# What are bulk superconducting magnets?

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# Why are bulk superconducting materials interesting?

- They can 'trap' or 'screen' large magnetic fields > 17 T (between a stack of 2 disks)
  - Allowing a considerable increase in the power density of electric motors
- We anticipate that they are the key to a major technological breakthrough
- Replacing the classic NdFeB permanent magnets...

M. Tomita et M. Murakami., « High-temperature superconductor bulk magnets that can trap magnetic fields of over 17 tesla at 29 K », Nature 421, pp. 517-520, 2003.





#### **Bulks HTS synchronous motors** 50 kW, 5000 rpm @30 K (still under test @ GREEN)



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#### **Presentation Outline**

- Superconductivity basics
- Bulk High-Temperature Superconductors (HTS)
  - Types of bulk materials, fabricating, characterizing, magnetization
- How does Pulsed Field Magnetization process work?
  - In real life cases





Zero resistance

Discovered at the university of Leiden in 1911 by Kamerlingh Onnes who was working on the resistivity of Mercury.

• *T*<sub>c</sub>: Critical temperature concept





Heike Kamerlingh Onnes







• Meissner effect (1933)

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- Characterized by a perfect diamagnetism (B = o)
- Below critical field  $H_c$  (or first critical field  $H_{c1}$ )





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- Levitation by Meissner effect ≠ Flux pinning effect
  - <u>https://youtu.be/JIjzJKnpahA</u>



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#### Walter Meißner

# Superconductivity basics

- Meissner effect
  - Eddy currents flow on surface of superconductor to maintain internal B = o

{Ref.} Brandt, E. H., & Mikitik, G. P. (2000). Meissner-London currents in superconductors with rectangular cross section. *Physical review letters*, 85(19), 4164.

- These currents are reversible (related to B or not dB/dt)
- The typical size of these currents corresponds to the London's penetration depth  $\lambda$

Туре	Material	$T_{\rm c}$ [T]	<i>H</i> <sub>c</sub> (o) [T]	<i>H</i> <sub>c1</sub> (o) [T]	H <sub>c2</sub> (o) [T]	λ(o) [nm]
Ι	Pb	7.2	0.080	-	-	48
II	Nb	9.2	0.200	0.170	0.4	40
II	Nb <sub>3</sub> Sn	18	0.540	0.050	30	85
II	NbN	16.2	0.230	0.020	15	200
II	MgB2	40	0.430	0.030	3.5	140
II	YBCO	93	1.400	0.010	100	150



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#### Mixed state

- Only practical operating state possible for superconductors
- Above first critical field Hc1 (a few mT for HTS)
- Below second critical field  $H_{c_2}$  (from a few T to a hundred T)
- The magnetic field penetrates the material in multiples of flux quantum  $\Phi_0 = 2,067\,833\,667 \times 10^{-15}\,\text{Wb}$
- The fluxons (or vortices) are not independent, they form a triangular network,  $\frac{2\Phi_0}{2}$ known as the Abrikosov network a =





field in NhSes at 1.8 K. The scan range is about 6000 Å. The Kévin Berger, 5th Superconductivity school held in Mexico, 6 November 2021



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# Superconductivity basics $\frac{\text{Magnetic flux}}{\text{density}} \boldsymbol{B} = \mu_0 \left( H + \boldsymbol{M} \right) \text{Magnetization}$



- Reversible effect
  - No pinning
- Irreversible effect
  - Some vortices are pinned
  - **Charles Bean** Explained by Bean's critical state model which introduces a critical current density  $J_c$

{Ref.} Bean, C. P. (1962). Magnetization of hard superconductors. Physical review letters, 8(6), 250.



Applied magnetic field









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- Influence of vortex motion
  - (1) Zero resistance
  - (2)  $V \propto I^n$
  - (3)  $V \propto (I I_{\rm ff})$
  - (4)  $V \propto I$
  - Only true for isothermal experiment

{Ref.} Osorio, M. R., Morales, A. P., Rodrigo, J. G., Suderow, H., & Vieira, S. (2012). Demonstration experiments for solid-state physics using a table-top mechanical Stirling refrigerator. European journal of physics, 33(4), 757.





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- *E*(*J*) models vs Experiments
  - Percolation model

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$$E(J) = \begin{cases} 0, & \text{if } J \leq J_{c,\min} \\ E_c \left( \frac{J - J_{c,\min}}{J_c - J_{c,\min}} \right)^n, & \text{if } J > J_{c,\min} \end{cases}$$

• Power law  

$$E(J) = E_c \left(\frac{J}{J_c}\right)^n$$
 with  $E_c = 1 \,\mu\text{V/cm}$ 



Fig. 1. Experimental data points measured on a 4 mm wide SuperPower tape, as well as E - J curves for the four different models defined in Section III-A. On this scale, the power law and the percolation models are undistinguishable.





