Connections Between Magnetism and Preferred Atomic Arrangements in Multicomponent Alloys

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Context ●	Modelling Approach	Results 00000	Next Steps O	Conclusions

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Context ●	Modelling Approach	Results 00000	Next Steps O	Conclusions

► Steels, *e.g.* Fe₇₀Cr₂₀Ni₁₀.

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Steels, *e.g.* Fe₇₀Cr₂₀Ni₁₀.

► High Entropy Alloys (HEAs), *e.g.* CrMnFeCoNi, VNbMoTaW.

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Co

- ► Steels, *e.g.* Fe₇₀Cr₂₀Ni₁₀.
- ► High Entropy Alloys (HEAs), *e.g.* CrMnFeCoNi, VNbMoTaW.





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

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- ► High Entropy Alloys (HEAs), *e.g.* CrMnFeCoNi, VNbMoTaW.





- At what temperature will order emerge? What is the nature of order? Short-range? Long-range? Materials properties?
- Challenging for modellers. Huge number of potential compositions and atomic configurations. Magnetic state for alloys containing Fe, Mn, Co?

Context O	Modelling Approach ●000	Results 00000	Next Steps O	Conclusions

Have lots of lattice-based configurations.



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

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Context O	Modelling Approach ●000	Results 00000	Next Steps o	Conclusions

- Have lots of lattice-based configurations.
- Specify these by $\{\xi_{i\alpha}\}$.



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- Have lots of lattice-based configurations.
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- Interested in the average value of these, i.e. work with partial occupancies:

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



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

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$$c_{ilpha}\equiv \langle \xi_{ilpha}
angle$$

Construct an effective medium representing the average electronic structure of the disordered alloy; use the KKR-CPA to do this¹².

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 ²Woodgate, Staunton, Phys. Rev. Mater. **7** 013801 (2023)

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Key Idea			00000	

Perturb high-*T*, homogeneous state c_{iα} = c_α + Δc_{iα} and see what favourable correlations are¹².



¹Woodgate, Staunton, Phys. Rev. B 105 115124 (2022)
 ²Woodgate, Staunton, Phys. Rev. Mater. 7 013801 (2023)
 ³Khan, Staunton, Stocks, Phys. Rev. B 105 115124 (2022)

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Key Idea			00	00000

- Perturb high-*T*, homogeneous state c_{iα} = c_α + Δc_{iα} and see what favourable correlations are¹².
- Take advantage of translational symmetry and do this in reciprocal space.

$$c_{ilpha} = c_{lpha} + \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{R}_i}\Delta c_{lpha}(\mathbf{k})$$
 (1)



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 ²Woodgate, Staunton, Phys. Rev. Mater. **7** 013801 (2023)
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$$c_{i\alpha} = c_{\alpha} + \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{R}_i}\Delta c_{\alpha}(\mathbf{k})$$
 (1)

 Get energetic costs via a perturbative analysis of CPA reference medium³; think DFPT.

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 ²Woodgate, Staunton, Phys. Rev. Mater. **7** 013801 (2023)
 ³Khan, Staunton, Stocks, Phys. Rev. B **105** 115124 (2022)

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Understanding Ordering Tendencies

Two options:

 Apply Landau theory to free energy constructed via perturbative analysis¹².

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Results 00000 Next Steps

Understanding Ordering Tendencies

Two options:

- Apply Landau theory to free energy constructed via perturbative analysis¹².
- Map derivatives of internal energy on to pairwise atomistic model:

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



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 Generate physically-motivated configurations for subsequent study.

