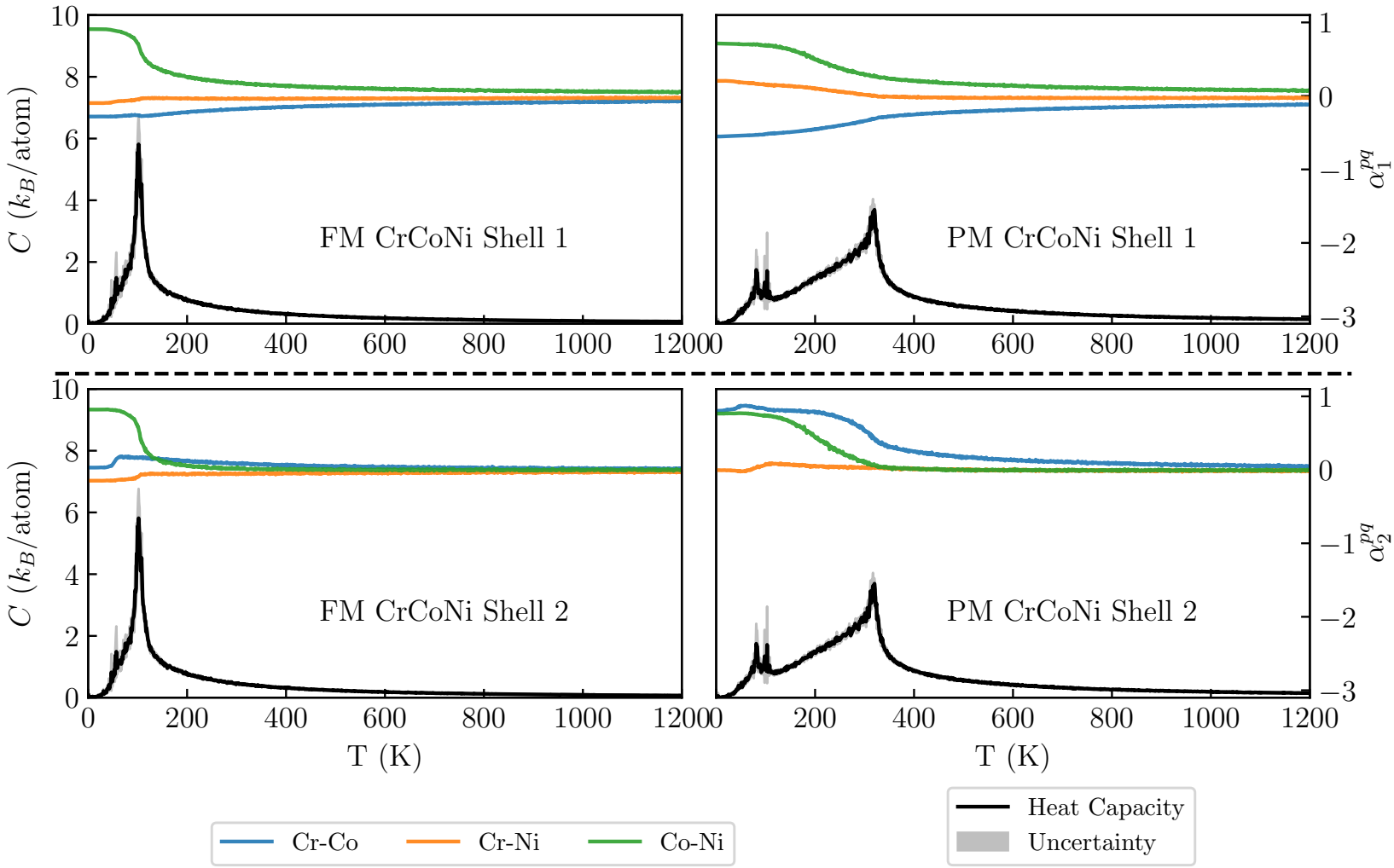




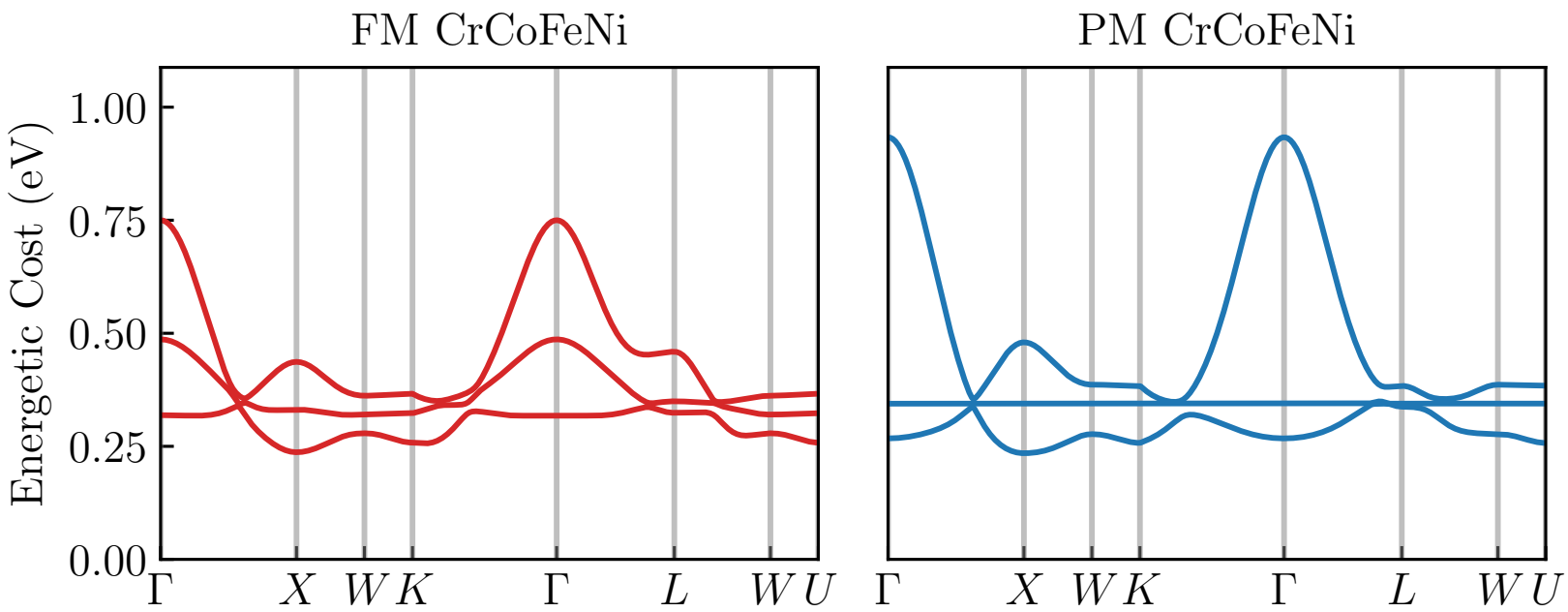
- Different predicted chemical orderings based on magnetic state! Can we observe this experimentally in some systems?

