Modelling the Effect of Alloying Additions on Atomic Arrangements and Subsequent Magnetic Anisotropy in $L1_0$ FeNi

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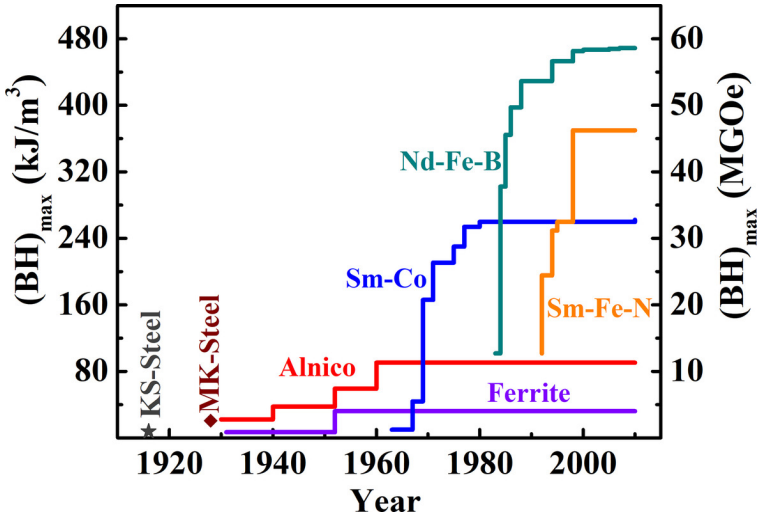
Why Do We Need New Magnets?

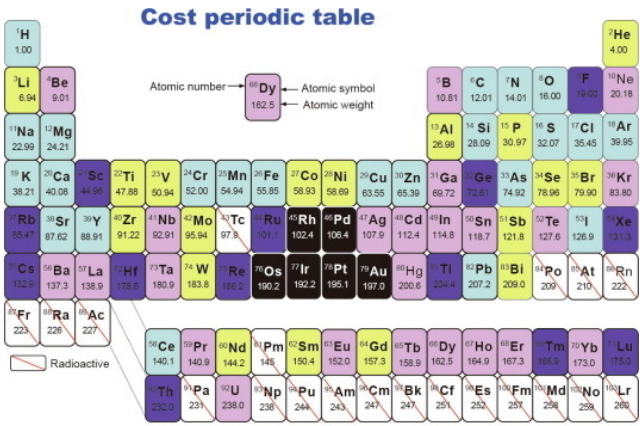
- Permanent magnets find myriad applications, particularly for "green" technoligies.
  $\Longrightarrow$ **increasing demand**
- Existing 'gap' in performance range of permanent magnets for advanced applications[1].
  $\Longrightarrow$ **need materials to fill the 'gap'**



[1] Coey, Scr. Mater. 67 524-529 3-8 (2012)

Why Transition Metals?

- Most permanent magnets for *advanced* applications currently use rare-earth (RE) elements—provide large MAE.
  $\Longrightarrow$ **constrained resources, environmental concerns**

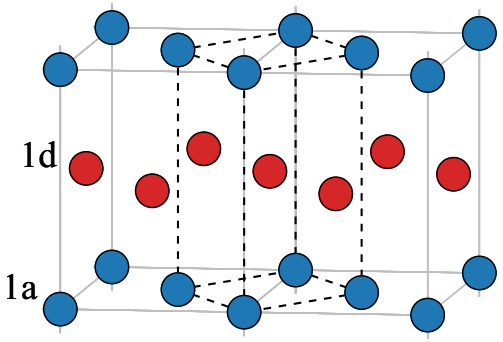


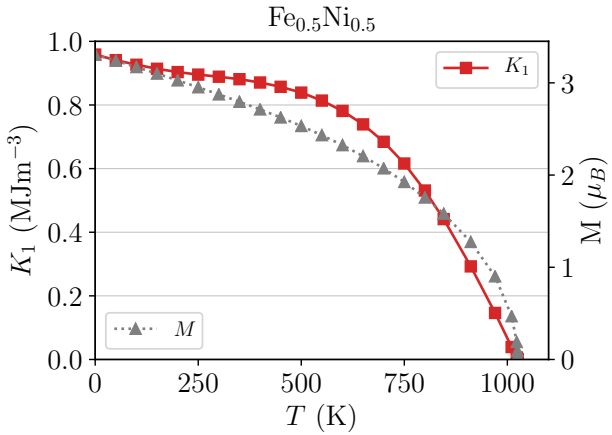
Recipe for a Good Permanent Magnet

1. Large M_S, high T_C.
   $\Longrightarrow$ **Use elements such as Fe, Co, Ni, Mn**
2. Large magnetocrystalline anisotropy energy (MAE).
   $\Longrightarrow$ **Uniaxial crystal structure, spin-orbit coupling, appropriate composition**
3. Need to understand underlying electronic structure.
   $\Longrightarrow$ **Treat atomic ordering and MAE in same framework**

