



ULTRA-LOW TEMPERATURES NUCLEAR ADIABATIC DEMAGNETIZATION

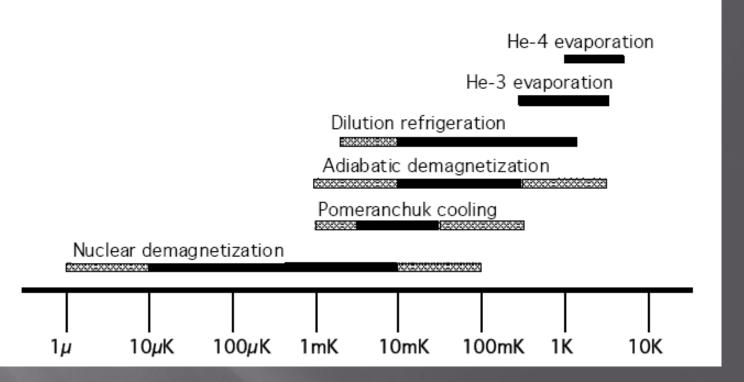
European Advanced Cryogenics School

Henri Godfrin (and Juha Tuoriniemi in absentia)

European Advanced Cryogenics School - Chichilianne 2011

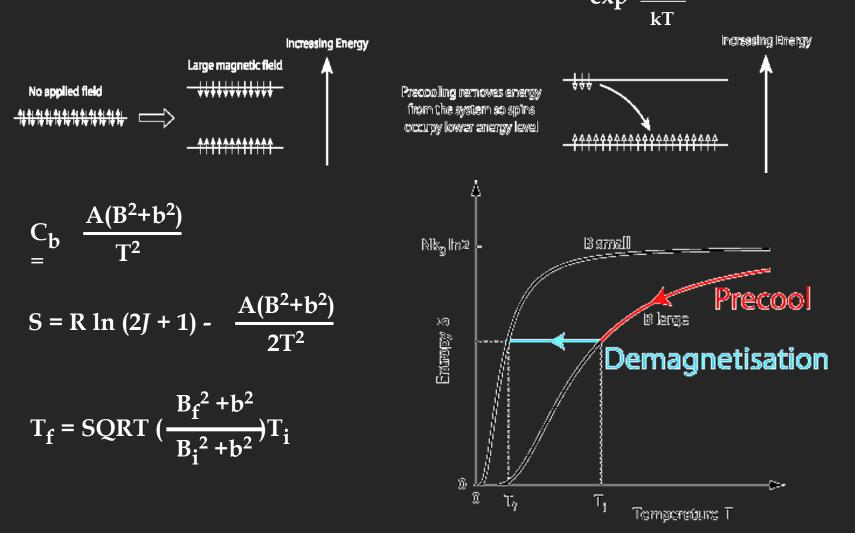
Refrigeration below 1K

Dilution refrigerator
 Adiabatic demagnetization
 Pomeranchuk cooling

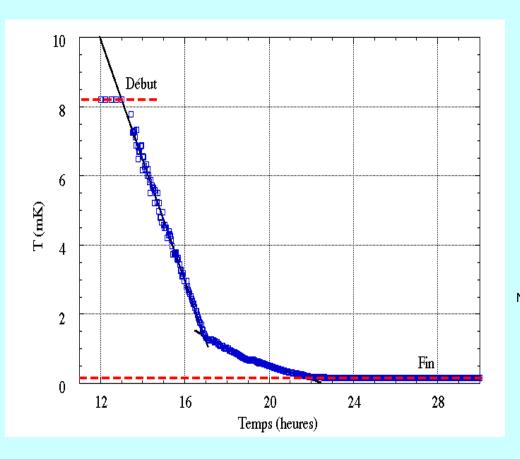


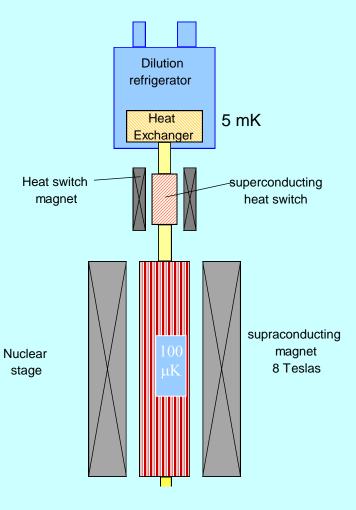
Nuclear demagnetization

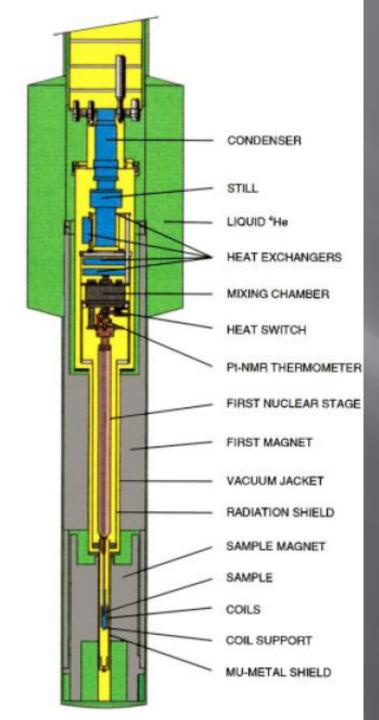
Suggested by Gorter (1934) and Kurti and Simon (1935) Realized by Kurti, Robinson, Simon and Spohr (1956) Cooled Cu nuclear spins to about 1 μ K, while lattice and electrons remains in 12 mK. exp <u> μ H</u>

