Adiabatic **nuclear** demagnetization

– Many isotopes carry a nonzero nuclear spin
– **Very small moment**, of order nuclear magneton

$$\mu_n = \frac{e\hbar}{2m_n} = \frac{m_e}{m_n}\mu_B \approx 5.05\cdot 10^{-27}\ J/T \qquad \mu_B = \frac{e\hbar}{2m_e} \approx 9.27\cdot 10^{-24}\ J/T$$

=> **T_c ~ pK ... µK**

- Very low temperatures can be achieved
- Moments need not be diluted
  => good moment density (more heat capacity)
- Metals can (must) be used
  – Good thermal conductivity
  – Reasonable thermal contacts

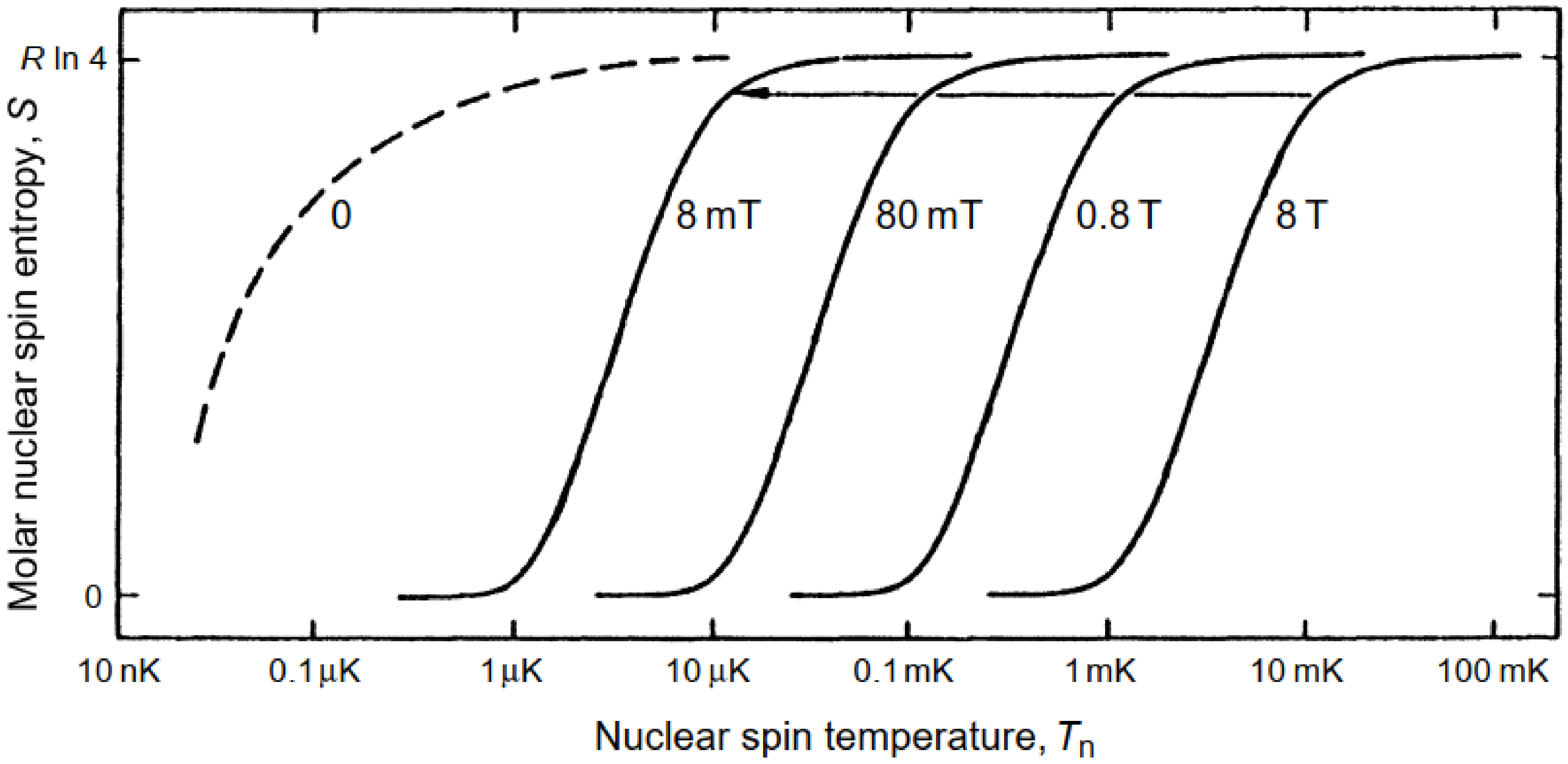


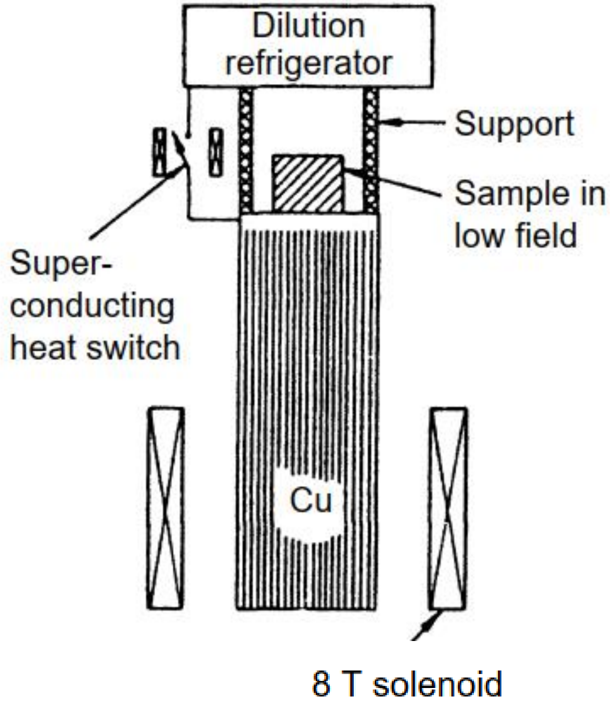
Fig. 10.1. Molar nuclear spin entropy S of Cu nuclei in various magnetic fields as a function of temperature. The arrow indicates an adiabatic demagnetization process from 8 T to 8 mT. The full nuclear spin disorder entropy of copper nuclei with $I = 3/2$ is $R \ln(4)$ as indicated on the vertical axis. The dashed line is the nuclear spin entropy of Cu in zero field according to the data of [10.1]; it indicates the spontaneous antiferromagnetic ordering of the Cu nuclear spins

The challenge

Small μ => need very **large B_i/T_i**

For example: $\begin{cases} T_i = 10 \text{ mK} \\ B_i = 6 \text{ T} \end{cases}$

=> $\boldsymbol{\Delta S/S_{max}}$ **= 5% for Cu**



To maintain sufficient heat capacity, one cannot demag. to zero field (for nuclear magnets $b \sim 0.1$ mT)

Usually $B_f \sim 10 \ldots 100$ mT (limits achievable T_f)

DR & SM

To meet the required initial conditions, one needs

– dilution refrigerator (DR), $T_i \sim 10$ mK

– superconducting magnet (SM), $B_i \sim 10$ T

The magnet has to be in **PERSISTENT current** state during precooling to T_i at B_i ($I_i \sim 100$ A) and during experiments for best possible stability and to **minimize boil-off**