FeNi—Potential for High Anisotropy

- FeNi known to crystallise in $L1_0$ structure.
  - ✓ Large M_S
  - ✓ High T_C
  - ✓ Significant uniaxial anisotropy[2]
- Computationally predicted uniaxial MAE at $T = 0$: $K_U = 0.95$ MJm^{-3}



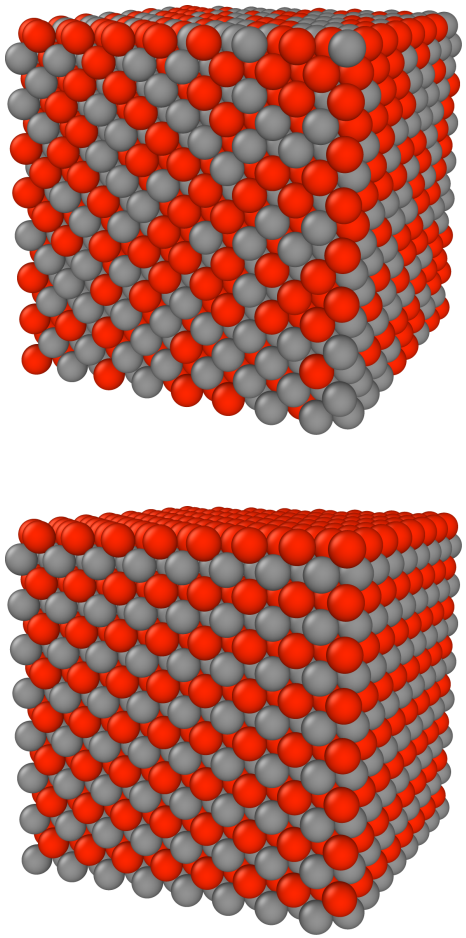


[2]Woodgate, Patrick, Lewis, Staunton, J. Appl. Phys. **134** 163905 (2023).

FeNi—Phase Behaviour

- Model using DFT-based approach[34].

Paramagnetic			Ferromagnetic		
Material	T_{ord}	$L1_0$?	Material	T_{ord}	$L1_0$?
FeNi	175	No	FeNi	507	Yes



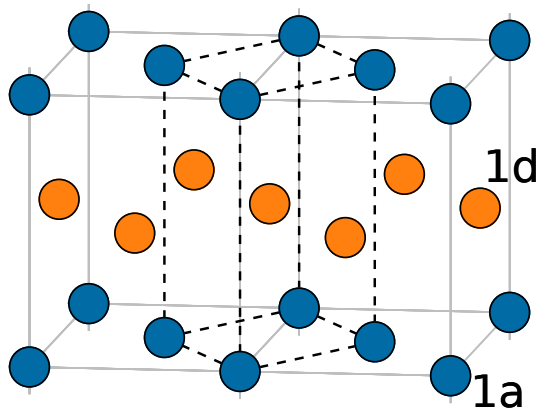
- Magnetic state *crucial* to $L1_0$ ordering.
- **Good, but can we do better?**
  - Higher T_{ord}?
  - Larger K_U?

[3]Woodgate, Hedlund, Lewis, Staunton,Phys. Rev. Mater. **7** 053801 (2023)
[4]Woodgate, Lewis, Staunton, arXiv:2401.02809

Fe_4Ni_3X—Naive Calculation

- Put additive X exclusively on Ni site and calculate magnetic anisotropy.
- Additive 1: $X = Pt$
  - MAE: 3.44 MJm^{-3}
- Additive 2: $X = Al$
  - MAE: 1.66 MJm^{-3}
- **Looks good! Problem solved?**

(FePt has large MAE, strong ordering tendencies)