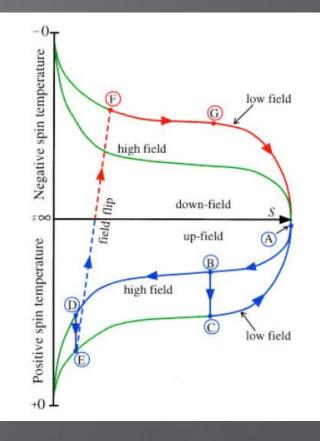


Nuclear demagnetization refrigerator











Material Cu PrNi₅

Geometry Nuclear stage can be done from wires, bulk road or plates.

Heat switch Sn Zn Pb Al



Cooling superfluid ³He down to 100 μK





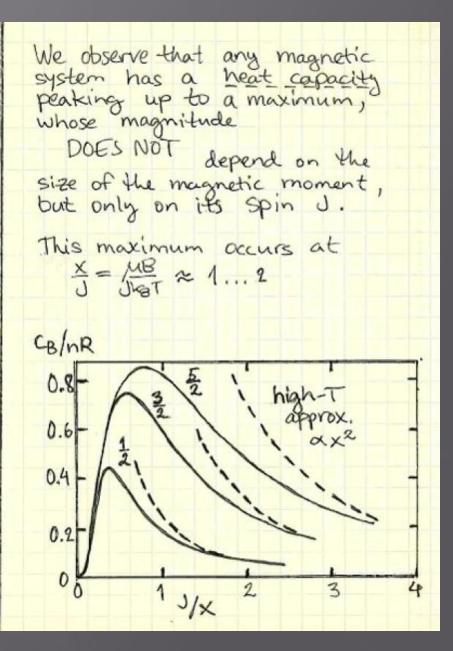
- ③ Reduce the magnetic field (adiabatically)
 Bi → Bf
 - The same entropy is obtained when $T_f = \frac{B_f}{B_i} T_i < T_i$!

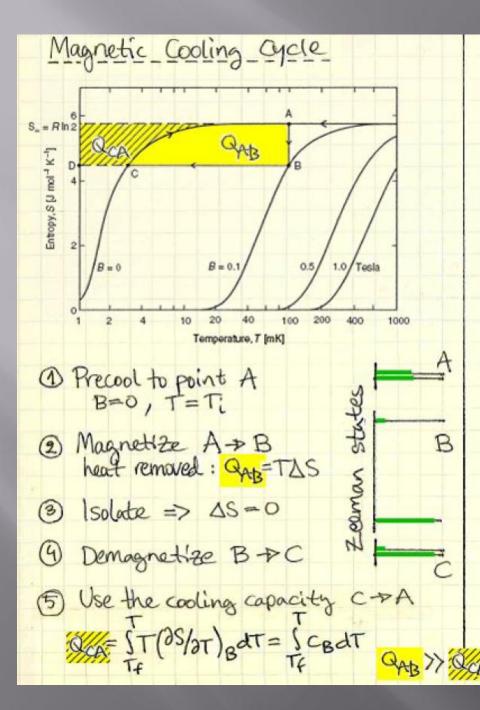
what if BF = 0? At that limit effective fields other than the external applied field come into play:

Spin-spin interactions contiducte by b~ <u>kBTc</u>; Btot~ $\sqrt{B^2 + b^2}$ where Tc is the magnetic ordering temperature

Other useful quantities: · susceptibility X X=MOM ~ JM Momsatx = 7 where $\lambda = J + 1 n R \mu_0 \mu^2_{k^2}$ is the Curie constant $([\lambda]=k)$

Now, eg. $C_{B} = \frac{J+1}{3J} nRx^{2} = \frac{V\lambda}{M_{0}} \left(\frac{B}{T}\right)^{2}$





To succeed:

- magnetic entropy must dominate
 Ti << To (Debye, phonons)
 - Tikk TF (Fermi, electrons)
 - any other entropy in the system causes <u>nonadiabaticity</u>
- · magnetic ordering temperature Tc must be lower than the target T
 - To due to dipole-dipole interactions scales as μ^2/r^3
 - metals exhibit exchange interaction also; can be large
- initial condition (Bi, Ti) should be such that the spin entropy has been reduced as much as possible. This gives cooling capacity.