- Both models agree on a common area
- The differences are significant
  - in the low electric field zone
- What can change the current's relaxation

{Ref.} Sirois, F., Grilli, F., & Morandi, A. (2018). Comparison of constitutive laws for modeling high-temperature superconductors. *IEEE Transactions on Applied Superconductivity*, 29(1), 1-10.



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Experimental data — Power law model



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- 3 critical quantities
  - Critical temperature *T*<sub>c</sub>
  - Critical field  $H_c$  (or  $B_c$ )
  - Critical current density  $J_c$  (or  $I_c$ )
    - Related to pinning forces!
    - There are several definitions...
      - Electrical voltage criterion  $E_c = 1 \,\mu\text{V/cm}$
      - Magnetization loop width
- It defines a critical surface





- The most famous are probably the (RE)BaCuO
  - (RE) = Rear Earth elements
    - Y, Gd, Nd, Eu, Dy...
- Trapped magnetic field is achieved by pinning penetrated magnetic field (quantized flux lines)
- By means of induced macroscopic electric currents



A large, single grain YBaCuO bulk superconductor from <u>ATZ GmbH</u> (top side machined)





- Trapped field and magnetization increase with sample volume
  - *t* = thickness, *a* = radius, and *z* is the height above the top surface



#### Trapped field analytical models

- Easier to deal with & faster
- Based on Bean's model and Biot-Savart law
- Simplified geometries
- Constant and uniform J<sub>v</sub> is assumed
- Magnetostatic approximation
  - Current's paths are assumed to be known
- In other cases, numerical simulations are required!
  - This is still a quite complicated task to do
  - Very difficult to precisely predict the whole HTS behavior





- Candidate materials of possible interest should
  - Carry large current density over large length scales
  - Be "insensitive" to application of large magnetic fields
    - Field dependence of critical current density  $J_c(B)$
  - Have high critical temperature
    - to reduce cooling system constraints



Example of magnetic field dependence of the critical current density of a YBCO



- Currently, there are 2 main candidates for bulk HTS magnets
  - (RE)BaCuO
    - $T_{\rm c} \approx 90 96 \, {\rm K}$
    - Manufacturing is complicated / slow
    - Homogeneity difficult to achieve
  - MgB2
    - Discovered in 2001,  $T_c = 39$  K
    - Easy to manufacture
    - Cheap and light-weight



MgB2 cylinders by Mg-RLI process (Edison SpA)











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• Applications are determined by high  $J_c$  and  $H_{irr}$  rather than by high  $T_c$ 



{Ref.} Koblischka-Veneva, *et al.* (2019). Comparison of Temperature and Field Dependencies of the Critical Current Densities of Bulk YBCO, MgB2, and Iron-Based Superconductors. *IEEE Transactions on Applied Superconductivity*, 29(5), 1-5.



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#### Processing Bulk HTS (courtesy of BSG, Cambridge, UK)



Sintered YBCO

2 μm

- Simple sintering of (RE)BCO powder in bulks does not result in the best possible material
- Early attempts at sintered bulk materials were disappointing
  - Low  $J_c$
  - Granularity is a problem & grain boundaries = 'weak-links'
  - Microcracking





# **Processing Bulk HTS**

- Grain boundaries can be avoided using a seeded peritectic growth process
- All (RE)BCO melt processes are based on the following peritectic reaction that occurs around 1015°C:

$$2(\text{RE})\text{Ba}_{2}\text{Cu}_{3}\text{O}_{7-\delta} \leftarrow (\text{RE})_{2}\text{Ba}\text{Cu}_{5} + (\text{Ba}_{3}\text{Cu}_{5}\text{O}_{8})$$
(123)
(211)
Liquid





# **Processing Bulk HTS**

- Top Seeded Melt Growth (TSMG):
  - Seed with the same lattice structure
  - Phase stability with the BaCuO melt
  - Higher melting temperature → initializes growth & controls orientation
  - $T_p(\text{Sm-123}) \sim 1054 \text{ °C or } T_p(\text{Nd-123}) \sim 1068 \text{ °C} > T_p(\text{Y-123}) \sim 1015 \text{ °C}$



Namburi, D. K., *et al.* (2018). A robust seeding technique for the growth of single grain (RE)BCO and (RE)BCO–Ag bulk superconductors. Superconductor Science and Technology, 31(4), 044003.





