# Tuning $L1_0$ Order and Magnetocrystalline Anisotropy in Rare-Earth-Free Transition Metal Magnets

C. D. Woodgate[1], L. H. Lewis[2], J. B. Staunton[1]

[1]University of Warwick, Coventry, UK
[2]Northeastern University, Boston, USA

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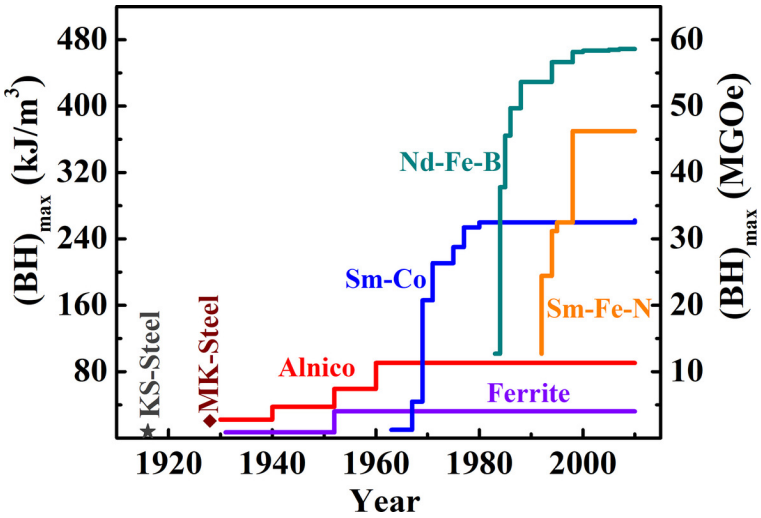
# Why Do We Need New Magnets?

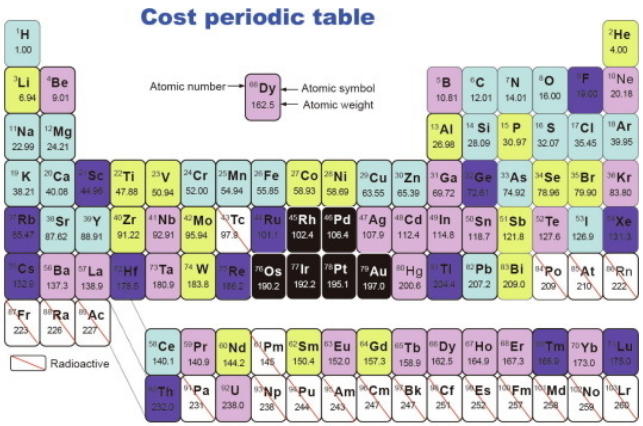
- Permanent magnets find myriad applications, particularly for "green" technoligies.
  $\Longrightarrow$ **increasing demand**
- Existing 'gap' in performance range of permanent magnets for advanced applications[1].
  $\Longrightarrow$ **need materials to fill the 'gap'**