In practice AS:~ 5... 50% Smax

Paramagnetic Satts
(magnetic metals are ruled out
because of the strong exchange
interaction
$$\Rightarrow$$
 Tc is high)
We play with the
electronic moment
 $\mu = g\mu_{B}J$
with the Landé factor
 $g = \frac{3}{2} + \frac{1}{2} \frac{S(S+1) - L(L+1)}{J(J+1)}$
and the Bohr magneton
 $\mu_{B} = \frac{eK}{2m_{e}} \approx 9.27 \cdot 10^{24} J/T$

Elements with unpaired electrons at inner shells produce local magnetic moments in the solid state

- 3d transition elements - 4F vare earth metals

Often their magnetism is too strong (high Tc) so that they are diluted by forming an ionic compound (salt) with loads of crystal water Tc $\propto \frac{\mu^2}{r^3}$ goes down

Ion to be used can be

Mn2+, Fe 3+, Cn3+, Ce3+, ...

Nuclear demagnetization
Often the nucleus carries a
magnetic moment - al you
need is an unpaired proton
or neutron

$$\mu \sim \mu_N \approx 5.05 \cdot 10^{27} \text{ J/T}$$

 $\left(\frac{\mu_N}{\mu_B} = \frac{\text{me}}{\text{mn}} \sim \frac{1}{1840}\right)$
 $\Rightarrow T_C \leq 0.1 \mu \text{K}$
Therefore:
- one can reach conciderably
lower temperatures
- no need to dilute the moments
 \Rightarrow good moment density
 μ more heat capacity

- metals can be used good thermal conductivity easy thermal contracts The challenge: - small $\mu \Rightarrow$ need large Bi/Ti e.g. STi = 10mkBi = 6T. => AS= 5% Smax for Cu - to maintain heat capacity one cannot demag to zero or even close to b (typically b~ 0.1 mT) Often Bf~ 10 ... 100 mT

Material candidates?

A) Pure metal for sufficient thermal conductivity

B) Reasonable moment, abundance (maybe large spin)

$$\begin{array}{c|c} e.g. & \mu/\mu_{N} & I \\ \hline 27Al & 3.64 & 5/2 \\ \hline 63,65 (\mu & 2.3 & 3/2 \\ \hline 113,115 In & 5.5 & 9/2 \end{array}$$

STMn & 59Co are very nice nuclei, too, but the host metal is ferromagnet => these cannot be demagnetized

93Nb & 51V are also very good isotopes, but they are in superconductors, Bc = 0.2/0.1T ⇒ demag only that far

All & In are superconductors, too, but Bc~ 10/30.mT only

c) Cubic lattice or spin I= 1/2

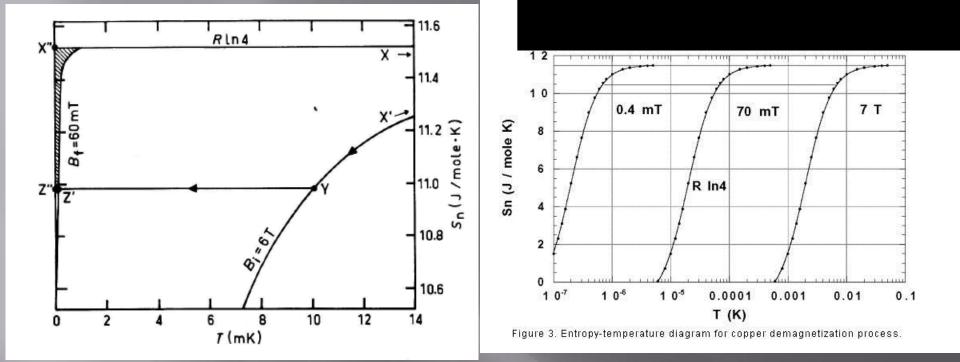
in other cases electric quadrupole interaction may be strong and contributes to the effective field

· In (TETR) by ~ 0.25T is thus valed out

We are left with

() Copper

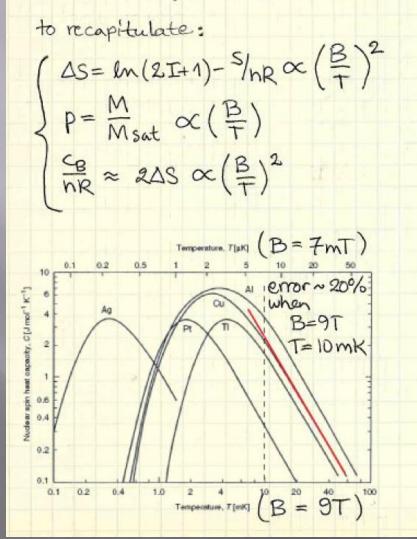
(Al might do in some cases)

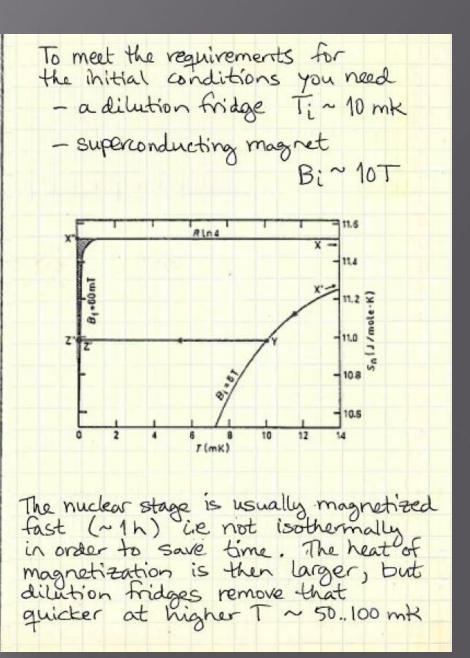


1. Adiabatic

- 2. Large Curie constant Cu 0.57 μ K (Pt 0.019, In 1.11); PrNi₅ x 17
- 3. In equilibrium with electrons (small Korringa constant) Cu 1.1 secK (Pt 0.03)
- 4. No superconducting transition
- 5. Small residual field (PrNi5 ~ 1mK)
- 6. High thermal conductivity
- 7. Desirable mechanical and metallurgical properties (heat leak)