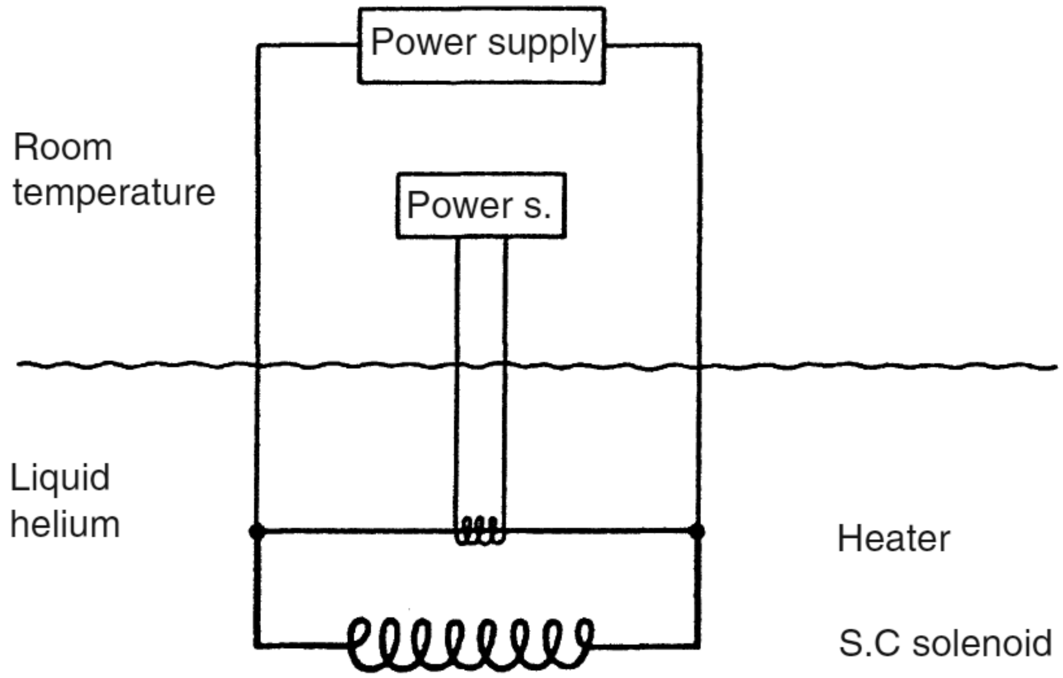
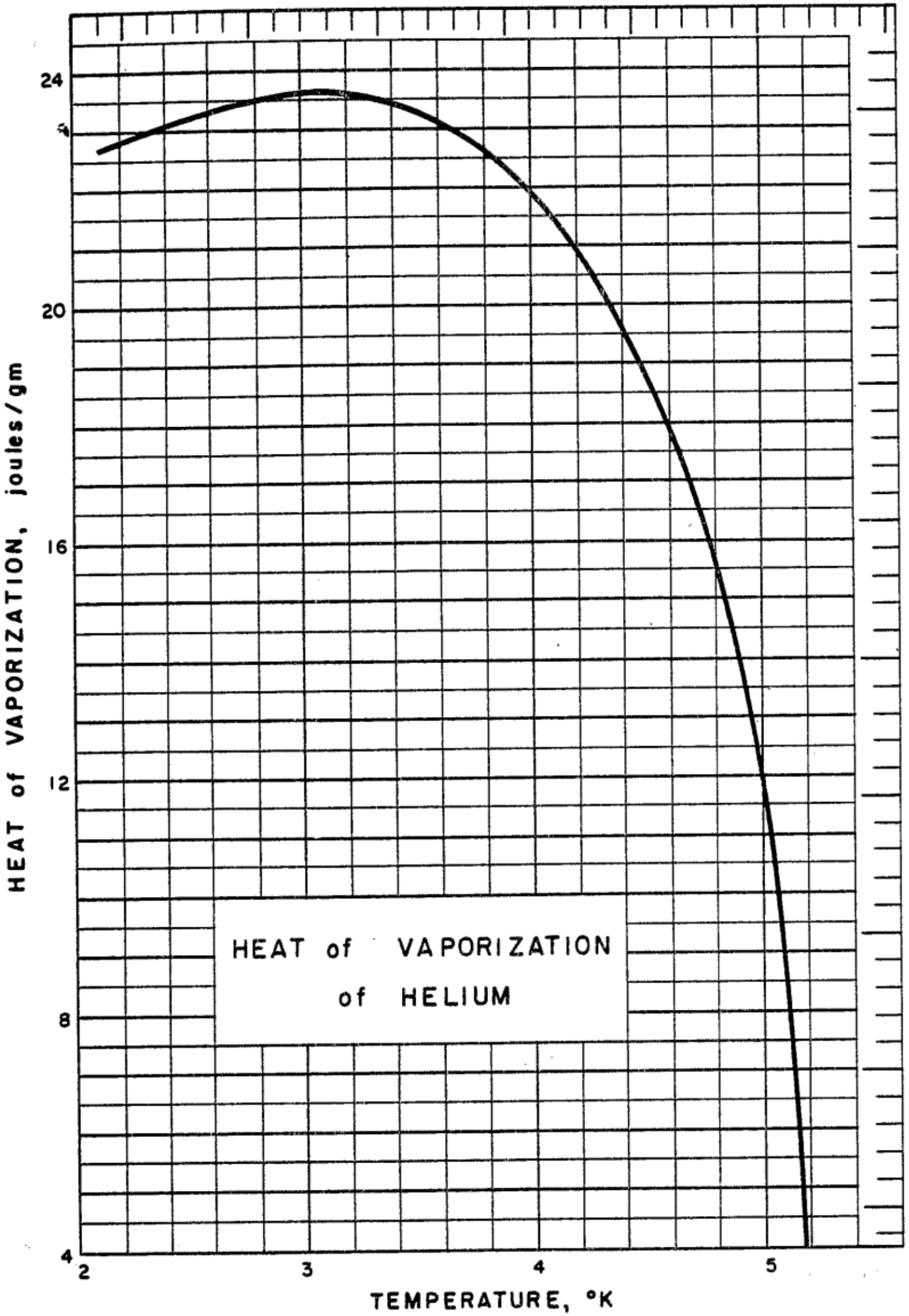


Fig. 10.6. Wiring for a superconducting solenoid to be operated in the persistent mode (see text). The heavy lines indicate the superconducting wire of the magnet and its persistent shunt



