Jumping the Gap: Can Tetrataenite become a "Hard" Permanent Magnet?

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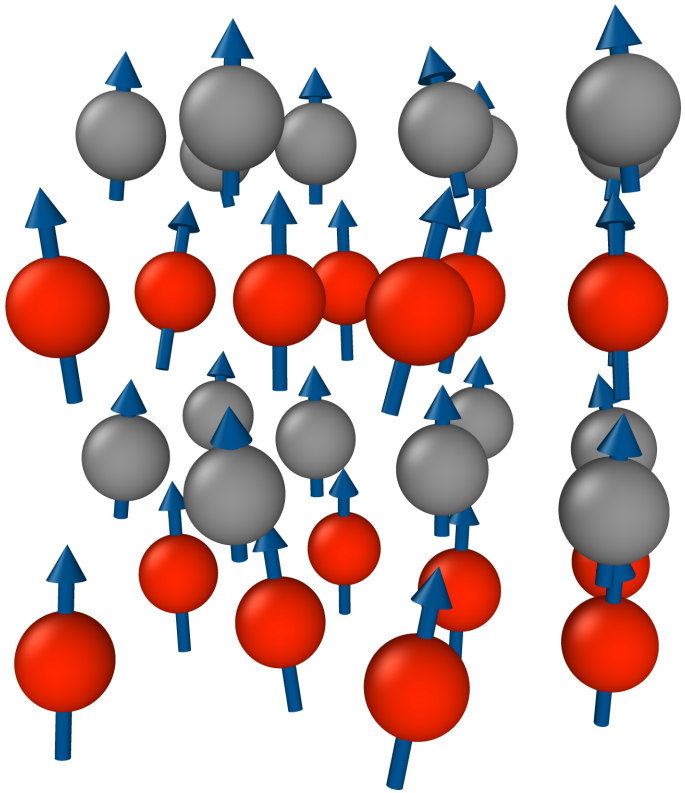
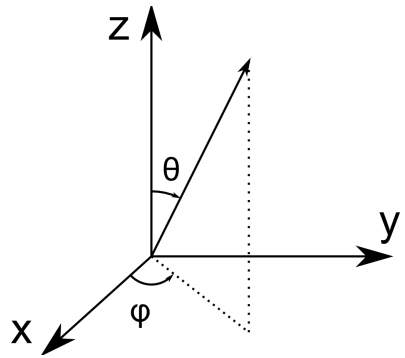
Motivation

- $L1_0$ FeNi (tetrataenite) is a under consideration as a candidate rare-earth-free permanent magnet, provided structure can be optimised.
- Consists entirely of two abundant $3d$ transition metals.
- Dispute in the literature as to the value of its uniaxial anisotropy energy.



Magnetocrystalline Anisotropy Energy (MAE)



- Tetragonal sample with magnetisation $\mathbf{M} = |\mathbf{M}|(\cos\theta\cos\phi, \cos\theta\sin\phi, \sin\theta)$.
- Magnetocrystalline anisotropy energy written

$$\begin{aligned} K(\theta,\phi) =& K_1 \sin^2\theta + K_2 \sin^4\theta + K_3 \sin^4\theta \cos 4\phi + \ldots \\ \approx& K_1 \sin^2\theta \end{aligned}$$

- Conventional to measure/calculate *uniaxial* MAE:

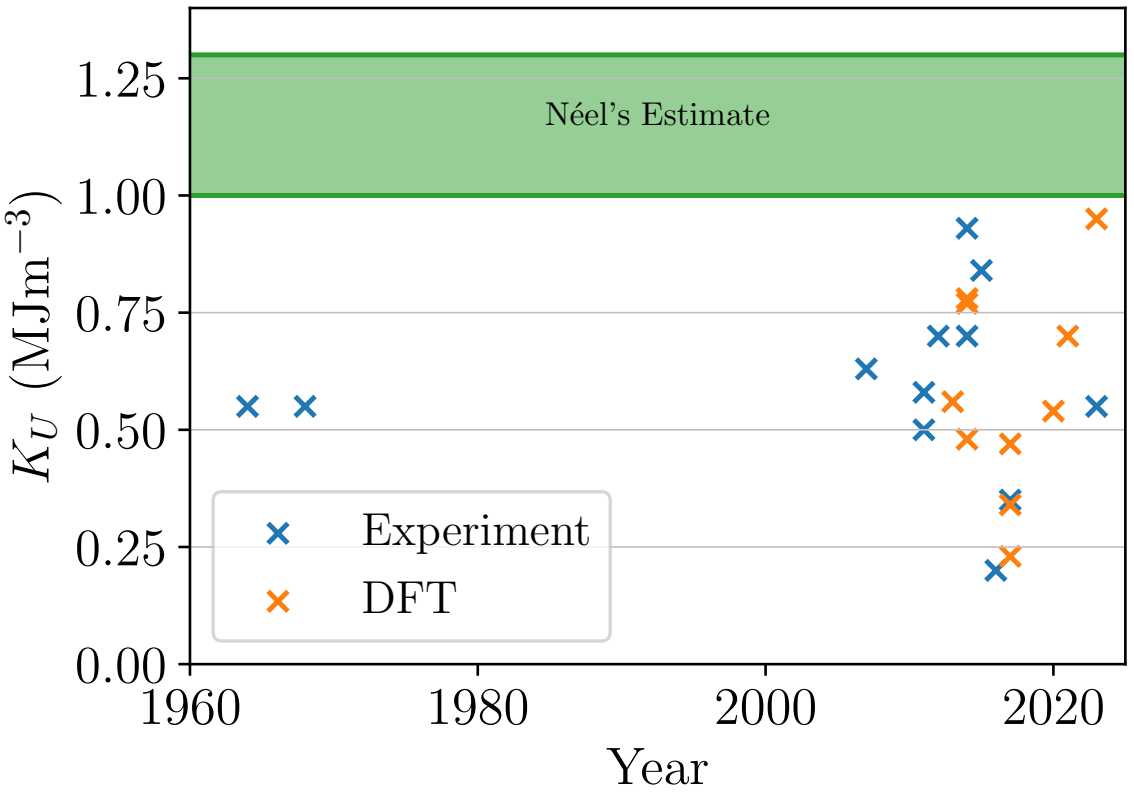
$$K_U = K(\hat{\mathbf{z}}) - K(\hat{\mathbf{x}}) = K_1 + K_2 + K_3$$

History of $L1_0$ FeNi

- $L1_0$ phase first reported in 1960s by Néel and co-workers[1][2].
- Also found to occur naturally in metallic meteorites, mineral name tetrataenite.
- Challenging to synthesise. Recent experimental efforts have focussed on manufacturing thin films or on targeted processing.
- Extensively studied using density functional theory calculations, *e.g.* Ref. 3. Primarily for perfectly ordered structures at $T = 0$ K, though.

[1] Néel *et al.*, J. Appl. Phys. **35** 873 (1964)
[2] Paulevé *et al.*, J. Appl. Phys. **55** 989 (1968)
[3] M. Werwiński, W. Marciniak, J. Phys. D: Appl. Phys. **50** 495008 (2017)

Determinations of K_U



- Problem: DFT studies primarily study perfectly ordered structures at $T = 0$ K.

Determinations of K_1, K_2, K_3

- Only two studies resolve K into its separate coefficients.
- Both find large K_2, K_3.
  - Néel *et al.*: $K_1 = 0.32$, $K_2 = 0.23$.
  - Paulevé *et al.*: $K_1 = 0.3$, $K_2 = 0.17$, $K_3 = 0.08$.

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Magnetic Properties of an Iron–Nickel Single Crystal Ordered by Neutron Bombardment

L. Néel, J. Pauleve, R. Pauthenet, J. Laugier, and D. Dautreppe
Centre d'Etudes Nucléaires et Laboratoire d'Electrostatique et de Physique du Métal, Grenoble, France

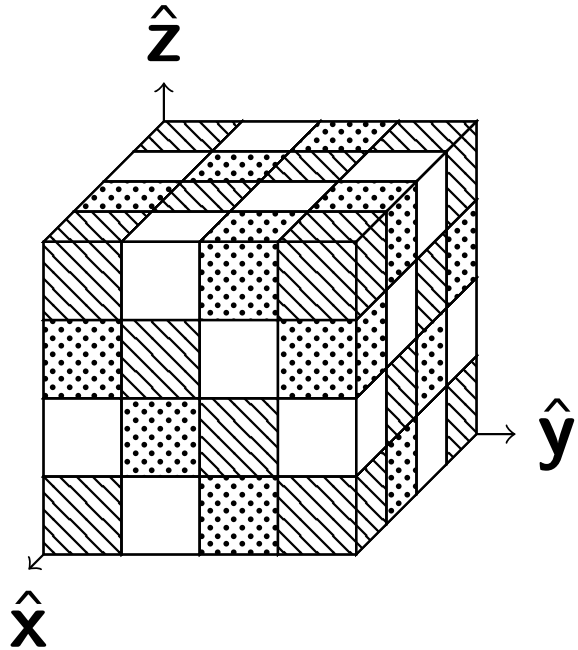
JOURNAL OF APPLIED PHYSICS VOLUME 39, NUMBER 2 1 FEBRUARY 1968

Magnetization Curves of Fe–Ni (50–50) Single Crystals Ordered by Neutron Irradiation with an Applied Magnetic Field

J. Paulevé, A. Chamberod, K. Krebs, and A. Bourret
Centre d'Etudes Nucléaires de Grenoble, France

Potential Origins of Large K_2 and K_3?

