Permanent Magnets and the “Rare Earth crisis”
Mark van Schilfgaarde, King’s College London

- A brief survey of the early history of magnetism
- Hard and Soft magnets --- what are they for?
- Materials abundance issues in the best hard magnets ... the “rare earth crisis”
- Origins of Ferromagnetism, key role of demagnetising fields
- Figures of merit: magnetic energy, moment, exchange, and anisotropy
- Relation of key magnetic properties to electronic structure
- Domain walls, relation to anisotropy and demagnetising fields, and hysteresis
- What causes anisotropy? What makes it large?
- Reducing rare earth content in permanent magnets
Early History of Magnetism

Magnets have been around a long time:

Thales of Miletus (6th century BC) is credited with discovering lodestone's attraction to iron (lodestone is mostly magnetite).

Even earlier: a grooved magnetic bar was found from artifacts of the Olmecs in Central America ~1000 years before Thales.

Sir William Gilbert did experiments on magnetism and wrote *De Magnete* (1600). He concluded that the Earth was magnetic. Some credit him as the father of electricity and magnetism. Gilbert also studied magnetic properties of carbon steel.
Uses of Magnetism

Until recently magnets were mostly confined to specialised applications as electromagnetic energy converters (motors and generators).

Before the 1950s, magnets tended to assume “interesting” shapes, because they were not strong enough to overcome the demagnetising field.

Today magnets are used everywhere, such as in a typical household (which has ~100 magnets, e.g., as microwave generators) though we are often not aware of many of them.
A magnet is typically classified by “hardness---” the amount of hysteresis it exhibits.

Cycle the magnet around a loop ⇒ energy dissipated in proportion to loop area.

The ideal hard magnet forms a square hysteresis loop. Both hard and soft magnets have widespread uses.
Applications of Hard and Soft Magnets

Soft magnetic materials
- Used when the magnetization is cycled rapidly (transformer coils, generators and some motors)
- Low hysteresis minimizes energy loss.
- Popular magnet: Permalloy (Ni/Fe alloy), $H_c \sim 2 \cdot 10^{-7}$ Tesla

Hard magnets (or permanent magnets)
- Used in applications where you don’t want material to demagnetise e.g. loudspeakers, motors, magnetic recording
- The hardest magnets contain rare earths, e.g. $\text{Nd}_2\text{Fe}_{14}\text{B}$, with a coercive field $H_c \sim 1.2$ Tesla.
- Rare earths make the best magnets, but issues of materials abundance become important (later).
Inexpensive Hard Magnets

Ferrite family: e.g. BaFe$_{12}$O$_{19}$, SrFe$_{12}$O$_{19}$, (the seal on your refrigerator, or the magnetron in your microwave oven). Comprises most hard magnets, but only ~1/3 by cost.

First developed in the Netherlands in [J. Went, G.W. Rathenau, E.W. Gorter, G.W. van Oosterhout, Phys. Rev. 86, 424 (1951)] and marked the onset of the modern era of magnetic materials.

A big drawback of the Ferrites is that the net magnetic moment $M$ is moderate because some spins align antiferromagnetically.
Neodymium Magnets


The combination of Fe (a transition metal with large moment and large exchange coupling) and Nd (a \(4f\) element with very large anisotropy) make this a nearly perfect magnet!

Pure \( \text{Nd}_2\text{Fe}_{14}\text{B} \) has limited applicability because hysteresis falls off rapidly with \( T^0 \). \( \text{Nd}_2\text{Fe}_{14}\text{B} \) can be stabilised up to ~200°C by replacing 3-10wt.% of Nd by Dy (or Tb)
Breakdown of the Hard Magnet Market

Ferrites are more commonly used because of low cost (<$5/kg). But energy density is low (<38 kJ/m$^3$).

$\text{Nd}_2\text{Fe}_{14}\text{B}$ is a vastly superior magnet: some lab magnets have energy density $\sim 470$ kJ/m$^3$; commercial magnets a bit lower.

**Cost** is a big issue, because of the Nd (next slide).

But even though $\text{Nd}_2\text{Fe}_{14}\text{B}$ can be stabilised by substituting some Nd for Dy, Dy is much more rare and costly than Nd!