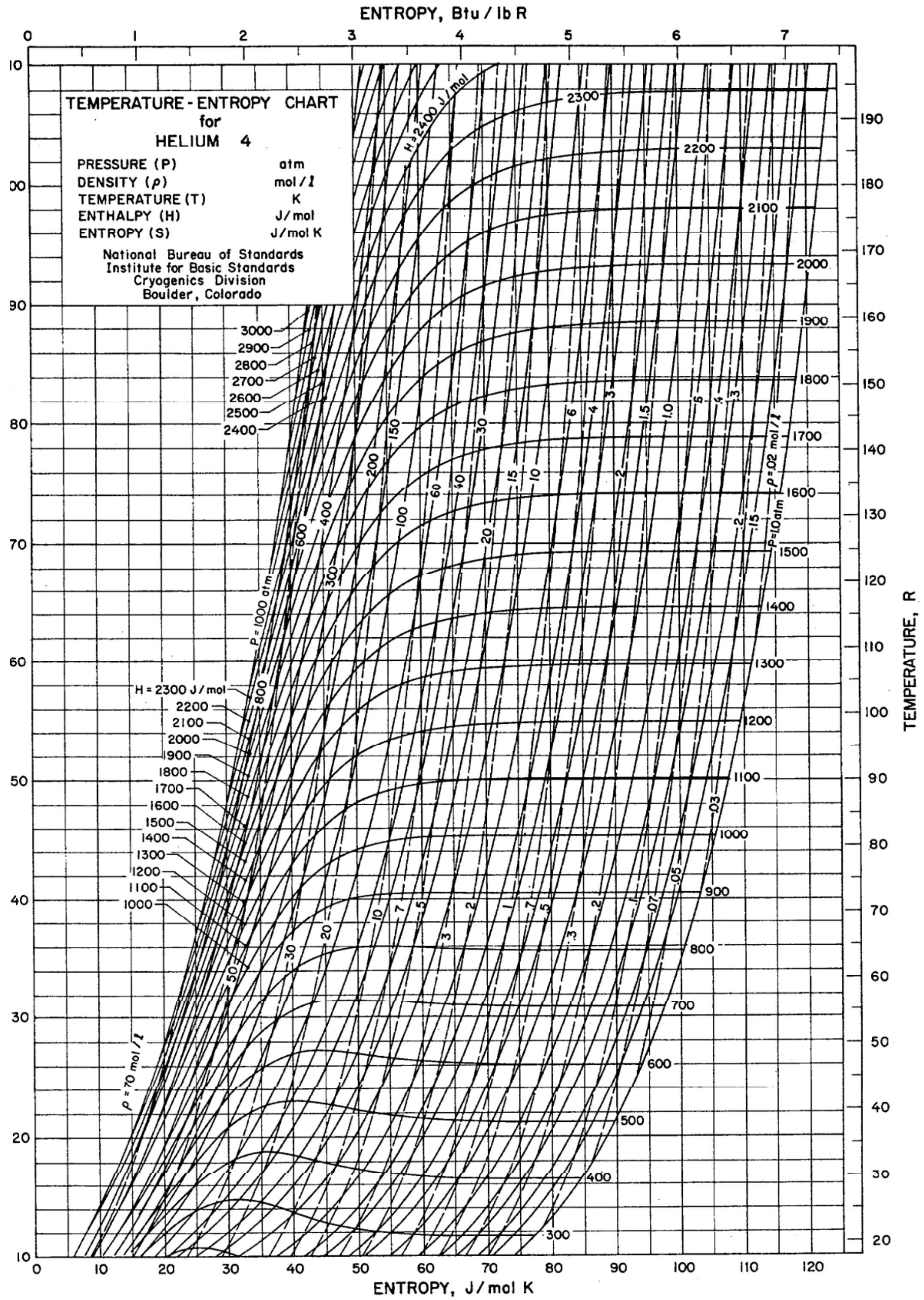


FIGURE 37. Temperature-entropy diagram for helium between 10 and 110 K

Possible material choices

- **Pure metal** for sufficient thermal conductivity
- **Large nuclear moment**, high isotope abundance
- No strong superconductivity or electronic magnetism
- **Spin lattice coupling** has to be sufficient
- Large spin?

$$V = \frac{8\pi}{3}\gamma_e \gamma_n \mathbf{I}\cdot\mathbf{S}\delta(r)$$

$$\frac{1}{T_1} = \frac{64\pi^3}{9}\hbar^3 \gamma_e^2 \gamma_n^2 \left\langle \left| u_k^2(0) \right| \right\rangle_{E_F}^2 \rho(E_F) kT$$

Candidates with ~ 100% abundance:

isotope	μ/μ_n	I	note
^{27}Al	3.64	5/2	SC, B_c = 10 mT, tolerable
^{63,65}Cu	2.3	3/2	
^{113,115}In	5.5	9/2	SC, B_c = 30 mT, challenging
^{51}V	5.14	7/2	SC, B_c = 0.1 T, ruled out
^{93}Nb	8.07	9/2	SC, B_c = 0.2 T, ruled out

Metal & nucleus	Abund. (%)	λ/V Nucl. Curie const./molar volume (μK)	B_c Critical ind. of supercon-ductor (Vs/m^2)	K Korringa's constant (sK)	V Molar volume (cm^3)	τ_2 Spin-spin relax. time (μs)	b Local ind. due to di-pole inter-actions (Vs/m^2)	b_Q Local ind. due to qua-drupole inter-actions (Vs/m^2)
Al^{27}	100	0.87	0.01	1.8	9.98	~30		
V^{51}	100	1.91	0.13	0.8	8.34			
Cu^{63}	69.1	0.57		1.1	7.11	80	0.0003	
Cu^{65}	30.9							
Nb^{93}	100	1.99	~0.25	0.19	10.9			
In^{113}	4.2	1.11	0.03	0.086	15.7			0.25
In^{115}	95.8							
Sn^{115}	0.35	0.015	0.03	0.030	16.3	~100		
Sn^{117}	7.61							
Sn^{119}	8.58							
+ non magn. isot.								
Pt^{195}	33.8	0.019		0.030	9.1	1000		
+ non magn. isot.								
Tl^{203}	29.5	0.21	0.02	0.006	17.2	30		
Tl^{205}	70.5							

Further requirement

- **Cubic lattice or spin *I* = 1/2**

  otherwise electric field gradient at nuclear positions may produce large **quadrupole splitting** (unless $I = 1/2$), which contributes to b, possibly making it too large

For example, indium has **tetragonal lattice**, whereby the quadrupole moment produces an effective field

$b_q \sim 0.25$ T => demagnetization is limited to this

All in all, copper is the best choice
(aluminum might do in some cases)

Thermodynamics

- High temperature expansions are sufficient in most cases

$$\frac{S}{R(N/N_A)} \cong \ln(2J+1) - \frac{J+1}{6J}x^2$$

$$\frac{C_B}{R(N/N_A)} \cong \frac{J+1}{3J}x^2$$

$$\frac{M}{M_{\text{sat}}} \cong \frac{J+1}{3J}x \qquad x = \frac{\mu B}{k_B T}$$

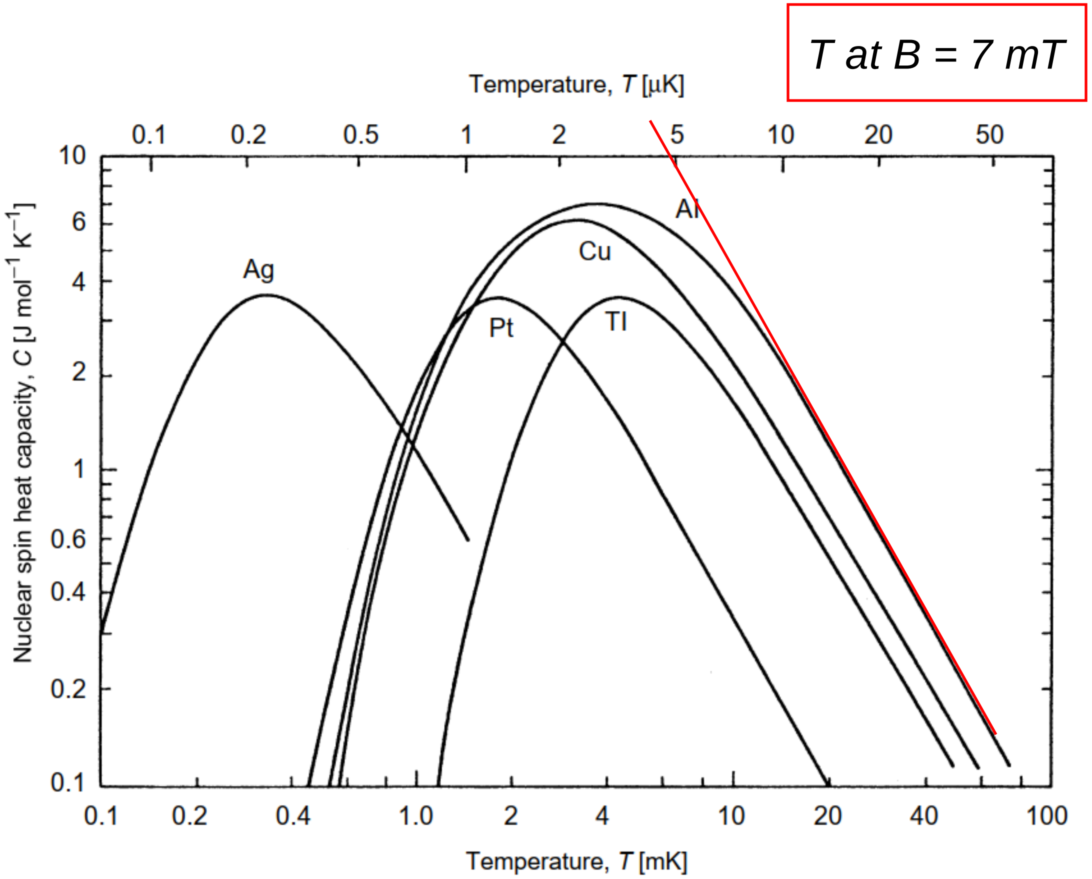
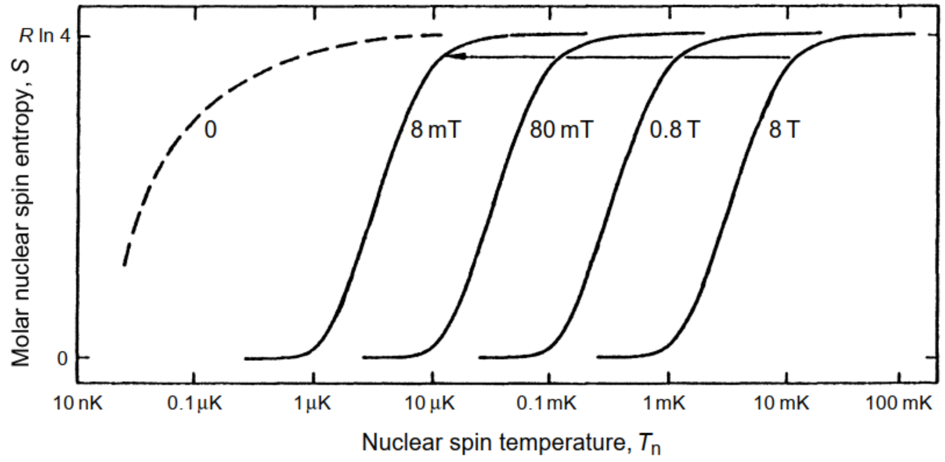