When working with nuclear spins it is usually sufficient to use the simple high-T expansions



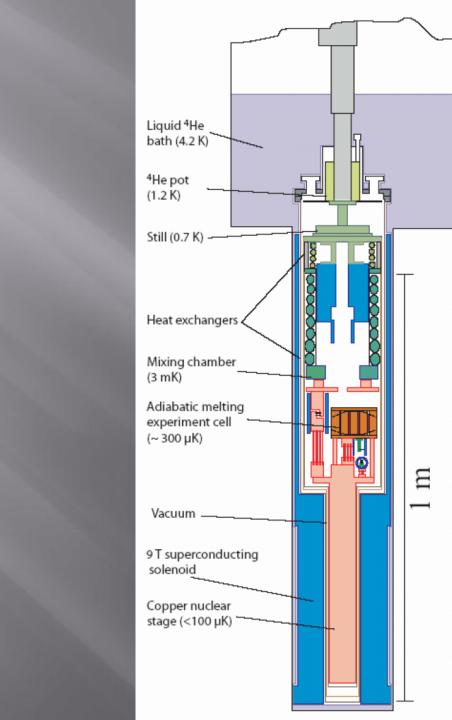


A copper nuclear stage n≈10100 mol (1 ser	
1) Precooling with a dill in B=69T to Ti takes some 15	~ 10 mt ~ 10 mt
2) Thermal isolation is by a superconducting	
3) Demagnetization to I takes ~ 10 h (1 day limited by eddy cur	Br~10100 mT); speed rent heating
4) Depending on the heat can maintain T& 1 1 day over a	
The superconducting magnet must be persistable. During phase 1) there is a	Power supply Room temperature Power s.
Current of order 100A. During phase 4) the greatest stability is required.	Liquid helium Heater COLOCOLO

Phase 1) Precooling · power of the dilution fridge Qd = ata-qd where a = 84 ns /(molk2) and gd is the background load of the Fridge (depends on ing too) Base temperature To = Vg1/a · thermal resistance of the heat switch (normal state) R~ 2... 20 K2 (Appically) Whereby T2-T2= 2RQNS-d · heat of magnetization QNS= - CB dt ; $C_B = \frac{C}{T_{NS}^2}; C = \frac{1}{32} \left(\frac{M_B}{K_B}\right) nR = \frac{M_B}{M_B} B^2$ These determine the precooling time

... Precooling time $t(T) = (\frac{1}{\alpha} + 2R) = (\frac{1}{2}(\frac{1}{2T_0} lm \frac{T+T_0}{T-T_0} - \frac{1}{T})$ $T \approx \left(\frac{1}{a} + 2R\right) \frac{c}{3T3}$ B=9T nG = 100 mol Esijäähdytys YKI, 20.-22.4.1998, 1500 µmol/s Model: Precool $1/a = (5.6 \pm 0.2) \text{ K}^2/\text{W}$ $R = (10.54 \pm 0.04) \text{ K}^2/\text{W}$ c = 26 mJK (fixed) $T_{0} = (5,7 \pm 0,2) \,\mathrm{mK}$ T(mK)2000000000000 TMC 10 TNS dn/dt = 1.5 mmol/sdn/dt = 0.6 mmol/s 10 100 t (h)

For example: $\dot{n}_3 = 1 \frac{mmol}{s} \Rightarrow \frac{1}{a} = 12 \frac{k^2}{w}$ $R=10\frac{K^2}{M}$; $T_0=3mK$ nNS = 100 mol (Ca) Bi= 9T >> c=26,5 mJK results in ±(10mK)=3.105 s ~ 3.5 days Phase 2) heat switch We assumed R~10 K2 in normal state A good switch has a switching ratio > 105 (T=To) We can then estimate the leakage through the SC switch Q~ 2:105R = 4.5 PW This is perfectly ok



superconducting Heat switch Good electrical conductivity in normal state (Wiedemann - Frantz -> thermal conductivity) Poor phonon anductivity i.e high TD (Debye temperature) Field coil an Cu Nb-shield em W Possible materials: BeCu - AI Bc Tp TC AL 1.2K 10mT 394 K Zh 0.9 5 Sh 3.7 30 234 ~200 Alyminum has "extremity pure" Cu ----exellent material properties but K[Wem^{*}K¹] bui must overcome the oxide problem on the surface 10 · diffusion welding 10" AL-Ch @ 500°C 10-5 Epibond 121 for ~ 15 min AO in vacuum 100 200 6.00 1000 under compression Temperature, 7 [mK]