The SmCo market is aimed mainly at high-$T_c$ applications where the Nd magnets do not perform well.
The more Dy, the better the magnet


**Figure 1**

$H_{cJ}$ and $B_r$, as a function of approximate Dy content

<table>
<thead>
<tr>
<th>Dy (%)</th>
<th>$H_{cJ}$ (Oe)</th>
<th>$B_r$ (kG)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>35,000</td>
<td>15</td>
</tr>
<tr>
<td>2</td>
<td>30,000</td>
<td>14</td>
</tr>
<tr>
<td>4</td>
<td>25,000</td>
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<tr>
<td>6</td>
<td>20,000</td>
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<tr>
<td>8</td>
<td>15,000</td>
<td>11</td>
</tr>
<tr>
<td>10</td>
<td>10,000</td>
<td>10</td>
</tr>
</tbody>
</table>

- **M**: 80°C, 100°C
- **H**: 120°C
- **SH**: 150°C
- **UH**: 180°C
- **EH**: 200°C
- **AH**: 220°C

Typical uses for each grade are shown along the top columns. Data supplied by Arnold Magnetic Technologies, http://www.arnoldmagnetics.com.
Volatility of the RE Market

The price of Nd and Dy underwent a huge spike in 2011. China adopted a new policy that it would no longer export raw material. Dy peaked at $1400/kg in 2011; it is ~$500/kg today, apparently largely as a result of illegal production in China (China mines 99% of worldwide production, ~100 tonnes/year).

This solved the immediate crisis, but …

RE’s are Emerging Critical Materials

Regardless of Chinese policies (China has only ~30% of all RE reserves worldwide), a critical shortage of Dy is shaping up, owing to anticipated large increases in demand for motors in electric cars and wind turbines.

~100 grams of Dy go into in every electric car motor. If Toyota were to manufacture 2 million cars per year, the available Dy supply would be exhausted.

According to the US DOE, Dy is the single most critical element for emerging clean energy technologies, since no good alternative has yet emerged.

Basic Science of a Ferromagnet: origin of FM

In classical magnetism we associate a magnetic moment \( \mu \) with a current loop. An electron at point \( \mathbf{r} \) generates a magnetic moment

\[
\bar{\mu} = -\frac{e}{2} \mathbf{r} \times \mathbf{v} = \left(-\frac{e}{2m}\right)\mathbf{r} \times \mathbf{p} \equiv \gamma \mathbf{L}
\]

But ferromagnetism does not originate from circulating \( e^- \) ...

Instead, an \( e^- \) has an intrinsic angular momentum (spin) which emerges from the intersection of relativity & quantum theory. Magnetism is a consequence if the fact that \( e^- \) must obey the Pauli exclusion principle.

Formally: the many-particle wave function must be antisymmetric when two particles are exchanged.
Origin of FM II

The total wave function $\psi$ consists of a product of orbital part $r$ and spin part $\sigma$.

Consider a two-particle $\psi(r_1\sigma_1, r_2\sigma_2)$ under particle exchange $r_1\sigma_1 \leftrightarrow r_2\sigma_2$. It is not difficult to show that:

- If $\sigma_1$ and $\sigma_2$ are parallel ($\uparrow, \uparrow$), the orbital part $\psi$ of must be antisymmetric
- If $\sigma_1$ and $\sigma_2$ are antiparallel ($\downarrow, \uparrow$), the orbital part $\psi$ of must be symmetric

Let $\phi$ be the orbital part of $\psi$. Consider the symmetric case when $r_1 \rightarrow r_2$. We must have simultaneously:

$$\lim_{r_1 \rightarrow r_2} \phi(r_1, r_2) = - \lim_{r_1 \rightarrow r_2} \phi(r_2, r_1) \quad \text{and} \quad \lim_{r_1 \rightarrow r_2} \phi(r_1, r_2) = + \lim_{r_1 \rightarrow r_2} \phi(r_2, r_1)$$

These can both be true only when $\phi(r_1, r_2) \rightarrow 0.$
Origin of FM III

Last slide: these can both be true only when $\varphi(r_1, r_2) \to 0$.