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

CrCoNi: Atomistic Modelling



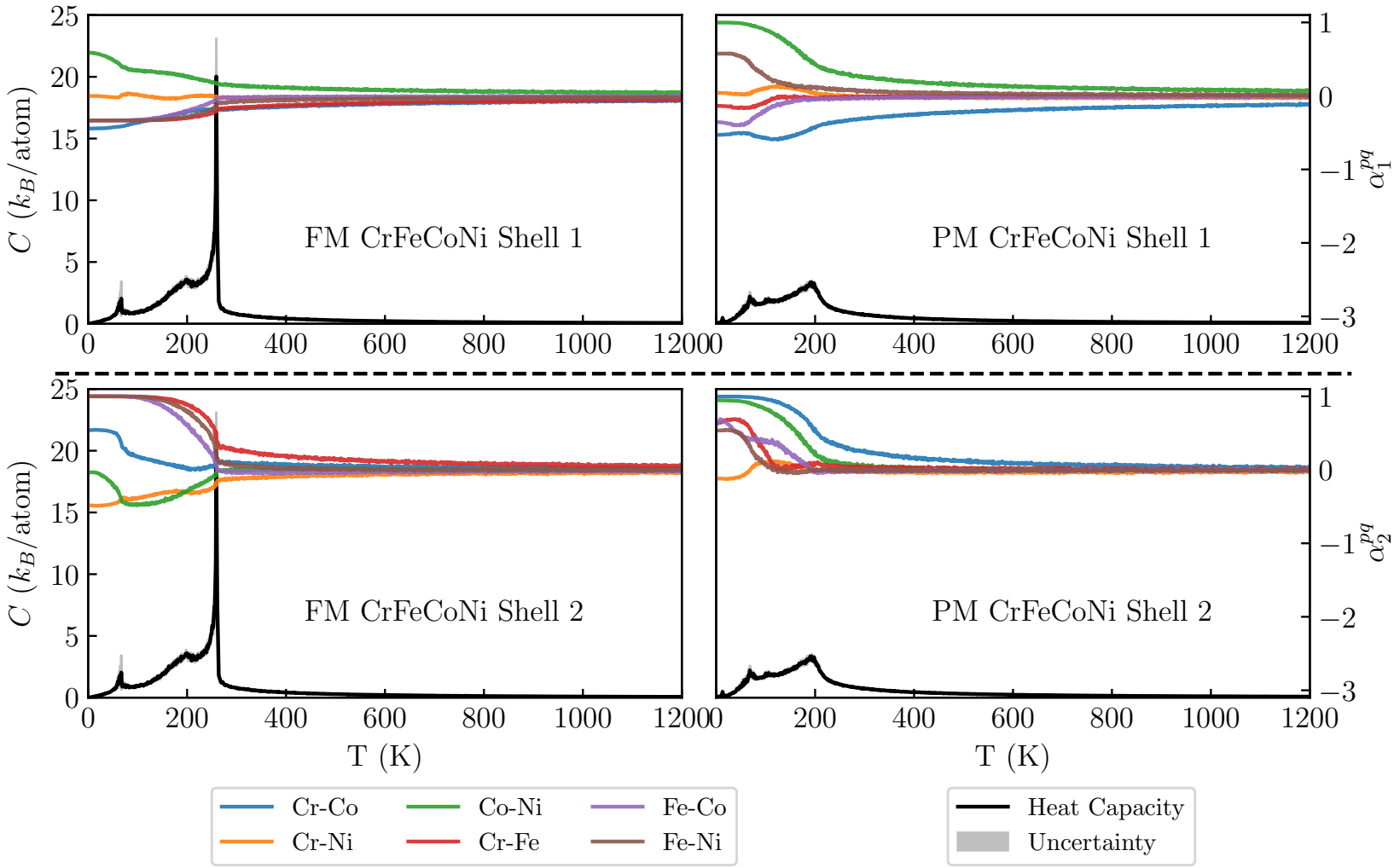
CrFeCoNi: Linear Response



- Correlations involving Fe significantly strengthened.

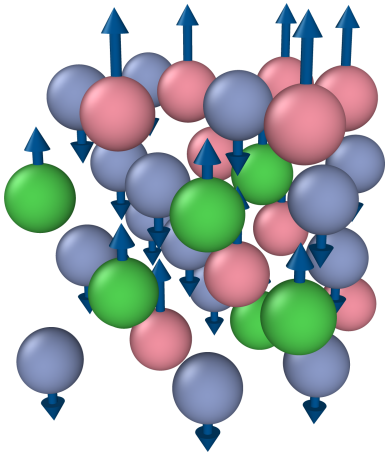
[4] Woodgate, Hedlund, Lewis, Staunton, Phys. Rev. Mater. **7** 053801 (2023)

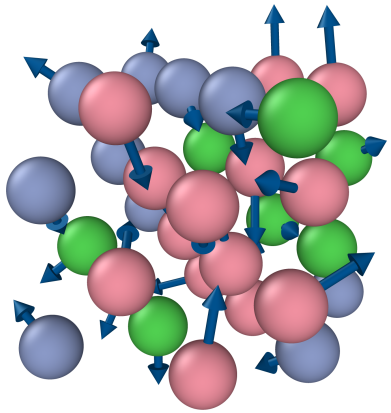
CrFeCoNi: Atomistic Modelling



Next Steps and Future Work

- Multicomponent alloys represent a *huge* playground.
- Approach is highly computationally efficient; all figures shown today can be reproduced in < 1000 CPU-hours. Materials discovery?
- Feed into more sophisticated techniques, *e.g.* use rapidly-generated configurations in training sets for machine-learned interatomic potentials[5].





[5] Shenoy, Woodgate, Staunton, Bartók, Kermode, in preparation.

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.

Experimental Implications

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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