Fig. 10.4. Nuclear heat capacities as a function of temperature for Al ($I = 5/2$), Cu ($I = 3/2$), Tl, Pt and Ag ($I = 1/2$) (however, remember that Pt contains only 33.8% of ^{195}Pt, its only isotope with a nuclear moment). The lower temperature scale corresponds to heat capacity data in a magnetic field of 9 T, the upper temperature scale to data in a field of 7 mT

Cooling cycle



1) **Magnetize** to B_i (1 ... 2 h), usually **as quickly as the magnet permits** (large heat of magnetization, DR copes with it better, $T_1 \sim 50 \ldots 100$ mK)

2) **Precool** to T_i (one to several days) nuclear stage (NS) size + efficiency of DR + quality of heat switch

3) **Thermal isolation** of the nuclear cooling stage (15 min) superconducting heat switch between the DR & NS made SC

4) **Demagnetization** to $B_f \sim 10 \ldots 100$ mT (1/2 ... 1 day) speed limit is set by **losses due to eddy current heating**

5) Thermalize the experiment (hours ... days)

6) **Maintain the low *T* (days ... weeks), small demag/remags**

7) **Connect NS to DR** (heat switch made normal by magnetic field) for the next precool; GOTO 1)

Precooling

Dilution refrigerator absorbs heat $\dot{Q} = aT_{DR}^2 - \dot{q}$ where $a = 84\dot{n}\ \mathrm{J/(mol\,K^2)}$

and $\dot{q}$ is the background heat load

Base temperature without extra load $T_0 \cong \sqrt{\dot{q}/a}$

Thermal resistance of the link between NS and DR (heat switch in normal state) is characterized by the coefficient R ~ 2 ... 20 $\mathrm{K^2/W}$, giving

$$2R\dot{Q} = T_{NS}^2 - T_{DR}$$

The power $\dot{Q}$ originates from heat of magnetization of nuclear spins

$$dQ = -C_B dT_{NS} \qquad C_B \cong \frac{V\lambda}{\mu_0}\left(\frac{B}{T}\right)^2$$

Precooling time to temperature T can be solved from these:

$$t(T) = \left(\frac{1}{a} + 2R\right)\frac{V\lambda B^2}{\mu_0 T_0^2}\left(\frac{1}{2T_0}\ln\frac{T+T_0}{T-T_0} - \frac{1}{T}\right) \approx \left(\frac{1}{a} + 2R\right)\frac{V\lambda B^2}{3\mu_0 T^3}$$

Example

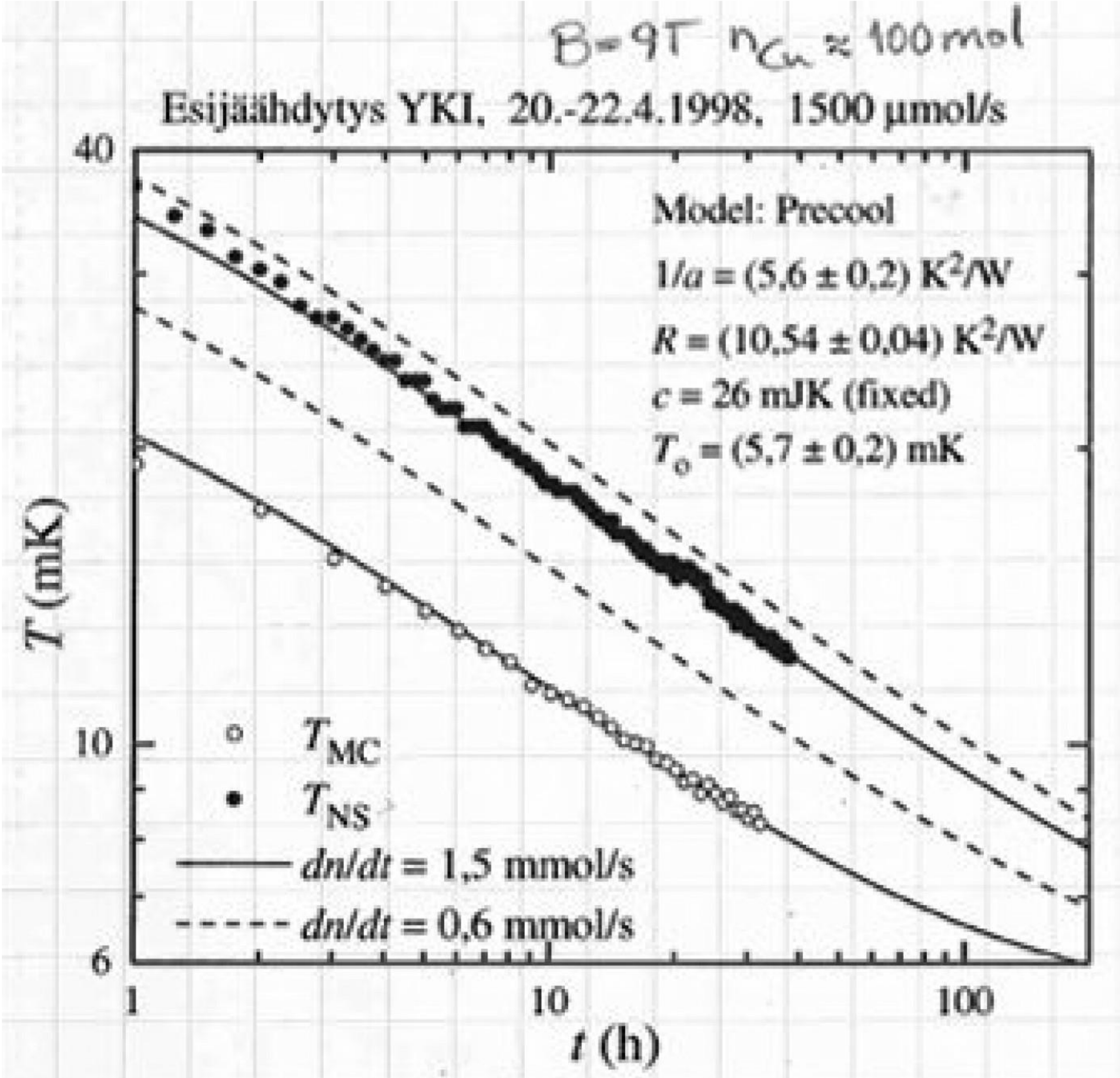
$\dot{n} = 1.0\,\text{mmol/s}\,;\; 1/a = 12\;\text{K}^2/\text{W}$