This implies that when the spins are aligned parallel $(\uparrow, \uparrow)$, the probability of two $e^-$ meeting each other vanishes.

The exclusion principle imposes no equivalent constraint when spins are antiparallel $(\downarrow, \uparrow)$.

Thus, $e^-$ with $(\uparrow, \uparrow)$ spins tend to stay away from each other while $e^-$ with $(\downarrow, \uparrow)$ spins have no such built-in correlation.

Remember that $e^-$ are charged and repel each other.

Thus, correlations keeping $e^-$ apart reduce their repulsion: $e^-$ can lower their energy by aligning their spins.

This interaction is called the “exchange interaction” and can be cast in terms of an effective magnetic field called the “exchange field” or “exchange-correlation field” $B_{xc}$.
Demagnetising Fields

True magnetic dipolar fields do play an important role in real magnets. They are tiny compared to $B_{xc}$. But in a macroscopic magnet Avogadro's number of them combine to generate a field whose energy scales in proportion to the volume. The effects of this field become important at the boundary of the magnet.

The total and external fields are related:

$$B = \mu_0(H+M).$$

From E&M we know $\nabla \cdot B = 0$, so $\nabla \cdot H = -\nabla \cdot M$.

$M$ originates from the $e^-$ through $B_{xc}$. $M$ changes abruptly at the surface and therefore $\nabla \cdot M$ and $\nabla \cdot H$ become very large. The resultant field, called the demagnetizing field $H_d$, is present both inside and outside the magnet, opposing the internal field.
Stray Fields

Inside the magnet $H_d$ opposes $B$, destabilising the ferromagnetism. Now $H_d$ depends strongly on the shape in complicated ways (see later). Until the discovery of the Ferrite magnets in the 1950’s they had to be formed in funny shapes.

Outside the magnet $H_d$ are “stray fields.” For a magnetic memory device, they interfere with neighbouring bits and the trick is to avoid them as much as possible. For motors and generators, they are essential as they supply the torques which couple mechanical and electrical power.
Influence of Magnet Shape on Energy Density

The energy stored by the demagnetising field is given by

\[ E_d = \frac{\mu_0}{2} \int_V d^3 r \mathbf{B} \cdot \mathbf{H}_d \]

\( H_d \) is a very complicated function of the shape, so it is often parameterised by a factor \( N \) with \( H_d \approx -N M \).

(Note: \( N \) should be a function if \( r \); usually ignored)

Consider an ideal hysteresis loop.

Use \( B = \mu_0 (H_d + M_s) \) and \( H_d \approx -N M_s \):

\[ E_d = \frac{\mu_0 V}{2} M_s^2 (N - N^2) \]

Thus there is no energy contained in the magnet if no demagnetising field (\( N = 0 \)) or if it approaches the internal field (\( N = 0 \))
Energy Figure of Merit

For the optimum energy density, maximise wrt $N$:

$$\frac{dE_d}{dN} = 0 = \frac{\mu_0 V}{2} M_s^2 (1 - 2N) \Rightarrow N = 1/2$$

It turns out that a cylinder with radius/height $\approx 2$ yields $N = H_d/M \approx 1/2$.

Modern commercial magnets are often in this shape.

From Sunshine Magnets

The standard figure of merit is

$$|BH|_{\text{max}} \sim 2 E_d / V$$

As the hysteresis loop deviates from ideal, $|BH|_{\text{max}}$ drops.
Many Figures of Merit

In practical magnets, many factors to consider. Some key figures of merit are:

(1) Large maximum energy product $|BH|_{\text{max}}$

(2) high saturation $M_s$

(3) nearly rectangular hysteresis loop with $M_r \approx M_s$ and $H_c > 1/2 M_s$

(4) very high uniaxial magnetocrystalline anisotropy energy $K$ (see soon)

(5) Low cost

(5) Significant structural and/or composition fluctuations $\Rightarrow$ high $H_c$ from domain wall nucleation or pinning

(7) High Curie temperature $T_c$

(8) Good temperature stability

(9) Corrosion resistance

(10) Mechanical strength

(11) Machinability

No magnet has good figures of merit in all of these categories!
Local-Moments picture of Magnetism

The spin density \((n^{\uparrow} - n^{\downarrow})\) originates from \(d\) or \(f\) orbitals, and tends to be rather localized near the nucleus.