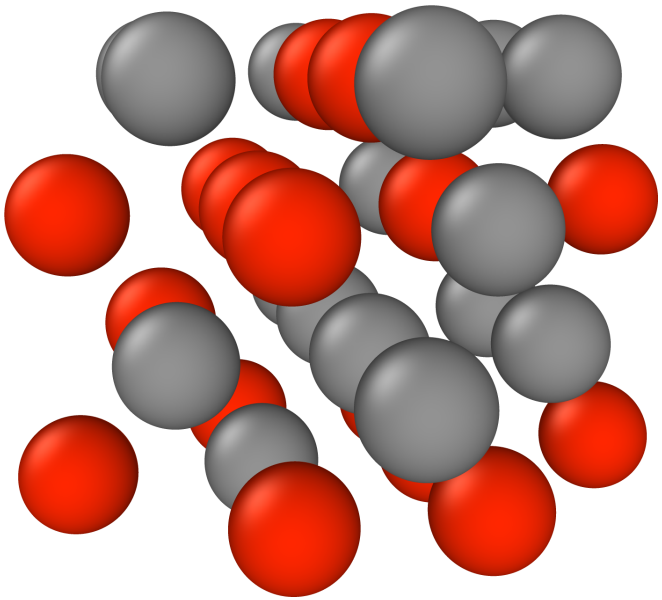
- In our DFT calculations, we find *negligible* K_2, K_3.
- Néel proposes bulk sample made up of individual $L1_0$ nanocrystals, layering directions not all aligned[1].
- Exchange interaction between nanoscale domains yields large, macroscopic K_2 and K_3.

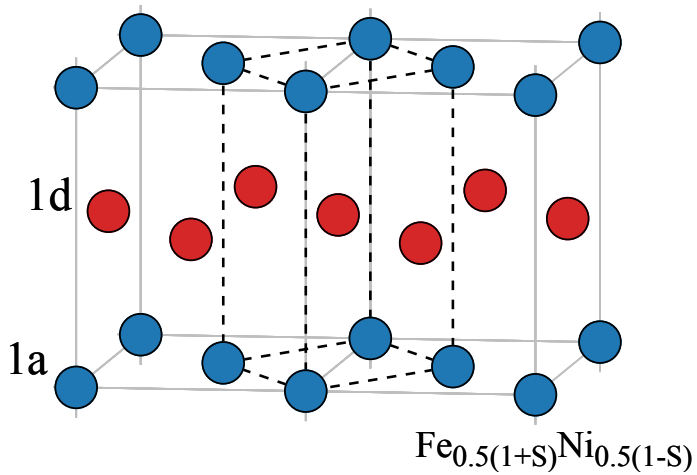


[1] Néel *et al.*, J. Appl. Phys. **35** 873 (1964)