$R = 12\,\text{K}^2/\text{W}\,;\; T_0 = 3\;\text{mK}$

$n_{NS} = 100\,\text{mol (Cu)}\,;\; B_i = 9\;\text{T}$

$$\frac{V\lambda}{\mu_0}B^2 = 26.5\ \text{mJK}$$

=> t (10 mK) = $3\cdot10^5$ s
~ 3.5 days



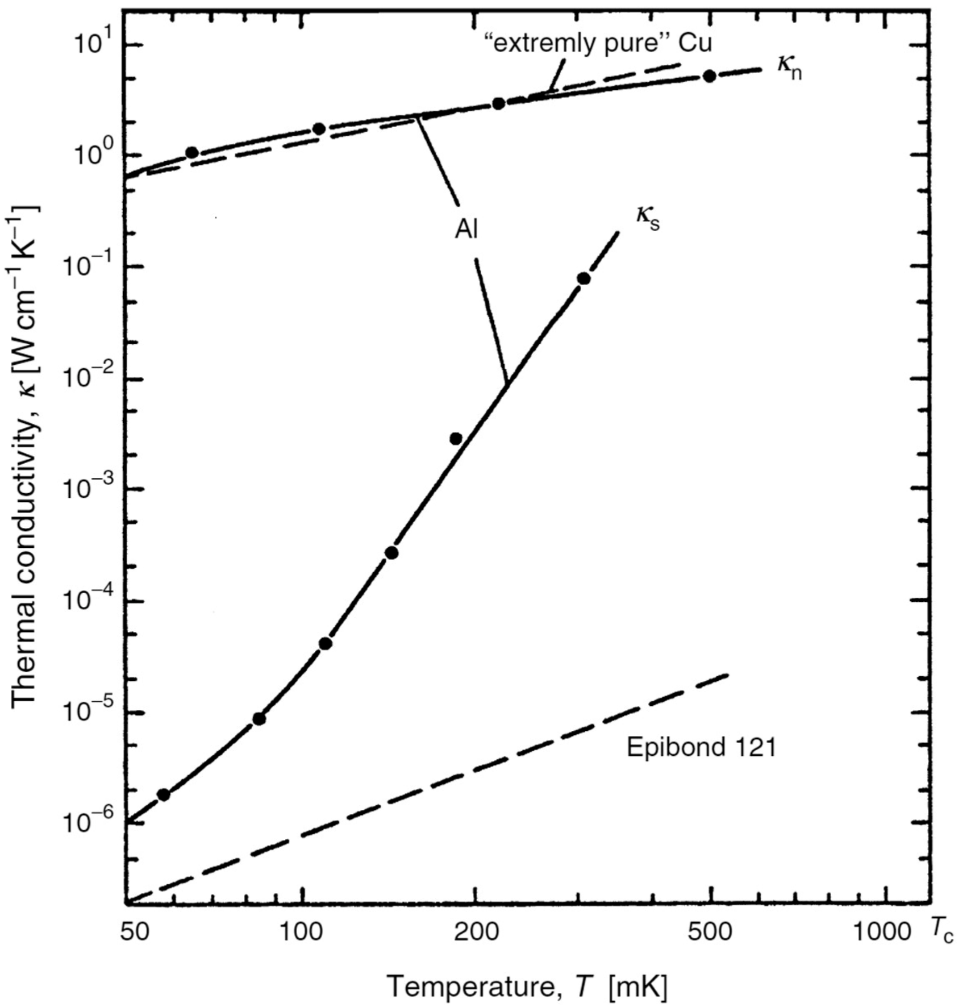
Heat switch

Assume $R \sim 10\ \mathrm{K^2/W}$ in normal state
Good heat switch has **a switching ratio $> 10^5$** $(T = T_0)$
=> Heat leak through superconducting heat switch at T_0:

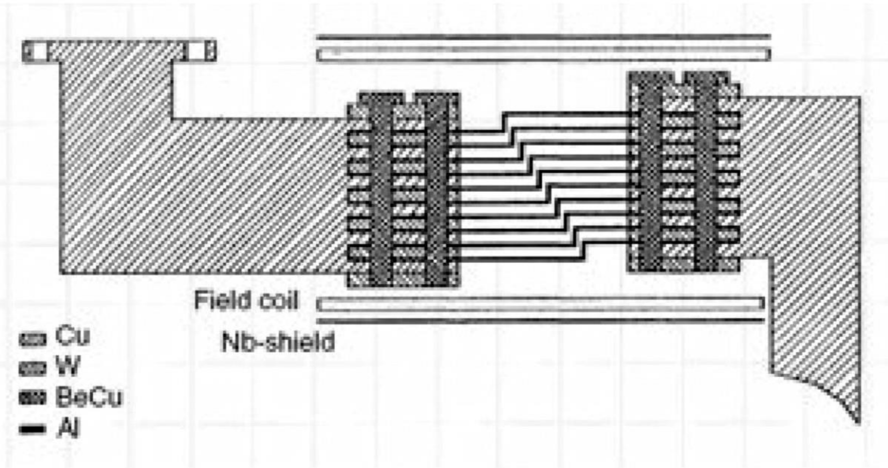
$$\dot{Q} \approx \frac{T_0^2}{2 \cdot 10^5 R} \approx 5\ \mathrm{pW} \quad \text{(perfectly OK)}$$

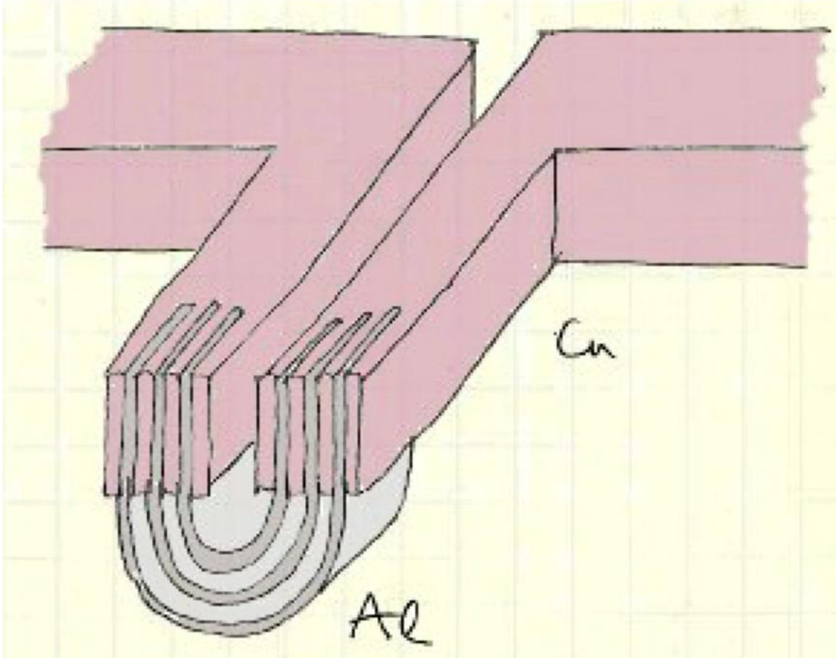
- Good ratio is achieved if the Debye temperature is high
- **Critical magnetic field** should not be too high but the T_c must be an order of magnitude higher than the operating temperature

Al	$\mathbf{T_c}$**=1.2 K**	$\mathbf{B_c}$**=10 mT**	$\mathbf{T_D}$**=394 K**
Zn	0.9 K	5 mT	234 K
Sn	3.7 K	30 mT	~ 200 K



HS designs





- Magnetic field should be oriented parallel to the foils to avoid **flux trapping** in the superconducting state
- Preferably, the field should **not be parallel to the direction of heat flow**
- It may become necessary to replace the coil – be prepared!

Demagnetization

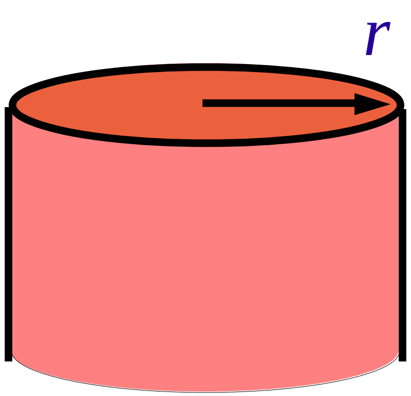
Change of magnetic field must be made as reversibly as possible
Losses may be assumed to result from

- constant background heat load $\dot{q}_{NS}$
- induced eddy current heating $\gamma \dot{B}^2$