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### Processing Bulk HTS – TSMG

#### 1. Mixing

Precursor powders of desired composition are mixed together using a mortar and a pestle. 2 hours are used for a mix of 200g.



Typical composition: 70wt% Y-123 + 30wt% Y-211 + 0.1wt %Pt (grain-refining agent)

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#### 2. Pressing + Seeding

The mixed precursor is weighed and poured into a die of desired dimensions. The powder inside the die is pressed using a press. A seed is then placed on top of the surface of the pellet. Seed



#### Pressed pellet

Pressure applied: 20 kN-50 kN for a pellet 20-40 mm in diameter

#### 3. Melt-processing

The pressed pellet with seed is then put into a furnace. The heating profile is as follows:

Temperature



Time

(1) Sintering ~ 940°C 15 min - 24h
(2) Decomposition ~1040°C 1h
(3) Nucleation from 1000°C to 960°C
~ 2h with respect to the crystal growth of that is about 0.1 mm/h (a slower rate of temperature decrease is better)

#### 4. Oxygenation

Necessary in order to obtain the orthorhombic structure (superconducting phase)

Heat treatment under oxygen for approx. 150h - 300h at 450°C







#### Processing Bulk HTS (Infiltration Growth and Top Seed Textured)

{Ref.} Chaud, X., Bourgault, D., Chateigner, D., Diko, P., Porcar, L., Villaume, A., ... & Tournier, R. (2006). Fabrication and characterization of thin-wall YBCO single-domain samples. Superconductor Science and Technology, 19(7), S590.

https://iopscience.iop.org/0953-2048/19/7/S33/media/video1.mpg





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#### Processing Bulk HTS – Growth Sector





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#### Processing Bulk MgB<sub>2</sub>

- Manufacturing process
  - Unconventional Sintering with SPS
  - = Spark Plasma Sintering
  - = Field Assisted Sintering Technology
  - = Pulsed Electric Current Sintering
  - Temperature: RT-2200 °C
  - Speed: o-700 °C/min
  - Force: 0.5-250 kN
  - Atmosphere: Air / N2 / Ar
  - Size (mm): 8 / 15 / 20 / 30 / 36 / 40 / 50 / 80





# Characterization of Bulk HTS

- Strong inhomogeneities for some materials
- Difficult to characterize the whole pellet
- Sample has to be cut in an orthorhombic shape
  - Typical size: 2 mm x 2 mm x 0.5 mm





{Ref.} Chen, D. X., & Goldfarb, R. B. (1989). Kim model for magnetization of type-II superconductors. *Journal of Applied Physics*, 66(6), 2489-2500.

{Ref.} Sanchez, A., & Navau, C. (2001). Critical-current density from magnetization loops of finite high-Tc superconductors. *Superconductor science and technology*, *14*(7), 444.

{Ref.} Philippe, M. M. (2015). *Magnetic properties of structures combining bulk high temperature superconductors and soft ferromagnetic alloys* (Doctoral dissertation, Université de Liège, Liège, Belgique).

$$U_{\rm c}(H) \left[ {\rm A/m^2} \right] = \frac{\Delta M(H) \left[ {\rm A/m} \right]}{a[{\rm m}] \left( 1 - \frac{a}{3b} \right)}$$

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for  $H_p < H < H_{max} - H_p$ 



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# Characterization of Bulk HTS

- Assessing the average performance of the entire sample
  - Force measurements with PM in front of a bulk
    - HTS is only magnetized on the surface
  - Determination of the critical current density from magnetic field measurements at the center of the top surface
    - Practical way to measure without cutting
    - Need to reach the full penetration field

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• Influences of the Hall probe location and sweep rate are taken into account



PM

{Ref.} Douine, B., Berger, K., *et al.* (2018). Determination of the complete penetration magnetic field of a HTS pellet from the measurements of the magnetic field at its topcenter surface. IEEE Transactions on Applied Superconductivity, 28(4), 1-4.





# Magnetization of Bulk HTS

- 3 magnetization processes
  - Zero Field Cooling (ZFC)
  - Field Cooling (FC)
  - Pulsed Field Magnetization (PFM)
- To trap 5 T, need at least 5 T or higher
  - FC and ZFC require large coils and long magnetizing times
  - PFM is the only practical process for applications / devices





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# Magnetization of Bulk HTS by FC