Degree of Atomic Order

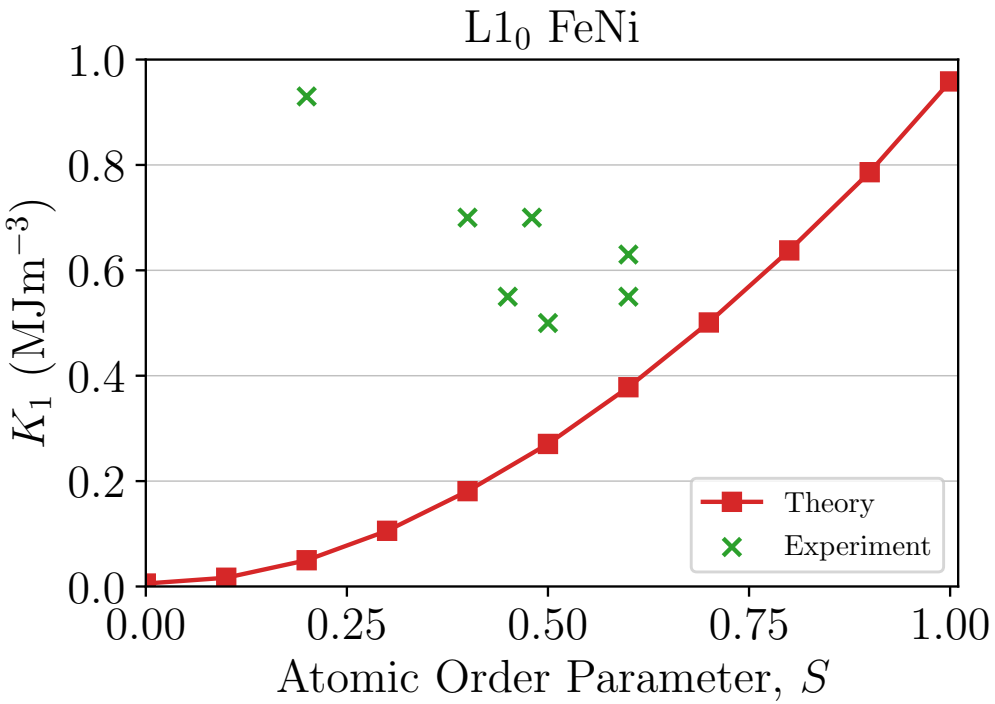
- Even if I have a single-variant nanocrystal, is is perfectly ordered?
- Define atomic order parameter, S to quantify how well-ordered sample is[4].





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

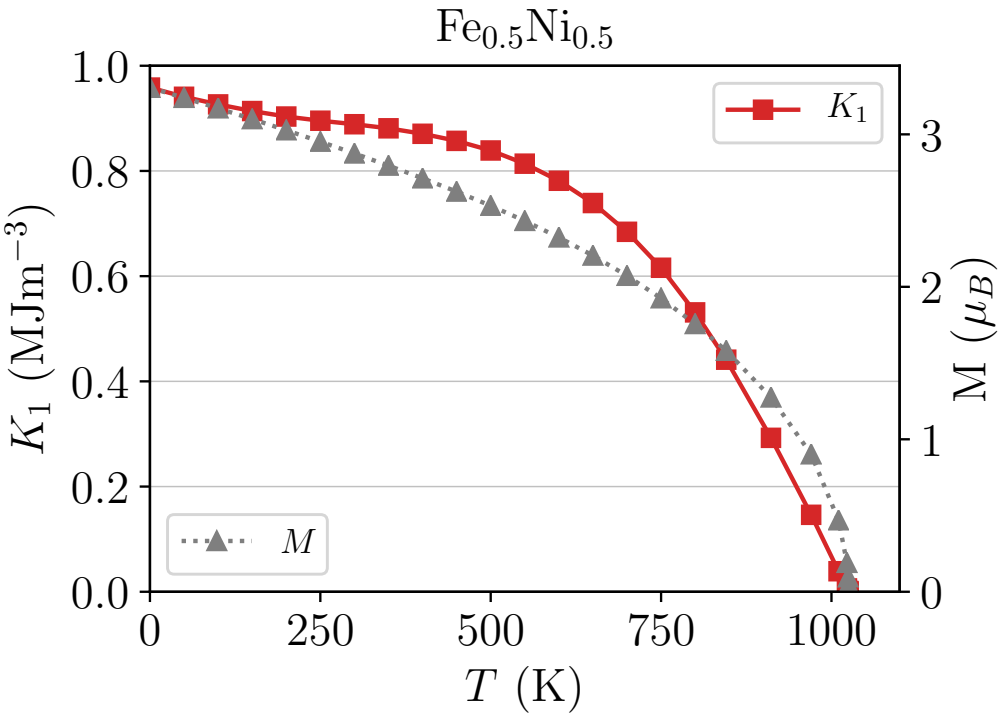
Degree of Atomic Order



- Clear disagreement between theory and experiment once effects of imperfect atomic order are considered[4].

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

Effects of Finite Temperature



- Robust computationally predicted finite temperature performance[4].

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

Summary: Aspects to Consider

Aspect 1: Polycrystalline, Multivariant Samples

Experimentally FeNi has significant K_2, K_3; these are *not* detected in DFT calculations. Nanoscale origins?

Aspect 2: Imperfect Atomic Order

Decreasing atomic order decreases computationally predicted MAE, makes for large discrepancy between theory and experiment.

Aspect 3: Finite Temperature

Tetrataenite has good finite temperature performance, but increasing temperature decreases MAE compared to $T = 0$.

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