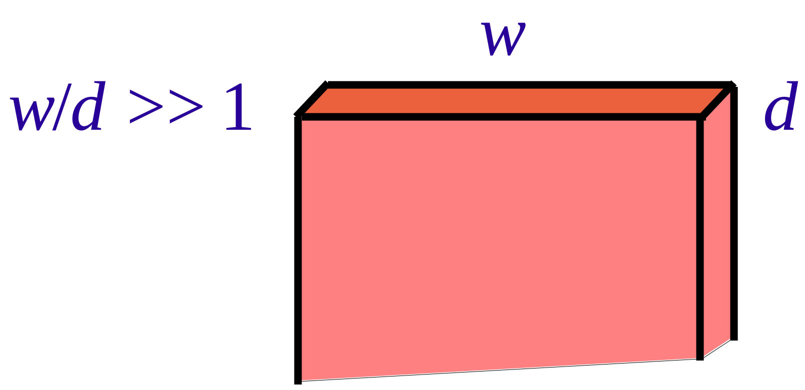
$$\dot{Q}_{NS} = \dot{q}_{NS} + \gamma \dot{B}^2$$

where $\boldsymbol{\gamma = \Gamma V\, RRR/\rho_0}$ depends on el. resistivity and geometry

for a cylinder
$\Gamma = r^2/8$



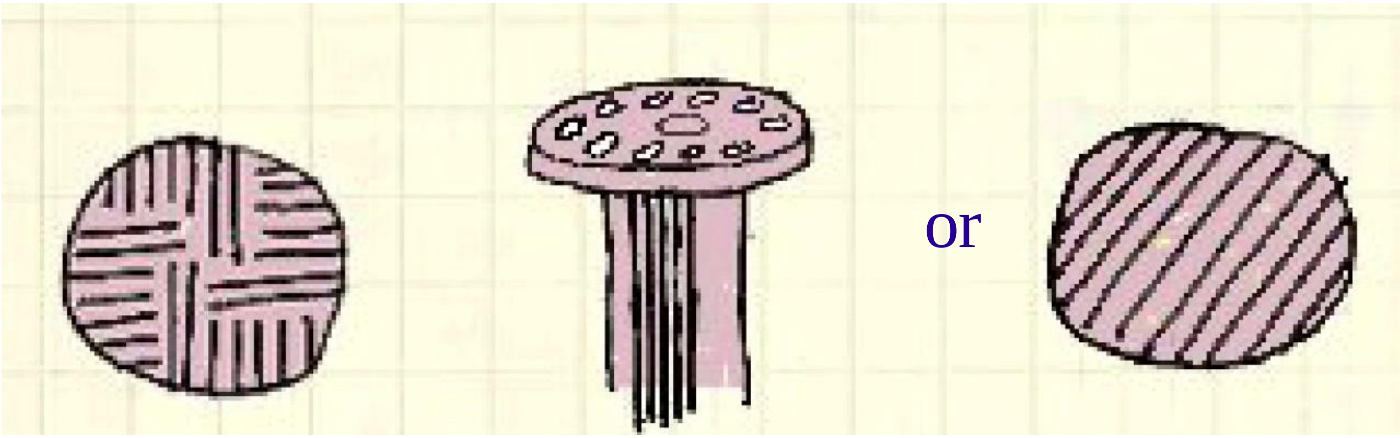
for a slab
$\boldsymbol{\Gamma = d^2/12}$



Eddy currents

To reduce detrimental influence of eddy currents, the nuclear stage must be constructed from **thin filaments or slabs**

$$2r \text{ or } d \sim 1 \text{ mm or less}$$



Filling factor should be good, but the slabs or filaments must be insulated from each other (from the flat side)

Demagnetization profiles

Optimal demagnetization rate is such that the two contributions to **losses are equal**: $\dot{q}_{NS} = \gamma \dot{B}^2$

=> linear demag. profile $\dot{B} = \sqrt{\dot{q}_{NS}/\gamma}$ $t_{\mathrm{opt}} = \sqrt{\dot{q}_{NS}/\gamma}\,(B_i - B_f)$

Resulting nonadiabaticity for linear sweep in time t_{dm}:

$$\Delta\left(\frac{B}{T}\right) = 2\frac{\mu_0}{V\lambda}\ln\left(\frac{B_i}{B_f}\right)\left[\frac{\dot{q}_{NS}}{B_i - B_f}t_{dm} + \frac{\gamma(B_i - B_f)}{t_{dm}}\right] \underset{t_{dm}=t_{opt}}{\longrightarrow} 2\frac{\sqrt{\gamma\,\dot{q}_{NS}}}{V\lambda/\mu_0}\ln\frac{B_i}{B_f}$$

Such a demag is **very SLOW** – several days for usual heat leaks

To save time ($t_{dm} < t_{opt}$), the **best profile is parabolic**:

$$B(t) = a(t/t_{dm})^2 - b(t/t_{dm}) + B_i$$

with
$$\begin{cases} a = B_i + B_f - \sqrt{4B_i B_f + (\dot{q}_{NS}/\gamma)t_{dm}^2} \\ b = 2B_i - \sqrt{4B_i B_f + (\dot{q}_{NS}/\gamma)t_{dm}^2} \end{cases}$$

Optimum demagnetization

Losses for best possible demagnetization in time t_{dm} (parabolic):

$$\Delta\left(\frac{B}{T}\right) = \frac{4\mu_0}{V\lambda}\left[\boldsymbol{a}\frac{\gamma}{\boldsymbol{t_{dm}}} + \sqrt{\gamma\,\dot{q}_{NS}}\ \mathrm{arsinh}\left(\sqrt{\frac{\dot{q}_{NS}}{\gamma B_i B_f}}\frac{\boldsymbol{t_{dm}}}{2}\right)\right]$$

For example: $n_{Cu} = 100$ mol, $d = 2$ mm, $w >> d$, $RRR \sim 1000$ } => $\gamma = 10.6\ \mathrm{Ws^2/T^2}$

$\dot{q}_{NS} = 10$ nW, $B_i = 9$T, $T_i = 10$ mK, $B_f = 50$ mT }

demag. time	profile	loss $\Delta(B_f/T_f)/(B_i/T_i)$
$t_{opt} = 3$ days	linear	1.2 % (too slow)
10 h	linear	5 % (reasonable)
10 h	parabolic	3 % (quite fine)

Good demag. protocol

Instead of spending three days for a demag, it is a good idea **to continue the precool for two more days** and then make a quicker demag during one day or so (say, in 10 h)

Optimum parabolic sweep 9 T $\rightarrow$ 50 mT in 10 h
would begin **at rate ~ 27 mT/min and end at ~ 3 mT/min**

One might also argue that the background heat load is larger at high fields (**vibrations $\rightarrow$ eddy currents**) and that the magnetic field sweep causes less harm in high fields because of **magnetoresistance effects**

=> **Start the sweep at somewhat higher rate and slow down towards the end**

Thermalization process

Nuclear demag **cools down the nuclear spin ensemble**.
How does this couple to your sample?

Spin system has internal relaxation time $\boldsymbol{\tau_2}$ **~ 0.1 ... 10 ms** (fast)

Energy exchange between nuclear spins and conduction electrons is characterized by spin-lattice relaxation time τ_1

metals obey the **Korringa law:** $\boldsymbol{\tau_1 = \kappa / T_e}$

typically $\boldsymbol{\kappa}$ **~ 0.01 ... 10 sK**

when T_e ~ 50 μK => τ_1 ~ 200 s ... 200 ks >> τ_2

THEREFORE $T_e \neq T_n$, conduction electrons may be much HOTTER than the nuclear spins

NMR equations

Thermal equilibrium can be analyzed in terms of NMR relations

Magnetization relaxes as: $$\frac{dM}{dt} = -\frac{1}{\tau_1}\left(M - M_0\right)$$

with $$M \cong -\frac{I+1}{3I}\frac{\mu B}{k_B T_n} M_{\mathrm{sat}} \propto \frac{1}{T_n}$$

towards equilibrium $$M_0 \cong -\frac{I+1}{3I}\frac{\mu B}{k_B T_e} M_{\mathrm{sat}} \propto \frac{1}{T_e}$$

=> $$\frac{d}{dt}\left(\frac{1}{T_n}\right) = -\frac{1}{\tau_1}\left(\frac{1}{T_n} - \frac{1}{T_e}\right)$$

Since $\frac{d}{dt}\left(\frac{1}{T}\right) = -\frac{1}{T^2}\frac{dT}{dt}$ and $\tau_1 = \kappa / T_e$

=> $$\frac{dT_n}{dt} = (T_e - T_n)\frac{T_n}{\kappa}$$

Warm up relations

Heat loads typically burden the conduction electrons, which eventually dump the heat to the nuclear spin system

Nuclei warm up as $$\frac{dT_n}{dt} = \frac{\dot{Q}}{C_B} = \frac{\mu_0 T_n^2}{V\lambda B_f^2}\dot{Q}$$ $$\frac{dT_n}{dt} = (T_e - T_n)\frac{T_n}{\kappa}$$

