

Tuning L₁₀ Order and Magnetocrystalline Anisotropy in Rare-Earth-Free Transition Metal Magnets

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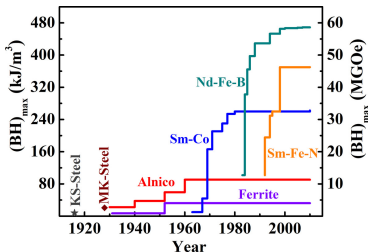
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Why Do We Need New Magnets?

- ▶ Permanent magnets find myriad applications, particularly for “green” technologies.
⇒ **increasing demand**
- ▶ Existing ‘gap’ in performance range of permanent magnets for advanced applications¹.
⇒ **need materials to fill the ‘gap’**



¹Coey, 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.
⇒ **constrained resources, environmental concerns**



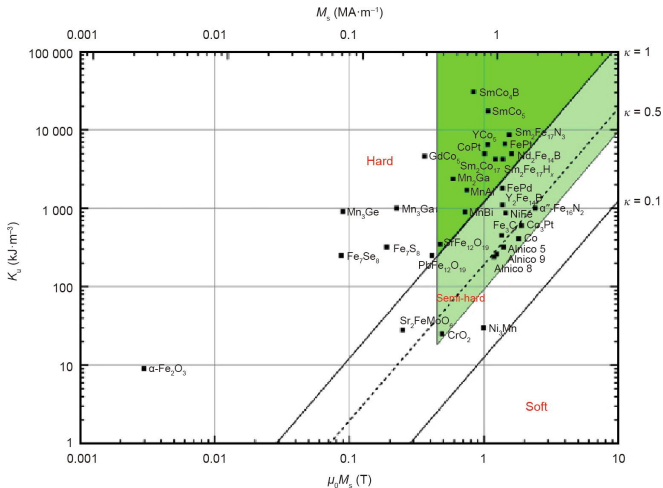
Cost periodic table

Atomic number — Dy — Atomic symbol
162.5 — Atomic weight

H 1.01																	He 4.00	
Li 6.94	Be 9.01											B 10.81	C 12.01	N 14.01	O 16.00	F 18.99	Ne 20.18	
Na 22.99	Mg 24.31											Al 26.98	Si 28.09	P 30.97	S 32.07	Cl 35.45	Ar 39.95	
K 39.10	Ca 40.08	Sc 44.96	Ti 47.88	V 50.94	Cr 52.00	Mn 54.94	Fe 55.85	Co 58.93	Ni 58.71	Cu 63.55	Zn 65.38	Ga 69.72	Ge 72.64	As 74.92	Se 78.96	Br 79.90	Kr 83.80	
Rb 85.47	Sr 87.62	Y 88.91	Zr 91.22	Nb 92.91	Mo 95.94	Tc 98.91	Ru 101.07	Rh 101.07	Pd 106.36	Ag 107.87	Cd 112.41	In 114.82	Sn 118.71	Sb 121.76	Te 127.60	I 126.91	Xe 131.29	
Cs 132.91	Ba 137.33	La 138.91	Hf 178.49	Ta 180.95	W 183.85	Re 186.21	Os 190.23	Ir 192.22	Pt 195.08	Au 196.97	Hg 200.59	Tl 204.38	Pb 207.2	Bi 208.98	Po 209	At 210	Rn 222	
Fr 223	Ra 226	Ac 227																
			Ce 140.12	Pr 140.91	Nd 144.24	Pm 145	Sm 150.36	Eu 151.96	Gd 157.25	Tb 158.93	Dy 162.50	Ho 164.93	Er 167.26	Tm 168.93	Yb 173.05	Lu 174.97		
			Th 232.04	Pa 231.04	U 238.03	Np 237.05	Pu 244.06	Am 243.06	Cm 247.07	Bk 247.07	Cf 251.08	Es 252.08	Fm 257.09	Md 258.10	No 259.10	Lr 260.10		

Radioactive

Why Transition Metals?



²Coey, Engineering 6 119-131 (2020)

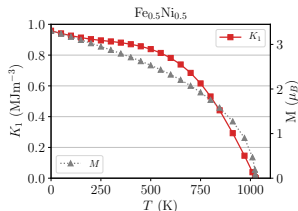
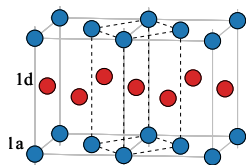
Recipe for a Good Permanent Magnet

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



FeNi—Magnetic Anisotropy

- ▶ FeNi known to crystallise in L1₀ structure.
 - ✓ Large M_S^3
 - ✓ High T_C^3
 - ✓ Significant uniaxial anisotropy³
- ▶ Computationally predicted uniaxial MAE at $T = 0$:
 $K_U = 0.95 \text{ MJm}^{-3}$



³C. D. Woodgate, C. E. Patrick, L. H. Lewis, J. B. Staunton, [arXiv:2307.15470](https://arxiv.org/abs/2307.15470)

FeNi—Phase Behaviour

- ▶ Use DFT-based approach to model phase behaviour⁴

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

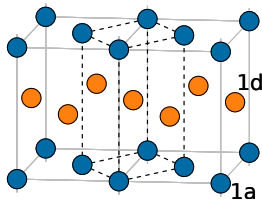
- ▶ Magnetic ordering *crucial* to atomic ordering.
- ▶ **Good, but can we do better?**
 - ▶ Higher T_{ord} ?
 - ▶ Larger K_U ?

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

Fe₄Ni₃X—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)

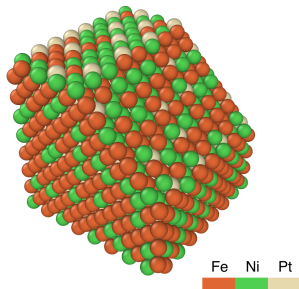


- ▶ 1a site occupancy: Fe₁₀₀
- ▶ 1d site occupancy: Ni₇₅X₂₅

Fe₄Ni₃X—Phase Behaviour

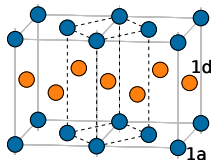
“All that glisters is not gold” - Shakespeare, Merchant of Venice

- ▶ Additive 1: Pt
 - ▶ L1₀-like ordering, but Pt-dominated.
 - ▶ Ferromagnetic state distinguishes Fe and Ni better.
- ▶ Additive 2: Al
 - ▶ Predicted ordering not even L1₀!
- ▶ **Anisotropies of predicted orderings?**



Fe₄Ni₃X—‘Holistic’ Magnetic Anisotropies

- ▶ Addition of Pt, annealing in *paramagnetic* state:
 - ▶ 1a site occupancy: Fe₆₁Ni₃₉
 - ▶ 1d site occupancy: Fe₃₉Ni₃₆Pt₂₅
 - ▶ Predicted MAE of 0.96 MJm⁻³
- ▶ Addition of Pt, annealing in *ferromagnetic* state:
 - ▶ 1a site occupancy: Fe₆₈Ni₃₂
 - ▶ 1d site occupancy: Fe₃₂Ni₄₃Pt₂₅
 - ▶ 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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