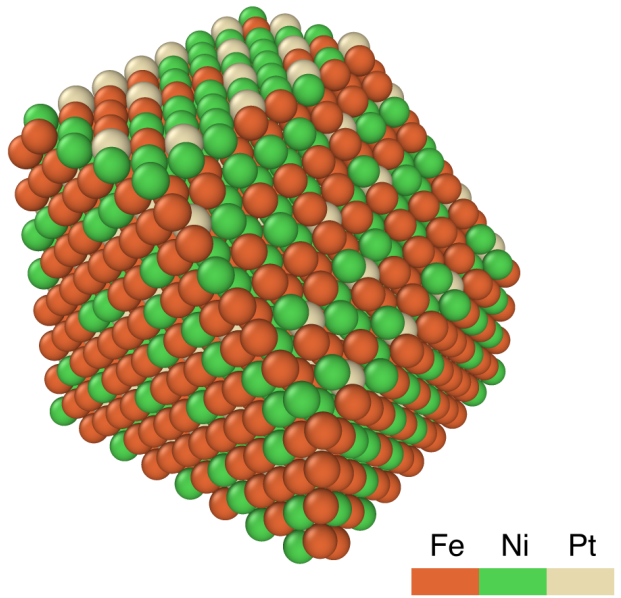
- 1a site occupancy: Fe_{100}
- 1d site occupancy: $Ni_{75}X_{25}$

[4] Woodgate, Lewis, Staunton, arXiv:2401.02809

Fe_4Ni_3X—Phase Behaviour

"All that glisters is not gold" - Shakespeare, Merchant of Venice

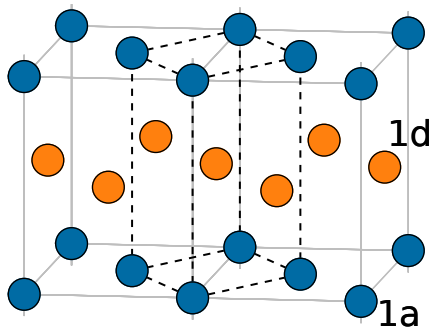
- Additive 1: $X = Pt$
  - $L1_0$-like ordering, but Pt-dominated.
  - Ferromagnetic state distinguishes Fe and Ni better.
- Additive 2: $X = Al$
  - Predicted ordering not even $L1_0$!
- **Anisotropies of predicted orderings?**



[4] Woodgate, Lewis, Staunton, arXiv:2401.02809

Fe_4Ni_3X—'Holistic' Magnetic Anisotropies

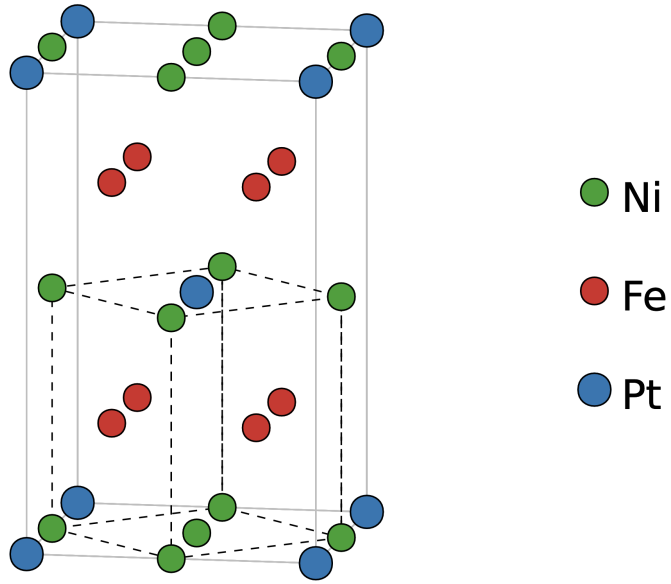
- Addition of Pt, annealing in *paramagnetic* state:
  - 1a site occupancy: $Fe_{61}Ni_{39}$
  - 1d site occupancy: $Fe_{39}Ni_{36}Pt_{25}$
  - Predicted MAE of 0.96 MJm^{-3}
- Addition of Pt, annealing in *ferromagnetic* state:
  - 1a site occupancy: $Fe_{68}Ni_{32}$
  - 1d site occupancy: $Fe_{32}Ni_{43}Pt_{25}$
  - Predicted MAE of 1.22 MJm^{-3}



[4] Woodgate, Lewis, Staunton, arXiv:2401.02809

Fe_4Ni_3Pt—'Ground-State' Structure

- Run Monte Carlo simulations—simulated annealing.
- Example: addition of Pt.
- Find a novel ground-state tetragonal structure.
- Anisotropy of compound is 2.77 MJm^{-3}.



[4] Woodgate, Lewis, Staunton, arXiv:2401.02809

Summary

Take-Home Message 1

Additives to FeNi system have the potential to improve atomic ordering tendencies (uniaxial structures) and magnetic anisotropy.

Take-Home Message 2

HOWEVER, it is necessary to understand on which lattice site(s) additives prefer to sit.

Take-Home Message 3

Enlarged, multicomponent, compositional search space is needed, not just FeNi plus alloying addition.

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Our Paper: arXiv:2401.02809

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