Phase 3) demagnetization
Optimal demag. profile?
a simple model:
- constant background heat
leak
$$\hat{q}_{NS}$$

- eddy current heating $y\hat{B}^2$
 $\Rightarrow \hat{a}_{NS} = \hat{q}_{NS} + y\hat{B}^2$
where $\begin{cases} r^2 \sqrt{RRR} & for a \\ \frac{d^2}{16} \sqrt{\frac{K^2}{16r}} & RRR & for a \\ \frac{d^2}{16r} \sqrt{\frac{K^2}{16r}} & \frac{K^2}{16r} & \frac{M}{16r} \\ \frac{d^2}{16r} & \frac{K^2}{16r} & \frac{K^2}{16r} \\ \frac{d^2}{16r} \\ \frac{d^2}$

so: typically a cylindrical block is sliced into vertical plates or it is bundled from plates ~1mm thick



in an optimal demag que = SB2 if quis is const. => linear profile B= V9NS/8 ; topt= / 1/gns (Bi-Bf)

The measure for nonadiabaticity: $A(\frac{B}{T}) = 2 \sqrt{\frac{3}{2}} \ln \frac{B}{B} \frac{1}{2}$

However, this is usually unacceptably slow - several days for reasonable \dot{q}_{NS} To save time (tam < topt) the best demag profile is <u>parabolic</u> B(t) = $a(\frac{1}{4}d_m)^2 - b(\frac{1}{4}d_m) + Bi$ with ($a = Bi + B_f - \sqrt{4}BiB_f + \frac{9}{4}NS + \frac{2}{4}d_m$ $b = 2Bi - \sqrt{4}BiB_f + \frac{9}{4}NS + \frac{2}{4}d_m$

Losses for parbolic demag: $\Delta(\stackrel{\text{P}}{=}) = \underbrace{4\mu_{0}}_{\lambda} \left[\alpha \underbrace{+}_{\text{dm}} + \sqrt{\hat{q}_{NS}} \operatorname{arroinh}\left(\sqrt{\frac{\hat{q}_{NS}}{s}} \underbrace{+}_{2\sqrt{B};B_{P}}\right) \right]$ For example: $\begin{cases} n_{CL} = 100 \text{ mol} \\ d = 2mm \\ W >> d \\ (RRR = 1000) \end{cases} = 7 V = 10,6 \frac{WS^2}{Tz}$ $q_{NS} = 10 \text{ nW}$ $rac{2}{3} = 10 \text{ nK}$ $rac{2} = 10 \text{ nK}$ $rac{2} = 10 \text{ nK}$ $rac{2} = 10 \text$ vs. parabolic tam= 10h => losses ~ 3% (or) F linear demag in time tam
⇒ ∆(^B/_T)=^{Ho}/_N ln(^{Bi}/_B)[^{gNS}/_B tamt ^V(Bi-B_f)] with the= 10h => losses ~ 5% (not bad)

9T -> SOMT linear in 10h ~ 15mT/min best parabolic in 10 h begin ~27 mT/min end ~ 3 mT/min Take into account also: - possible vibrational heating Q~B² -> sweep faster at beginning - magnetoresistance g=g(B) => f=f(B) =) sweep faster at beginning (Po can be >10 in IDT)

Phase 4) T= 100 MK Heat capacity to spend $Q = \sum_{T_F} c_B dT = \frac{T+1}{3T} \frac{nR}{T_F} \left(\frac{MB_f}{K_B}\right)^2$ ~ 10 mJ

Question: the cooling process operates on the nuclear spins, how is that connected to your sample?

The nuclear spins themalize among themselves due to spin-spin interactions in a time scale T2 (spin-spin relaxation time, remember NMR) Te is typically ~ 0.1 ... 10 ms (fast) => spin system can be concisidered to be in thermal equilibrium distribution

Energy exchange between the nuclear spins and conduction electrons (or lattice ? phonons) is characterized by spin-lattice relaxation time Ty

For motals Korringa law: $T_1 = \frac{K}{T_e}$

typically K~ 0.01 ... 10 sK when Te~50,4K => Ti~ 200 ... 2.105 s >> T2

THEREFORE : nuclear spin Th = Te electron temperature temperature

Electrons respond fairly quickly: Te ~ A Ko ~ 1ms $(c_e \propto T, k_e \propto T)$

Phonon specific heat is negligible (CRXT3) => TPh ~ Te

The Hermal Load (or heat leak) usually directs to the electron system > Te>Th

Lets analyze this problem: Nuclear relaxation is governed by $\frac{dM}{dt} = -\frac{1}{2}(M - M_0) \left(\begin{array}{c} NMR \\ B \omega ch \end{array} \right)$ where M≈ II Msat HB a th and the equilibrium value Mo ≈ I+1 MSAt MB x Te $\Rightarrow \frac{d}{dt}(\frac{1}{t_n}) = -\frac{1}{t_1}(\frac{1}{t_n} - \frac{1}{t_e})$ Since $d(\frac{1}{t}) = -\frac{1}{t^2} \frac{dT}{dt}$ and $T_1 = \frac{K}{T_e}$ $\Rightarrow \frac{dT_n}{dt} = (T_e - T_n) \frac{T_n}{\kappa}$