Suggests that magnetism can be well described in term of a local-moment picture, an ensemble of spins on a lattice that interact with each other.

A spin of size \(S_i\) resides at site \(i\) canted by some angle.

Heisenberg model: each \(S_i\) interacts with each \(S_j\) through an interaction \(J_{ij}\):

\[
J_{ij} S_i \cdot S_j = J_{ij} S_i S_j \cos(\theta_{ij})
\]

\(J_{ij}\) can be determined from electronic structure theory.
Temperature Dependence

If $J=0$, an ensemble of local moments will be randomly oriented.

Application of external field $H$ provides a driving force to align spins, which competes against entropy (favoring disorder).

Interactions $J$ effectively act as an internal field, analogous to $H$, causing the material to spontaneously magnetise.

$M$ drops with $T^0$, and vanishes at the Curie temperature.
Electronic Structure and Critical Temperature

Mean-field theory gives a simple (reasonably reliable) estimate of $T_c$. For one kind of atom $T_c$ is:

$$k_B T_c = \frac{2J_0 S^2}{3}, \quad J_0 = \sum_j J_{ij},$$

$$\hat{H} = -\sum_{ij} J_{ij} \hat{S}_i \cdot \hat{S}_j$$

To make $T_c$ large, $S$ and $J$ should be large. $S$ and $J$ are determined by the electronic structure.

For $J$ to be large, atomic wave functions must strongly overlap with neighbors.

The middle of the 3d series (half filled $d$ bands have pretty large $S$ and large $J$. $T_c$ can be quite high ($T_c = 1400K$ for Co)

Rare earths, with their $4f$ electrons, can have very large $S$, (Gd has $S=7$), but the wave functions are atomic-like: they weakly overlap with (and couple to) neighbors. $T_c = 292K$ for Gd
High $T_c$ is a necessary but not sufficient condition

If $T_c$ and $M_s$ were the only materials properties that mattered, transition metals would make excellent magnets

A third critical property is the uniaxial magnetocrystalline anisotropy $K$. Without anisotropy, there is no hysteresis.

<table>
<thead>
<tr>
<th>Material</th>
<th>$T_c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>1043</td>
</tr>
<tr>
<td>Co</td>
<td>1400</td>
</tr>
<tr>
<td>Ni</td>
<td>627</td>
</tr>
<tr>
<td>Gd</td>
<td>292</td>
</tr>
<tr>
<td>EuO</td>
<td>69</td>
</tr>
<tr>
<td>Dy</td>
<td>88</td>
</tr>
<tr>
<td>MnBi</td>
<td>630</td>
</tr>
<tr>
<td>MnSb</td>
<td>587</td>
</tr>
<tr>
<td>CrO$_2$</td>
<td>386</td>
</tr>
<tr>
<td>Fe$_2$O$_3$</td>
<td>948</td>
</tr>
<tr>
<td>Nd magnets</td>
<td>583-673</td>
</tr>
<tr>
<td>SmCo magnets</td>
<td>993-1073</td>
</tr>
</tbody>
</table>
Origin of Magnetocrystalline Anisotropy

Crystals possess a magnetic “easy axis” and a “hard axis.”

Originates from coupling between spin and orbital angular momentum (a consequence of relativity). Spin-orbit coupling links the electron spins to the crystalline lattice: the energy of the spin system depends on its crystallographic orientation.

As the spin system rotates between two axes of low energy (easy axes) they must pass through a harder axis. There is energy barrier to rotating the collective system, called the uniaxial Magnetocrystalline Anisotropy (MAE).

Anisotropy can arise from other sources (esp shape anisotropy) but MAE is the most important one.
Making MAE large

Roughly, $\Delta E(\theta) \sim K \cos(\theta)$.