=> $$\frac{T_e}{T_n} = 1 + \frac{\mu_0 \kappa \dot{Q}}{V\lambda B_f^2} = 1 + \frac{\dot{Q}}{\dot{Q}_n}$$

where $$\dot{Q}_n = \frac{V\lambda B_f^2}{\mu_0 \kappa}$$

is the "**characteristic load**", depending on the magnetic field B_f

– $T_e \sim T_n$ if $\dot{Q} \ll \dot{Q}_n$

Characteristic load

For example $\begin{cases} B_f = 50\ \text{mT} \\ \boldsymbol{n_{\mathrm{Cu}} = 100\ \mathrm{mol}} \\ \kappa_{\mathrm{Cu}} = 1.2\ \text{sK} \end{cases}$ => $\dot{Q}_n = \dfrac{V\lambda B_f^2}{\mu_0 \kappa} \approx 0.7\,\mu\text{W}$

For constant $\dot{Q}$ T_n^{-1} decreases linearly:

$$\frac{dT_n^{-1}}{dt} = -\frac{\mu_0 \dot{Q}}{V\lambda B_f^2} = -\frac{\dot{Q}}{\kappa \dot{Q}_n} \qquad \frac{dT_n}{dt} = \frac{\dot{Q}}{C_B} = \frac{\mu_0 T_n^2}{V\lambda B_f^2}\dot{Q}$$

and so does T_e^{-1}

$$\frac{dT_e^{-1}}{dt} = \frac{\dot{Q}_n}{\dot{Q}+\dot{Q}_n}\frac{dT_n^{-1}}{dt} = \frac{\dot{Q}}{\kappa(\dot{Q}+\dot{Q}_n)} \qquad \frac{T_e}{T_n} = 1 + \frac{\mu_0 \kappa \dot{Q}}{V\lambda B_f^2} = 1 + \frac{\dot{Q}}{\dot{Q}_n}$$

Warm up curves

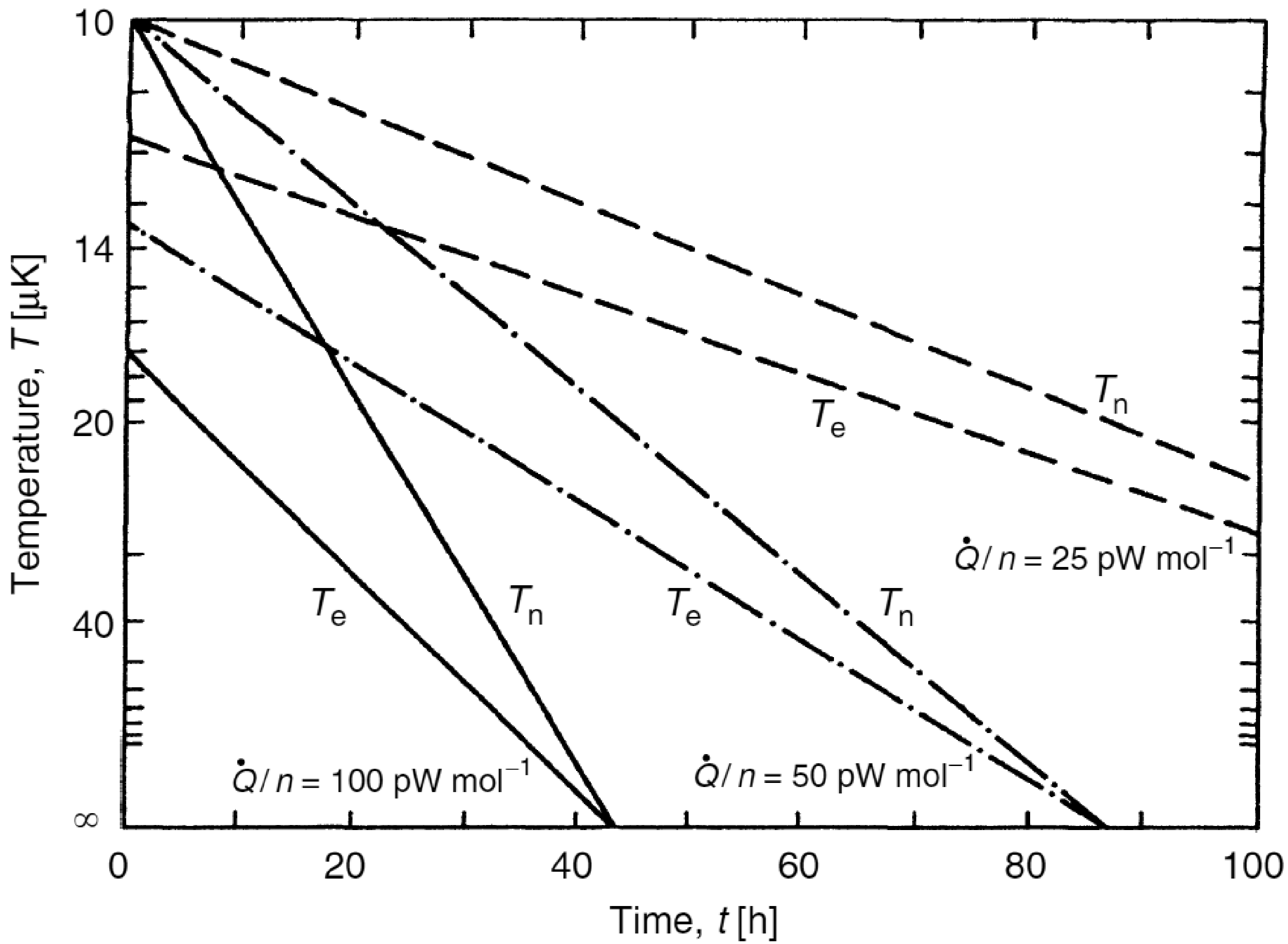


Fig. 10.8. Temperatures, see (10.21), of the nuclear and electronic systems of Cu after demagnetization. The data show the warm-up rates, see (10.26), for the given molar heat leaks $\dot{Q}/n$ to the Cu refrigerant in a final field $B_f = 7\,\mathrm{mT}$ [10.10]

Ph 81

UDC 621.59:536.711
621.576

ACTA POLYTECHNICA SCANDINAVICA

PHYSICS INCLUDING NUCLEONICS SERIES No. 81

Construction and operation of a nuclear refrigeration cryostat

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THESIS FOR THE DEGREE OF DOCTOR OF TECHNOLOGY ACCEPTED BY THE HELSINKI UNIVERSITY OF TECHNOLOGY (OTANIEMI).

