Nuclear demagnetization refrigeration

Journey to below 0.1 mK



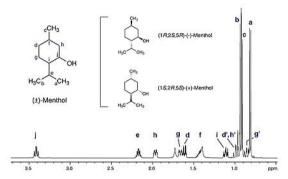
Juha Tuoriniemi Demag Day - Grenoble, France - 20.9.2021

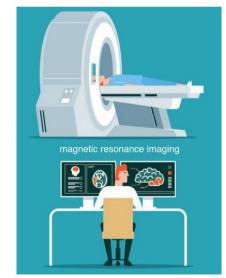
The setting

- Nearly half of the stable isotopes possess a nonzero nuclear magnetic moment (odd number of protons or neutrons)
- The most common application of this is in NMR spectroscopy or imaging

 As paramagnetic (disordered) ensembles such nuclei may also be suitable for thermometry and/or demagnetization cooling

1D PROTON SPECTRUM







Brief comparison of nuclear to electronic magnetism

Nuclei	Electrons
Large # of isotopes with $\mu \neq 0$	Transition elements with $\mu \neq 0$
Conceptually simple: pure spin I	Spin S and orbital angular momentum L must be considered
Weak: μ is of the order of nuclear magneton $\mu_n = e\hbar/2m_n$	Of the order of Bohr magneton $\mu_B = e\hbar/2m_e \sim 1836 \mu_n$
Simple thermodynamics: high temperature approximations apply	Complications due to tendency for spontaneous magnetic ordering
Spin-spin interactions can usually be ignored (except quadrupole effects)	Interactions are the crucial limiting factor



To perform demagnetization cooling, an initial state with decent entropy reduction ΔS must be prepared

- Maximum molar entropy for spin *I* is $S_m = R \ln(2I+1)$
- At temperature T in magnetic field B with $x = \mu B/k_BT$ we have $\Delta S \approx R(I+1)/(6I) x^2$
- Taking $\mu \sim 2\mu_n$ and I=3/2, we need $x \sim 0.7$ for $\Delta S/S_m \sim 10\%$ corresponding to B/T ratio of about 1000 T/K
- This translates to the need of $B \sim 10 \text{ T}$ and $T \sim 10 \text{ mK}$
- These figures can be achieved by a combination of a superconducting magnet and a good dilution refrigerator



- We seek pure ("nonmagnetic") metals with desired spin properties ($\mu \ge \mu_n$ and ~100% abundance)
- Some isotope candidates

element	Spin I	μ/μ_n	B_c if SC
²⁷ Al	5/2	3.64	10 mT
51 V	7/2	5.14	0.1 T
^{63/65} Cu	3/2	2.3	-
⁹³ Nb	9/2	8.07	0.2 T
$^{113/115}In$	9/2	5.5	30 mT



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- => use Cu (or Al)

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Typical figures

- 10-100 mol copper in 9 T field precooled to 10 mK
- Heat capacity up to $C \approx nR (I+1)/(3I) x^2 \approx 250 \text{ J/K}$
- To remove corresponding enthalpy in two days the dilution fridge must remove heat at the rate of ~1.5 μW
- Before demagnetization, the nuclear stage must be isolated from the precooling stage. This is done by a superconducting heat switch. (In its normal conducting state HS usually imposes a significant thermal impedance affecting the precooling performance)

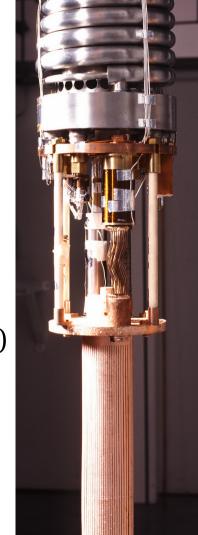




Demagnetization

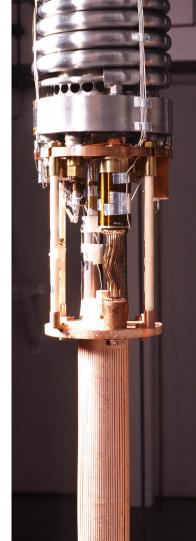
- Adiabatic change of magnetic field preserves the heat capacity (in practice to a few percent). Reduce the field to e.g. 30 mT to get nuclear temperature below 50 μ K
- Make the demag in 10-20 h to avoid excessive eddy current heating (the nuclear stage must be made from thin slabs to reduce the effect)
- At such low temperatures the coupling from nuclear spins to anything else (electrons, phonons, your sample) becomes an issue. Spin lattice relaxation time τ_1 in Cu will be $\kappa/T_e = 1.2 \text{ sK/50 } \mu\text{K} \sim 7 \text{ hours (Korringa law)}$
- Warming up under heat load of ≤ 5 nW takes weeks





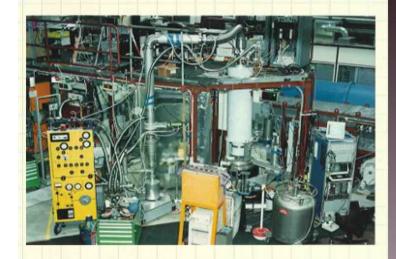
Heat leaks, thermal contacts, and thermometry

- These finally decide between the success or failure of the endeavor
- Potential sources of heat leak are many and often remain largely unknown
- Large thermal gradients may exist within the system and between the sample and the refrigerant
- Thermometry, too, is a big challenge below 1 mK
- For the extreme, it is possible to run two nuclear demagnetization stages in cascade

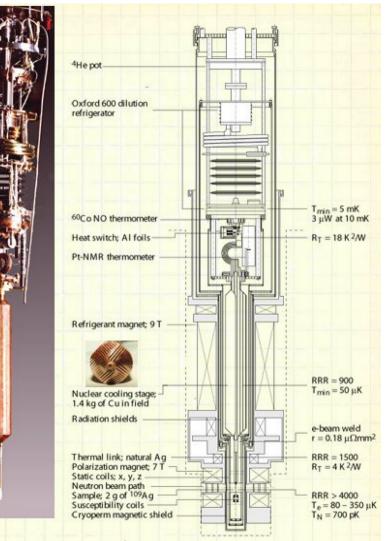




picokelvin installation at HMI in Berlin (operational from 1992 to 1996) Neutron diffraction on nuclear spin ordering in silver









Aalto University School of Science

Some other ways to utilize nuclear spins

- Dynamic polarization and demagnetization in rotating frame using dilute electronic magnetic impurities in dielectric solids
 - CaF₂, LiH, Ca(OH)₂, ...
- Hyperfine enhanced nuclear magnets (van Vleck paramagnets)
 - In some suitable compounds with electronic quadrupole ground state m = 0 external magnetic field is amplified by an induced electronic moment thus enhancing the magnetic field experienced by the nuclei up to factor of 10 100
 - Most famous example is PrNi₅



European Microkelvin Platform (EMP)

- See https://emplatform.eu for details
- Eight access giving sites in Europe

Aalto University, Helsinki
Basel University
CNRS Grenoble
Heidelberg University
Lancaster University
Royal Holloway University of London
Slovak Academy of Sciences Kosice
Technical University Vienna