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

²Woodgate, Staunton, Phys. Rev. Mater. **7** 013801 (2023)

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Context N O C	lodelling Approach ○○○●	Results 00000	Next Steps O	Conclusions
Successful A	pplications			
CrMnF	eCoNi and deriva	tives.		
C. Ph	D. Woodgate, J. B. S ys. Rev. B 105 11512	taunton, 4 (2022).		
VNbM	oTaW and derivat	tives.		
► C. Ph	D. Woodgate, J. B. S ys. Rev. Mater. 7 013	taunton, 8801 (2023).		
Influce	nce of Magnetism	ı on Atomic O	ordering.	
► C. Ph	D. Woodgate, D. Hed ys. Rev. Mater. 7 , 05	llund, L. H. Lewis, 3801 (2023).	, J. B. Staunton,	
Influen	ce of Ti additions	: Ti _x VNbMo ⁻	TaW	
► C.	D. Woodgate, J. B. S press I Appl Phys a	taunton, arXiv:2401 16243		
 Design C. ar) 	ing Magnetic Inte D. Woodgate, L. H. L Kiv:2401.02809.	ermetallics: Fe .ewis, J. B. Staunt	Ni + X	
► Al _× CrF	eCoNi Superalloy			
C.	D. Woodgate, G. A. M preparation.	Marchant, L. B. Pa	artay, J. B. Staunton,	
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Context 0	Modelling Approach 000●	Results 00000	Next Steps O	Conclusions 00
Succes	sful Applications			
	CrMpEcCoNi and dariy	atives		
	CrivinFeColvi and deriv	alives.		
	Phys Rev B 105 1151	24 (2022)		
	VNbMoToW and deriv			
	C D Woodgate I B	Staunton		
	Phys. Rev. Mater. 7 01	13801 (2023).		
	Influcence of Magnetis	n on Atomic C	Ordering.	
	C. D. Woodgate. D. He	dlund. L. H. Lewis	. J. B. Staunton.	
	Phys. Rev. Mater. 7, 0	53801 (2023).	, ,	
	Influence of Ti addition	s: Ti _x VNbMo	TaW	
	C. D. Woodgate, J. B.	Staunton,		
	In press, J. Appl. Phys.	arXiv:2401.16243.		
	Designing Magnetic Int	ermetallics: Fe	eNi + X	
	C. D. Woodgate, L. H.	Lewis, J. B. Staun	ton,	
	arXiv:2401.02809.			
	Al _x CrFeCoNi Superallo	у.		
	C. D. Woodgate, G. A.	Marchant, L. B. P	artay, J. B. Staunton,	
	In preparation.			
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Case Study: CrCoNi



Cr aligns antiparallel, Ni and Co parallel with total moment.

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

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CrCoNi: Perturbative Analysis



Shape of modes and location of minimum altered.

⁴Woodgate, Hedlund, Lewis, Staunton, Phys. Rev. Mater. **7** 053801 (2023) C. D. Woodgate Warwick Connections Between Magnetism and Preferred Atomic Arrangements in Multicomponent Alloys 9 of 14

CrCoNi: Inferred Orderings



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

⁴Woodgate, Hedlund, Lewis, Staunton, Phys. Rev. Mater. **7** 053801 (2023) C. D. Woodgate Warwick



CrCoNi: Atomistic Modelling



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Next Steps and Future Work

Multicomponent alloys represent a huge playground.

⁵Shenoy, Woodgate, Staunton, Bartók, Becquart, Domain, and Kermode, in press, Phys. Rev. Mater. arXiv:2309.08689

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Next Steps and Future Work

- Multicomponent alloys represent a *huge* playground.
- Approach is computationally efficient; all figures shown today can be reproduced in ~ 100 CPU-hours. Materials discovery?

⁵Shenoy, Woodgate, Staunton, Bartók, Becquart, Domain, and Kermode, in press, Phys. Rev. Mater. arXiv:2309.08689

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Next Steps and Future Work

- Multicomponent alloys represent a huge playground.
- ▶ Approach is computationally efficient; all figures shown today can be reproduced in ~ 100 CPU-hours. Materials discovery?
- Feed into more sophisticated techniques, e.g. use rapidly-generated configurations in training sets for machine-learned interatomic potentials⁵.





⁵Shenoy, Woodgate, Staunton, Bartók, Becquart, Domain, and Kermode, in press, Phys. Rev. Mater. arXiv:2309.08689

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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

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Laura H. Lewis

Our paper:

Woodgate, Hedlund, Lewis, Staunton,

Phys. Rev. Mater. 7 053801 (2023)





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