MAE is a complicated business!
No one single parameter characterizes it.
It originates from a relativistic effect that couples spin and orbital angular momentum ($\lambda L \cdot S$). $\lambda \sim Z^2$ (Landau) because $e^-$ move fastest near nuclei. Larger $Z$, faster $e^-$ accelerate near nucleus.
But ... the MAE is not $E(\lambda L \cdot S)$, but the change $\Delta E(\lambda L \cdot S)$ as the crystallographic orientation rotates through angle $\theta$.
Usually $\Delta E(\lambda L \cdot S) \ll E(\lambda L \cdot S)$ --- it is several orders of magnitude smaller in high-symmetry systems.

Rules of thumb:
Heavier elements have larger $E(\lambda L \cdot S)$: helps to increase $\Delta E(\lambda L \cdot S)$
Lower symmetry tends to increase $\Delta E(\lambda L \cdot S)$
Demagnetizing Fields Create Magnetic Domains

The “demagnetizing energy” (or the “dipolar” energy) $E_d$ is positive and increases with volume. $E_d$ can be lowered by splitting up the magnetic element into smaller regions because the dipolar fields from region of opposing $M$ partially cancel.

A region where the moments all point in a single direction is called a domain or a Weiss domain. The small region separating domains is called a domain wall.

The domain wall has an energy cost $2J$ in proportion to the wall area $A$. $2JA$ competes with the reduction in $E_d$; it is a complicated function of the shape of the domains. The globally optimum domain shape and thus global minimum energy structure depends on the shape of the specimen.
Size of a Domain Wall

The three energies, exchange coupling $J$, MAE $K$, and dipolar fields $H_d$ give rise to rich magnetic behavior. Dipolar fields depend on sample shape and can give rise to remarkable domain patterns.

Example: Interplay between $J$ and $K$ governs energy, mobility, shape of domain wall.

Energy cost to rotate neighbouring spins

$$-2JS_1 \cdot S_2 = -2JS^2 \cos \theta \approx JS^2 \theta^2 \text{ if } \theta \text{ is small.}$$

Net rotation: angle $\pi$ over $N$ sites $\Rightarrow \theta = \pi/N$.

Competes with MAE. Energy of layer $i$ depends on angle $\varphi_i$ as

$$E_i \approx K \sin^2 \varphi_i$$

Minimize

$$\sigma + \frac{NK}{2} \Rightarrow \text{DW width}$$

$$\delta = Na = \pi S \sqrt{\frac{2J}{Ka}}$$

Energy / area:

$$\sigma = \frac{JS^2 \pi^2}{N^2 a^2}$$

Sum MAE for $N$ sites

$$\sum_i K \sin^2 \varphi_i \approx \frac{NK}{2}$$
Hysteresis and Domain Walls

Application of external field $H$ causes the crystal to favor regions where spins are aligned with $H$.

Walls will move enabling domains parallel to the field to grow. But their motion is restricted because there are barriers to motion, because of the anisotropy. Barriers are the reason for hysteresis.

Barriers depend in a complicated way on the relative strengths of exchange coupling $J$, MAE $K$, and dipolar fields $H_d$, and also by inhomogeneities (e.g. defects), which can “pin” them. Thus domain-wall motion is intricately linked to microstructure.
MAE in MnBi: a detailed theoretical study

MnBi is one possible replacement for Neodymium magnets. A study of its magnetic properties highlights how challenging it is. Up to 628K MnBi is ferromagnetic and forms in the NiAs structure (hexagonal). $a$ and $c/a$ vary significantly with $T^\circ$.

Calculations from Antropov et al, PRB 90, 054404 (2014)
The MAE $K$, depends strongly on $T$, even changing sign. Around room temperature $T^0$, $K$ is respectable ($\sim 0.9 \text{mJ/m}^3$) but its $T^0$-dependence makes it unsuitable. $K$ originates from pockets of energy bands just above and below the (very complicated) Fermi surface.

Pockets contribute to $K$ with varying signs. As $T^0 \uparrow$, $c$ and $a$ change, which modifies pocket shapes and their individual contributions to $K$. 

PRB 90, 054404
What makes $K$ large?