- Trapped field records with FC process (between a stack of 2 disks)
  - 17.2 T @ 29 K, YBaCuO, two 26.5 mm Øx 15 mm (M. Tomita *et al.*, 2003)
  - 17.6 T @ 26 K, GdBaCuO, two 25 mm  $\Phi$ x 15 mm (J. Durrell *et al.*, 2014)
  - 3.05 T @ 77 K, GdBaCuO, single 65 mm Øx 19 mm (S. Nariki *et al.*, 2005)
  - 5.4 T @ 12 K, MgB2, single 20 mm Øx 8 mm (G. Fuchs *et al.*, 2013)
  - 5.6 T @ 10 K, MgB2, two 28 mm  $\Phi$ x 10 mm (T. Naito *et al.*, 2020)
    - 6.6 T expected @ 4.2 K without <u>flux jumps</u>

• 6.78 T @ 12 K (4.1 T rem.), MgB2, six 20 mm Øx 4 mm (B. Badica *et al.*, 2020)



### Magnetization of Bulk HTS by FC

#### • MgB<sub>2</sub> samples with $Ge_2C_6H_{10}O_7$ (Repagermanium) 6x MgB<sub>2</sub> Ø 20 mm, h = 6x4 mm

{Ref.} Badica, P., Aldica, G., Grigoroscuta, M. A., Burdusel, M., Pasuk, I., Batalu, D., ... & Koblischka, M. R. (2020). Reproducibility of small Ge2C6H10O7-added MgB2 bulks fabricated by ex situ Spark Plasma Sintering used in compound bulk magnets with a trapped magnetic field above 5 T. Scientific Reports, 10(1), 1-11.





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# What are flux jumps?

- Magneto-thermal instabilities
  - Due to the low thermal diffusivity and heat capacity of some superconductors...
    - MgB2
    - But not only...
  - It's worse with
    - low temperatures
    - using large samples
    - high sweep rates





MgB2 pellet (Mg-RLI) of 50 mm  $\Phi$  x 20 mm





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### Magnetization of Bulk HTS by FC

- Trapped field records with FC process (between a stack of 2 disks)
  - 17.6 T @ 26 K, GdBaCuO, two 25 mm  $\Phi$ x 15 mm (J. Durrell *et al.*, 2014)







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# Magnetization of Bulk HTS by FC

- What limits performance?
  - At 17 T, internal stresses are ~ 90 Mpa
    - Stress scales as ~ 0.282 B<sup>2</sup>
  - Leads to practical maximum trapped field of 7-9T in unreinforced samples as tensile strength can be < 10 MPa
  - Common failure mode seems to be a simple crack across sample
- How to overcome this?
  - Add 15 wt% AgO converted to Ag during processing, filling voids/cracks
    - Can improve fracture strength by an order of magnitude (a few MPa  $\rightarrow$  10 MPa)
  - Shrink-fit Stainless Steel onto sample, achieves ~250 MPa interface pressure
  - Tomita et al. used Carbon Fiber/Epoxy Nature 421, 517-520 (2003)









# Magnetization of Bulk HTS by PFM

- Trapped field records with PFM process (top surface)
  - 5.2 T @ 28-50 K, GBaCuO, single 45 mm  $\Phi$ x 15 mm (H. Fujishiro *et al.*, 2006)
  - 3.2 T @ 40-65 K, GBaCuO, single 30 mm Ø x 15 mm (M. Ainslie *et al.*, 2016)
  - 1.1 T @ 20 K, MgB2, single 22 mm  $\Phi$ x 15 mm (H. Fujishiro *et al.*, 2016)
  - 1.61 T @ 20 K, MgB2, single 30 mm Øx 19 mm (T. Hirano *et al.*, 2020)





# Magnetization of Bulk HTS by PFM

- What limits performance vs FC?
  - Heat induced during PFM!
- Who to optimize dynamics of magnetic flux during PFM process
  - Pulse magnitude/ pulse duration,
  - Number of pulses,
  - Operating temperature(s),
  - Type of magnetizing coil(s),
  - Use of ferromagnetic materials...

{Ref.} Fujishiro, H., Naito, T., Furuta, D., & Kakehata K. (2010). Temperature measurements in small holes drilled in superconducting bulk during pulsed field magnetization. Physica C: Superconductivity and its applications, 470(20), 1181-1184.

drilled holes

75

GSE

bul

SUS

T1~T3





# Magnetization of Bulk HTS by PFM

- PFM is the only practical process for applications using bulk magnets
  - We have to deal with strong magnetic field variations
  - And induced currents in all conducting parts
- A coupled electro-thermal modeling is mandatory
- But how does PFM process work?





# How does PFM process work?

- Pulsed Field Magnetization
  - What do we usually need
  - How it works
  - Equation of current/applied field
  - Main characteristics
  - Examples of operational setups
  - Summary
- In real applications?