IF there is a heat load Q (a > electrons > nuclei; ce << cn) we get dt = a = MoTn² dt = cp = VABf² Q $\Rightarrow \overline{Te} = 1 + \frac{M_0 k Q}{VA B_f^2} = 1 + \frac{Q}{Qn}$ we shall call $\dot{Q}_n = \frac{VABf}{\mu_0 \kappa} \sim 1 \mu W \begin{cases} B_f = 50 mT \\ n_{cu} = 100 mol \\ K_{cu} = 1.2 s \kappa \end{cases}$ the "normative load" depending on the - material (2, k) - conditions (Bf) - size (V) Te deviates from The severely when a approaches an (or an approaches à)

Good advice: mount a heater on your fridge If a is constant, then Th-1 decreases linearly: It helps calibrating many things dTh-=-Mage=-Q We may have an uncalibrated thermo-meter following a know dependence, e.g. measured reading $m(Te) = a X_m(Te) = a \frac{Am}{Te} = \frac{a}{Te}$ So does Tet : t= anta te One takes readings with and without additional heating to obtain $\Rightarrow dt = \frac{\dot{a}_n}{\dot{a}t} dt = \frac{-\alpha}{\kappa(\dot{a}+\dot{a}_n)}$ $\int \dot{m}_1 = -\frac{\dot{\kappa}(\dot{a}+\dot{a}_n)}{\kappa(\dot{a}+\dot{a}_n)}$ =) {à $m_2 = -\frac{\alpha(\dot{a}+\dot{a}e)}{\kappa(\dot{a}+\dot{a}e+\dot{a}n)}$ 1/ Tn Heat input = 0.1Qn 1/Te If à, àe «an you get a plot nverse temperature m slope is a/kan (kan= VABf is known) Time To not have Te>>To we want small k (and of course smallest à) || KCu=1.2.5K || Kpt=0.035K ---- Dae

to keep cold you must eliminate all IF you desire the lowest possible. Te possible heat loads there is an optimum Be to stop the demag: - conduction (support, leads, heat switch, ...) · thermal anchoring is important { Te,f = 1+ Moka Th,f = 1+ VAB2 -thermal vadiation (shield at T<1K) - remnant gas in vac. (PHe < 10-10 Pa) - vibrations! (building, LHe indewar, pumps, ...) $\left(T_{n,f}=T_{i}\frac{B_{f}}{B_{i}}\right)$ · big muss (tons) · Mexible support (air springs, flexible babs,-) => Tef = Ti (Bf + Moka i Bf + VA Bf > eigenfrequencies < 1HZ - radioactivity & cosmic radiation This has a minimum at - electric and magnetic tields · shielding, filtering Bf= / Worka i.e. Qn=Q - internal time dependent loads · H2 ortho-para conversion At this point Te,F=2Tn,F · amorphous substances (plastics), Typically 10-50pW/mol (at best 5-10pW/mol) Usually, however Temperature, T[µK] this is unpractically 20 low field. Often a compromise has 10 to be made to 20 F Sustain low le long enough 12 18 24 $(C \propto B^{2})$ Magnetic field, B [mT] Qin= 50 pW mol-1 Remember, we assumed Q/n= 100 pW mol (B>>> ; for Cu b= 0.34mT) 40 20 80 100 60 Time, t[h]

Hyperfine enhancement

In some metal compounds of rare-earth ions (like Pr^{3t} in PrNits) the electronic moment vanishes in Zero field -J=1 (or 2,3,...) - lattice is non-cubic and the ground state is m=0 Due to the guadrupole effect, any titie magnetic field can induce a nonzero effectivement (<u>Van Vleck</u>) => the nucleus feels a strong magnetic field Bn= B(1+K) where K~ 10.... 100 is the hyperfine enhancement factor (K=11.2 for 141 Pr in PrNic) Then it is easy to polarize the nuclei even at modest temperature and external magnetic field For example: Ti = 25mt Z= AS ~ 70% PrNi5 Bi = 6T J= Smax 70% The nuclei can still be demagnetized because the electronic moment disappears in B=0

+ Benefits: • easy initial conditions • great heat capacity / volume

· fast spin-lattic relaxation T,

- <u>Prawbacks</u>: • difficult materials • bad conductivity • ordering temperature ~ 0.1 ... 1 mK This is an intermediate from

This is an intermediate from paramagnetic salts to purely nuclear spin systems

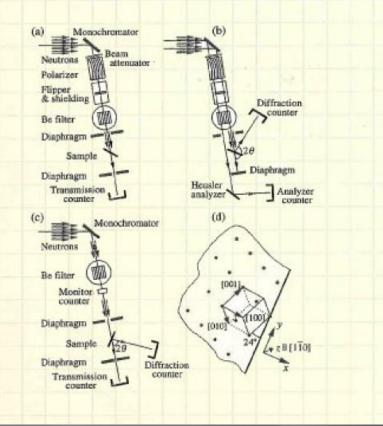
In early days these vere often used between dilution fridge and a copper nuclear stage to assist in lowering Ti