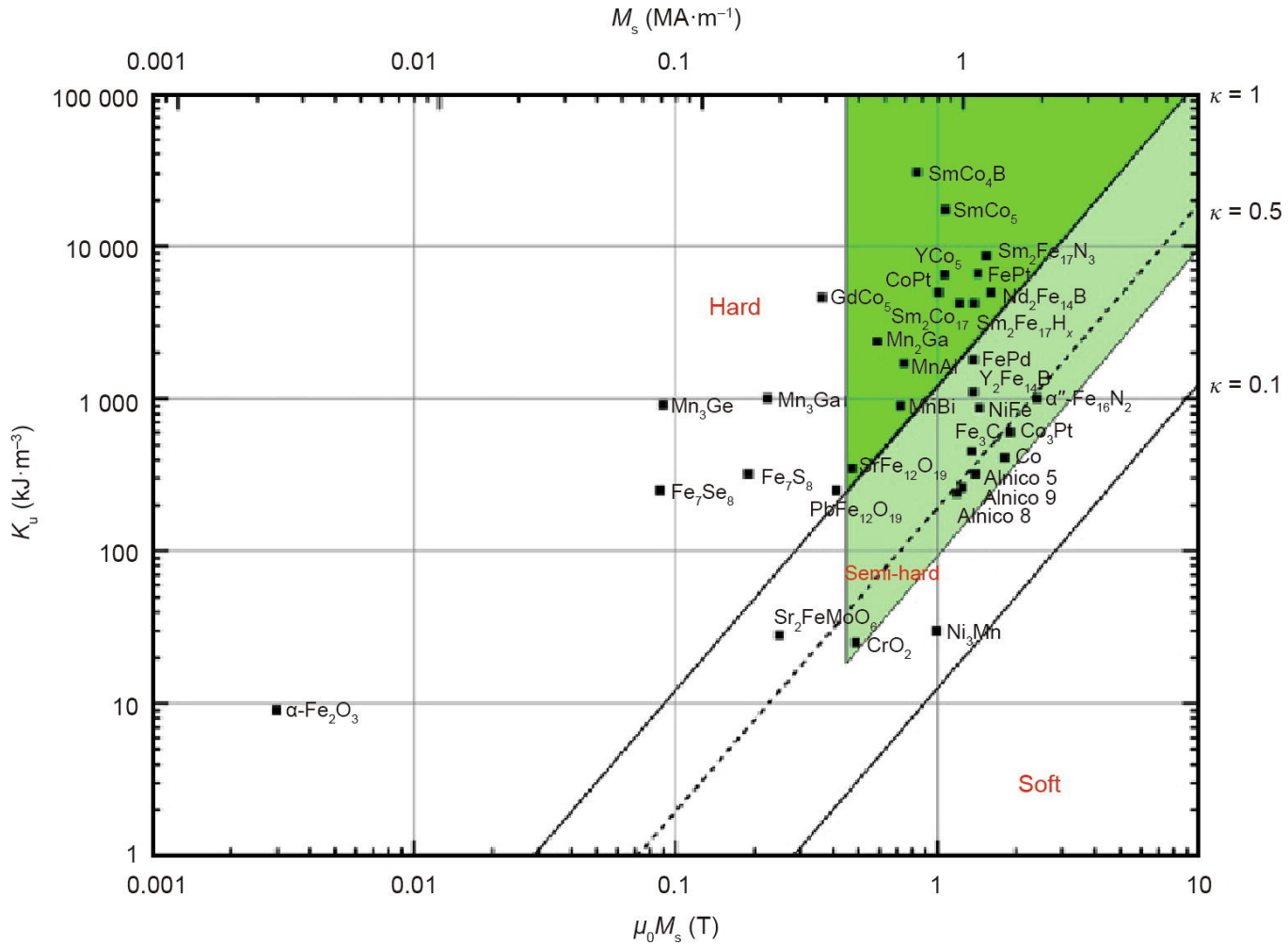

[1] Coey, Scr. Mater. 67 524-529 3-8 (2012)

# Why *Transition Metals*?

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



# Why *Transition Metals*?



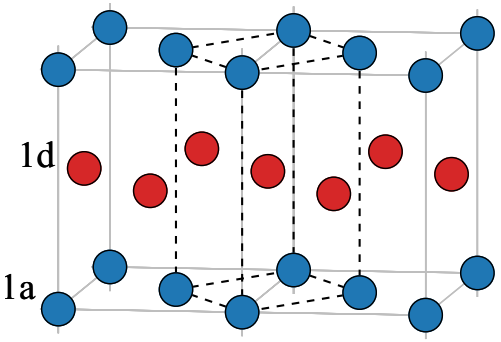

[2] Coey, Engineering 6 119-131 (2020)