# **Pulsed Field Magnetization?**

• Usually, charge of capacitors and then short circuit



- It can also be a short-circuited coil made on a
  - Transformer
  - Electric machine







#### Capacitors bank...











60 kJ – 10 kA 40 mF to 120 mF – 1000 V



5 mF – 2000 V



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#### About the current waveform



#### Influence of bulk HTS on inductance

Bulk pellet surrounded by a circular coil (circuit coupled problem)



{Ref.} Kapek, J., Berger, K., Koblischka, M. R., Trillaud, F., & Lévêque, J. (2019). 2-D numerical modeling of a bulk HTS magnetization based on H formulation coupled with electrical circuit. IEEE Transactions on Applied Superconductivity, 29(5), 1-5.

- From the point of view of the coil
  - A bulk HTS during PFM is almost equivalent to air



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# **Different regimes**

• Overdamped response  $\zeta > 1$ 

$$i(t) = A_1 \exp\left(-\omega_0\left(\zeta + \sqrt{\zeta^2 - 1}\right)t\right) + A_2 \exp\left(-\omega_0\left(\zeta - \sqrt{\zeta^2 - 1}\right)t\right)$$

• Underdamped response  $\zeta < 1$ 





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 $\zeta = \frac{R}{2} \sqrt{\frac{C}{I}}, \ \alpha = \frac{R}{2I}, \ \omega_0 = \frac{1}{\sqrt{IC}}$ 



#### Examples

#### Capacitors bank influence





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#### Underdamped regime





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#### Examples of operational setups



{Ref.} Ainslie, M. D., Fujishiro, H., Mochizuki, H., Takahashi, K., Shi, Y. H., Namburi, D. K., ... & Cardwell, D. A. (2016). Enhanced trapped field performance of bulk high-temperature superconductors using split coil, pulsed field magnetization with an iron yoke. *Superconductor science and technology*, 29(7), 074003.



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#### Niigata University (T. Oka and J. Ogawa)







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### Morioka University (H. Fujishiro)









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#### Magnetization by stator windings (4 poles)





motor. *IEEE transactions on applied* superconductivity, 21(3), 1171-1174.



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### **Operational test rig @ GREEN**

in situ magnetization







Bsat > 2.3 T Hc < 200 A/m  $\rho$  = 4.10<sup>-7</sup>  $\Omega$ .m density 8120 kg/m<sup>3</sup>  $\alpha$  = 10.10<sup>-6</sup> K<sup>-1</sup>

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# YBaCuO samples with SS Ring

#### • Sample and SS ring are glued together with Stycast 2850 FT

35 mm diameter - 12 mm high from CAN SUPERCONDUCTORS s.r.o.



31 mm diam. - 17 mm high from ATZ GmbH





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# Coil design and realization

- Coil should be as close as possible to the sample
  - Better magnetic coupling and optimized size
  - Copper foils of 0.2 mm thick
  - Kapton tape for turn-to-turn insulation
- Our first aim was to thermalize the coil with the 2nd stage of the crycooler using Stycast molding
  - Easy to manufacture and mount













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# Coil design and realization

- Some problems occurred during the experiences
  - Stycast can not handle the Lorentz force during the pulsed magnetization (~ 2.5 GPa @ 10 kA)
  - Heat is not well extracted (it's still epoxy resin...)
- Fiber Glass + Araldite impregnation
- Cooling using the 1st stage of the cryocooler











# **Other issues**

- High voltage up to 2 kV and high current densities > 1 kA/mm<sup>2</sup>
  - Working with HV in small environment that requires good thermal connections and compactness is not easy...
- Thermalization of the coil leads through the 1st stage of the cryocooler was made with Aluminium Nitride
  - (Cu / AlN / Cu) sandwich with copper plates of 20 mm x 80 mm
  - AlN properties: k > 150 W/(m.K),  $\rho > 1010$   $\Omega$ .m,  $E_{\rm d} = 15$  kV/mm, density of 3300 kg/m<sup>3</sup>,  $\alpha = 4.6 \times 10^{-6}$  K<sup>-1</sup>











# **Other issues**

- During PFM, the coil attempts to place at the center of the iron cylinder
  - The resulting force on the coil holder @ 10 kA reaches 13 kN

~ the weight of my Peugeot 5008

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#### Other homemade coils











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# Short summary

- PFM usually needs capacitors
- Current waveform/magnetization strongly depends on
  - The inductance and so of the environment
- HTS bulk needs to be tightly coupled with the coil
- About the use of iron
  - Increases the applied field for a given current
  - Increases the homogeneity of the applied field
  - Increases *L*, so  $t_{\max} \uparrow$  and  $i_{\max} \downarrow$
- Design of the coil is essential in HTS bulks applications







# Thank you for your attention!

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