What controls $K$ is a rather involved question, even for a simple two-level model system. For a discussion of $K$ in a model context, see the following paper:

PHYSICAL REVIEW B 92, 014423 (2015)

Band-filling effect on magnetic anisotropy using a Green’s function method

Generically speaking you need:

1. Large $\lambda L S \Rightarrow$ favors heavy elements (remember $\lambda \sim Z^2$)
2. Large orbital moment. $\lambda L S$ provides the driving force to induce orbital moments (it breaks symmetry between $m$ and $-m$) but it tends to be strongly quenched by the crystal field.
3. Large crystal field is needed to generate anisotropy in $\lambda L S$, i.e. change in $\lambda L S$ as spins rotate --- comes from anisotropy of the crystal field.
Why do f shell elements have large K?

f elements can have large anisotropy (generically) because:
(1) they are heavy (large $\lambda L \cdot S$)
(2) the crystal field is large enough to interact with $\lambda L \cdot S$ but
(3) it is not large enough to quench the orbital moments.

Hard to beat!

Anisotropic 3d compounds can yield respectable $K$, e.g. FeNi can be made to form in an ordered L1₀ structure (AKA tetrataenite) with $K=1.3$ mJ/m³.
So do CoPt, FePt, and MnAl with roughly similar $K$.

Compare to $K=4.7$ mJ/m³ ($Nd_2Fe_{14}B$) and $K=6.5$ mJ/m³ ($YCo_5$)
Reducing Rare Earths in Hard Magnets

Strategy 1: Reduce the Nd and Dy/Tb content in Neodymium magnets while retaining its good properties (e.g. preserving $H_c$ up to 200°C)

It may be possible to reduce the Nd and Dy content by ~50% largely by processing that better aligns grain boundaries (G.H. Yan et al, J. Phys. Conference Series 266, 012052 (2011)).

Domain wall motion and pinning are closely connected to inhomogeneities (microstructure) --- extremely complicated as small long-range forces interact with strong short-ranged ones. Optimise through Edisonian trial-and-error approach.
Finding Replacements for Neodymium Magnets

Strategy 2: Find a “good enough” replacement that doesn’t contain rare earths or other rare elements such as Pt.

Nd$_2$Fe$_{14}$B is already nearly ideal, so probably not realistic to equal it.
Transition Metal based Hard Magnets

A zoo of alternatives have been studied. Each is unique. Some (YCo$_5$, Mn$_2$Ga, FePt) have materials abundance issues. Others (Fe$_{16}$N$_2$, MnBi) have stability issues.

<table>
<thead>
<tr>
<th>Structure</th>
<th>MnAl</th>
<th>MnBi</th>
<th>Mn$_2$Ga</th>
<th>FeNi</th>
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<tbody>
<tr>
<td></td>
<td>$L_1^0$</td>
<td>$B_8^1$</td>
<td>$D_{022}$</td>
<td>$L_1^0$</td>
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<tr>
<td>$T_c$ (K)</td>
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<td>633</td>
<td>$&gt;770$</td>
<td>$&gt;820$</td>
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<tr>
<td>$\mu_0M_s$ (T)</td>
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<td>0.73</td>
<td>0.59</td>
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<tr>
<td>$K_1$ (MJ m$^{-3}$)</td>
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<tr>
<td>$\kappa$</td>
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<table>
<thead>
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<td>$\alpha''$</td>
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<td>0.56</td>
<td>0.45</td>
<td>1.7</td>
<td>2.7</td>
</tr>
</tbody>
</table>

Conclusions

A great deal is understood generically about what ingredients are needed to make a hard magnet.

The rare earths combined with Fe or Co are special because they have an optimum mix of high magnetisation $M_s$, essential for high $|BH|_{\text{max}}$.

Large $\lambda L \cdot S$ combined with enough, but not too strong, coupling to crystal fields to make possible large MAE $K$.

TM provides strong magnetic exchange interactions $\Rightarrow$ high $T_c$.

RE free magnets are not likely to be fully replaced soon. The search is on for intermediate quality magnets with good combinations of $(M_s, |BH|_{\text{max}}, K, T_c)$. Many promising candidates, each with its own unique set of challenges.

No magic bullet yet!