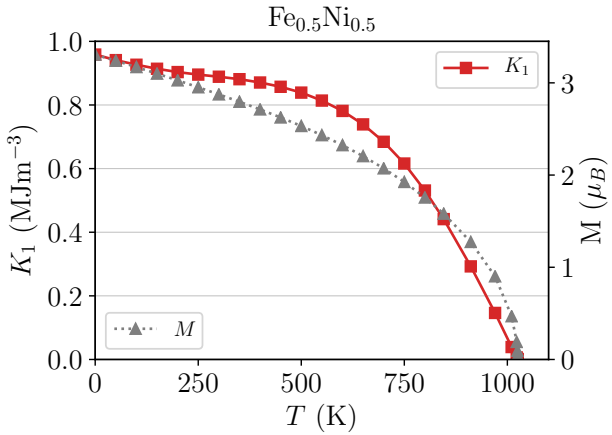
# Recipe for a Good Permanent Magnet

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—Magnetic Anisotropy

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




[3] C. D. Woodgate, C. E. Patrick, L. H. Lewis, J. B. Staunton, **arXiv:2307.15470**

# FeNi—Phase Behaviour

- Use DFT-based approach to model phase behaviour[4]

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

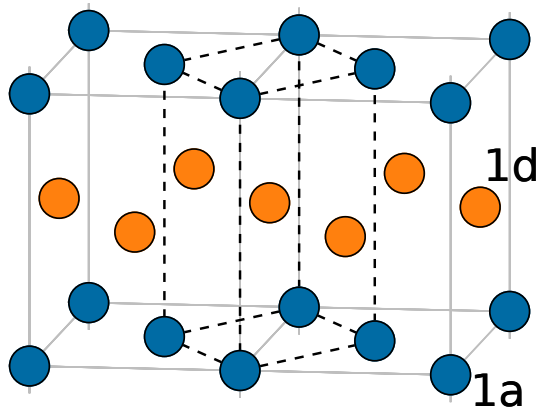
- Magnetic ordering *crucial* to atomic ordering.
- **Good, but can we do better?**
  - Higher $T_{ord}$?
  - Larger $K_U$?

[4] C. D. Woodgate, D. Hedlund, L. H. Lewis, J. B. Staunton, Phys. Rev. Mater. **7** 053801 (2023)

# $Fe_4Ni_3X$—Naive Calculation

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

(FePt has large MAE, strong ordering tendencies)

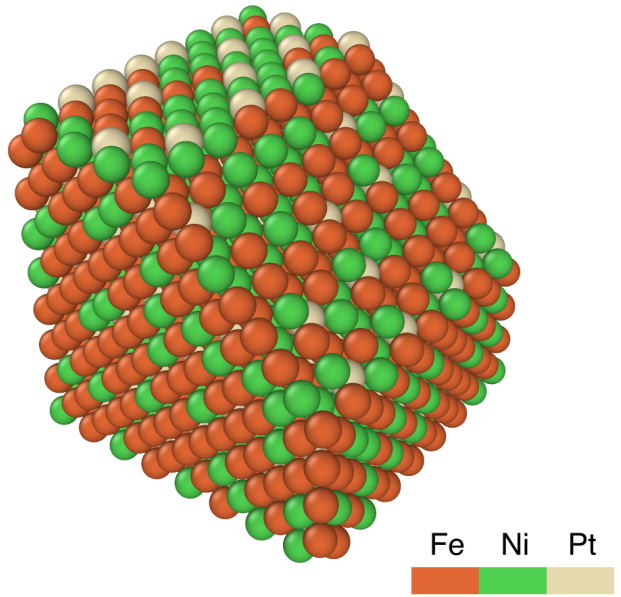



- 1a site occupancy: $Fe_{100}$
- 1d site occupancy: $Ni_{75}X_{25}$

# $Fe_4Ni_3X$—Phase Behaviour

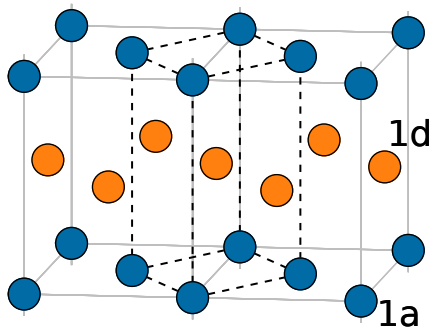
*"All that glisters is not gold"* - Shakespeare, Merchant of Venice

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



# $Fe_4Ni_3X$—'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 $MJm^{-3}$



# 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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