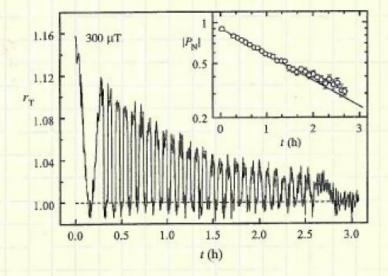
Not much in use nowadays due to increased power of dilution indges

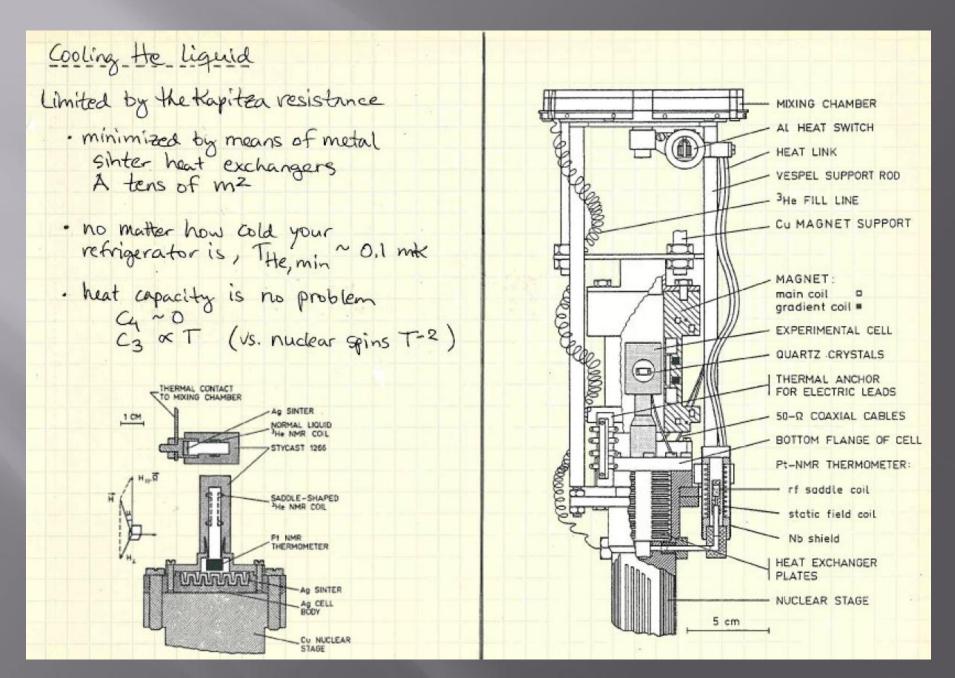
In a rough environment this might be your choice if you only need T<1mk . combined with a cryocooler & DR?

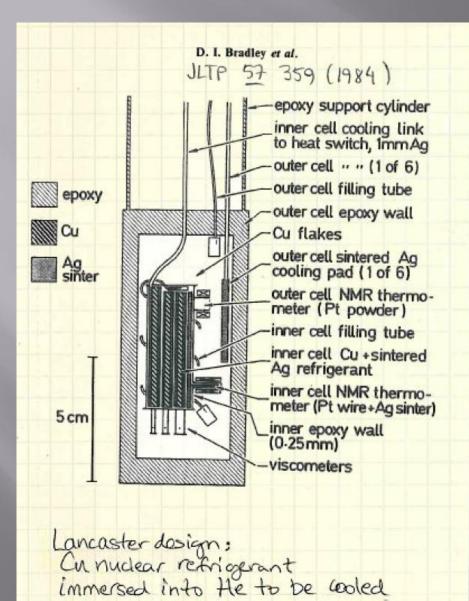
An example of heat load measurement: Cu stage of the HMI-cryostat: 21 molot Cu 110 Ag 1 = 24.6 s The specimen ~ 29 of silver, which was exposed to a neutron beam ~ 2.105/cm²s EB EY 95.2 2.9 0.3 (E) 22 4.4 4.5 0.7 110 46 Pd ~ 80% of neutrons become absorbed This produces a radioactive daughter nucleus 28 Epy 0.5 6.8 n, 7 27 wich decays emitting B & & 110 48 Cd 109 Ag These deposit heat to Lattice & electrons 30 Background ~ 1 nW Radioactivity (~ 2µCi) gives 14 nW 109 Ag 25 20 (MU) 15 1.0Neutron beam Ag 10 Open Closed 5 65Cu T,R 0.8 9.0 1/T (mK¹) dQ/dt = 14.8 nW3.0 3.5 4.0 4.5 2.5 2 (Å) dQ/dt = 1.0 nW0.4 5 10 15 20 25 30 0 t (h)

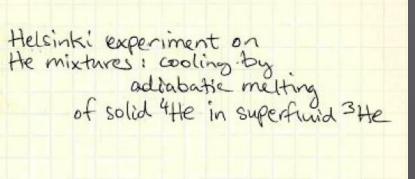
Special purpose thermometry by transmission of a neutron beam (T-nanok!)

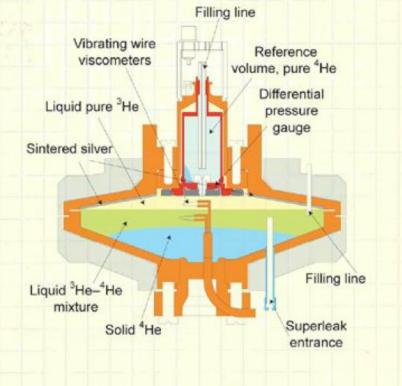




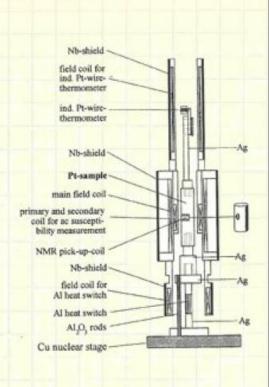


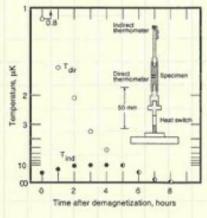






The record low temperatures have been achieved by operating two nuclear stages in cascade lowest nuclear spin temperatures: Silver ~ 500 pK (LTL, TKK) rhodium ~ 100 pK (LTL, TKK) Good to have reasonably large K, no heat switch between the Stages Lowest cond. electron temperatures: Copper ~ 5µK (Lancaster) platinum ~ 1µK (Bayrenth) Silver wires to Al heat Tin leads switch ¥ anchored Touc to warmest Tempererature, plate Warmest Cu olate PENMR 10 Heat switch thermometer wire buncle 15 20 Middle Cu plate Heater 100 Coldest Cu plate Silver Time after demagnetization, days support WITOS



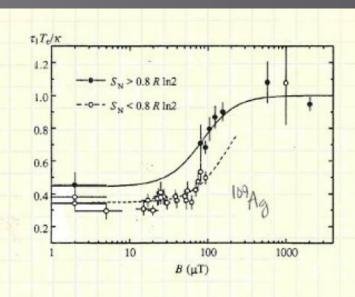


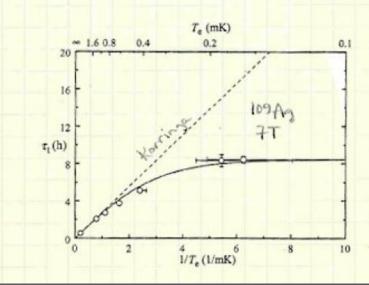
In cascade nuclear refrigeration the simple treatment above fails

· spin-spin interactions must not be omitted in low fields

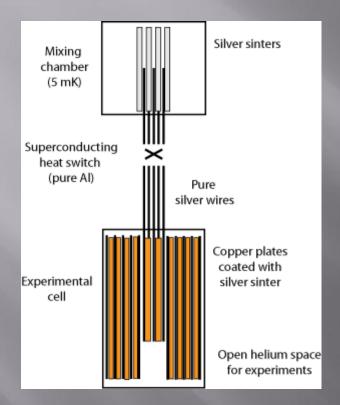
$$\left(e_{g}, T_{1} \approx \frac{k}{T_{e}} \frac{B^{2} + b^{2}}{B^{2} + \alpha b^{2}}; \alpha = 2...3\right)$$

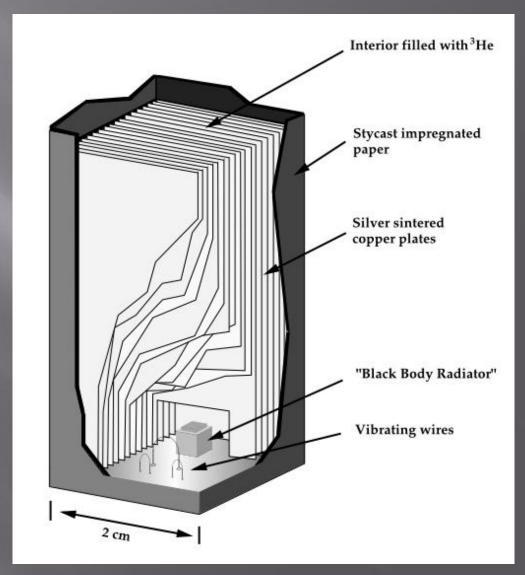
 µB is no longer small in comparison with kate
 ⇒ Korringia law is modified





Lancaster - type



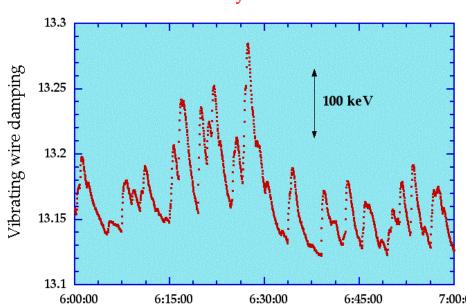


Superfluid ³He bolometry

Cosmic rays detection

6:45:00

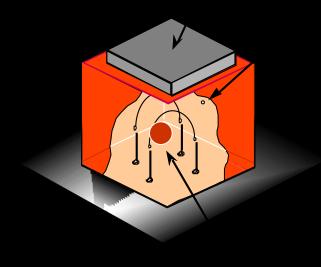
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Time (hours)

6:15:00

6:00:00



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