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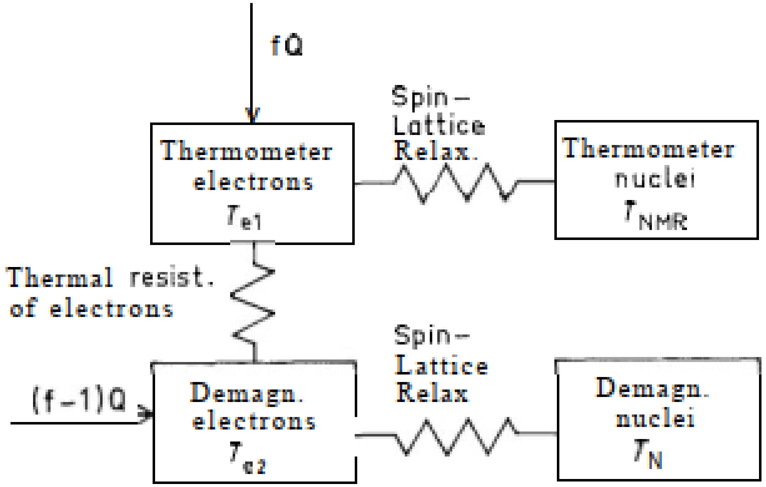
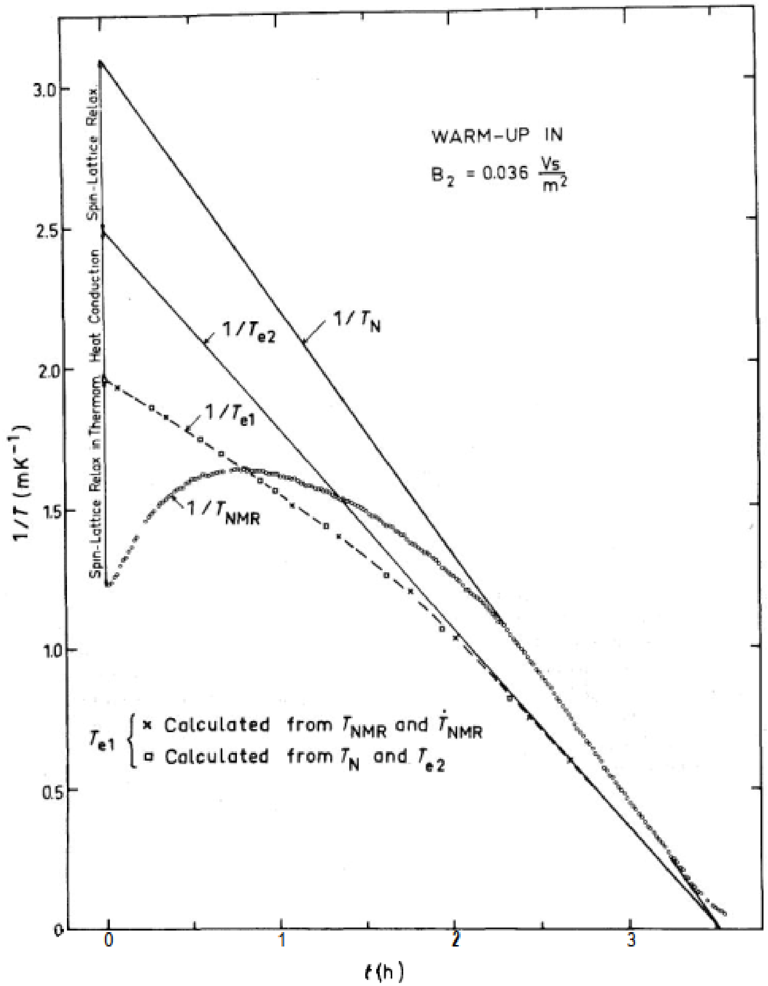




Lowest possible T_e

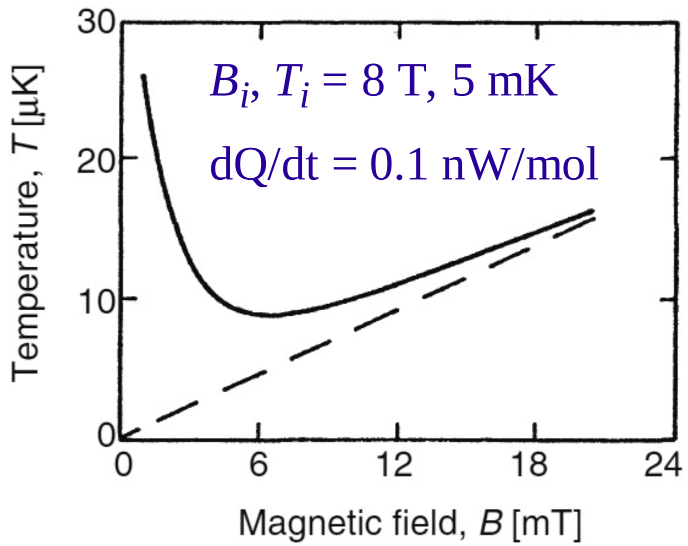
Conduction electron temperature cannot be made arbitrarily low

$$\begin{cases} \dfrac{T_{e,f}}{T_{n,f}} = 1 + \dfrac{\mu_0 \kappa \dot{Q}}{V \lambda B_f^2} \\ T_{n,f} = T_i \dfrac{B_f}{B_i} \end{cases} \quad \Rightarrow \quad T_{e,f} = \frac{T_i}{B_i}\left(B_f + \frac{\mu_0 \kappa \dot{Q}}{V \lambda B_f} \right)$$

This is at minimum, when

$$B_f = \sqrt{\frac{\mu_0 \kappa \dot{Q}}{V \lambda}} \quad \text{i.e. } \dot{Q}_n = \dot{Q}$$

Here $\boldsymbol{T_{e,f} = 2\, T_{n,f}}$



This is impractically low field for most purposes
To maintain more heat capacity, field is usually kept larger
(mind also that b = 0.34 mT for Cu)

Heat leaks

Success or failure of nuclear demag is determined by heat leaks ...

- **thermal conduction** (support, electric leads, heat switch, ...)
  - thermal anchoring is crucial
- **thermal radiation** (radiation shield at $T < 1$ K)
- **remnant gas** in vacuum space ($p_{\mathrm{He}} < 10^{-10}$ Pa)
- **mechanical motion** (pumps, boiling fluids, people, traffic, ...)
  - big mass (several tons)
  - flexible support (air springs, soft tubing, rubber fittings, ...)
- **radioactivity & cosmic radiation**
- electric and magnetic **interference**
  - passive & active shielding, filtering
- internal time-dependent heat leaks (e.g. **H_2 ortho-para conversion**)

Cryostat support

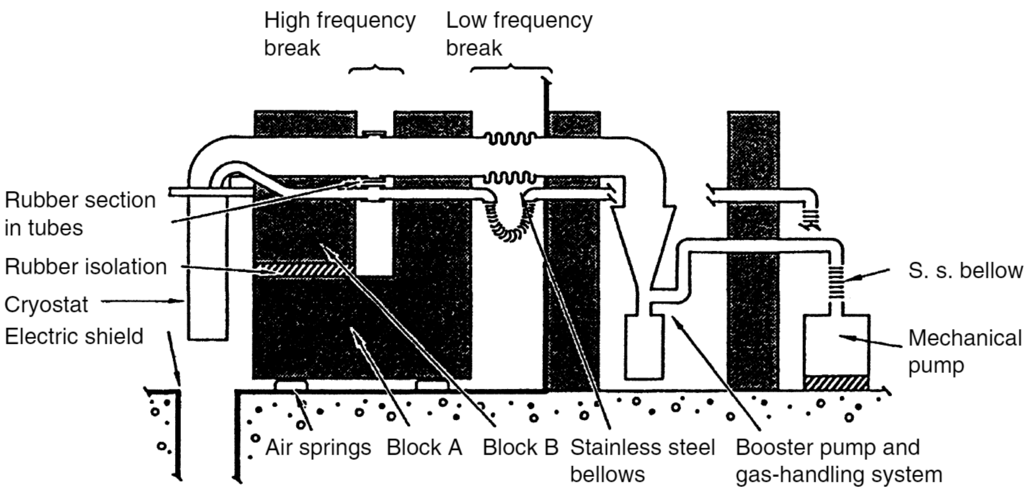


Fig. 10.9. Schematic diagram of a support system for a nuclear refrigeration cryostat. The cryostat is mounted on a concrete block (block A) supported by air springs. On top of block A are placed two smaller blocks (only one shown, block B) resting on thick pads of rubber which carry the wooden beams supporting the cryostat. The pumping tubes are, firstly concreted, into a massive block on the laboratory floor, to remove the vibrations of the pumps, secondly, taken through metal bellows, thirdly, concreted into the main block A, and finally led via a rubber section to be fixed to the sub-blocks B [10.10, 10.36]

Typically one achieves **10 ... 50 pW/mol**
(best cases 5 ... 10 pW/mol)

Hyperfine enhanced systems

Special class of materials (**van Vleck paramagnets**) has electronic moments with nonmagnetic ground state

– noncubic lattice => **quadrupole effects**

– even spin (J = 1, 2, ...) with **lowest energy state m = 0**

– **magnetic field induces an effective electronic moment, which is seen by the nuclei as larger applied field $B_n = B(1+K)$**

=> enhancement of nuclear magnetism by factor $1+K$

The enhancement coefficient **K may be as large as 10 ... 100**

– for example **$PrNi_5$** has $K = 11.2$ for ^{141}Pr nuclei

– nuclear polarization occurs at higher temperature

– **demagnetization is possible as the moment vanishes at $B = 0$**

Compounds: $PrNi_5$, $PrCu_6$, PrS, $PrPt_5$, ... (& TmX_n, TbX_n, HoX_n, ...)

$PrNi_5$

Most common hyperfine refrigerant

Sufficient initial conditions:

– T_i = 25 mK

– B_i = 6 T

$$\frac{\Delta S}{S_{\max}} \approx 0.7$$

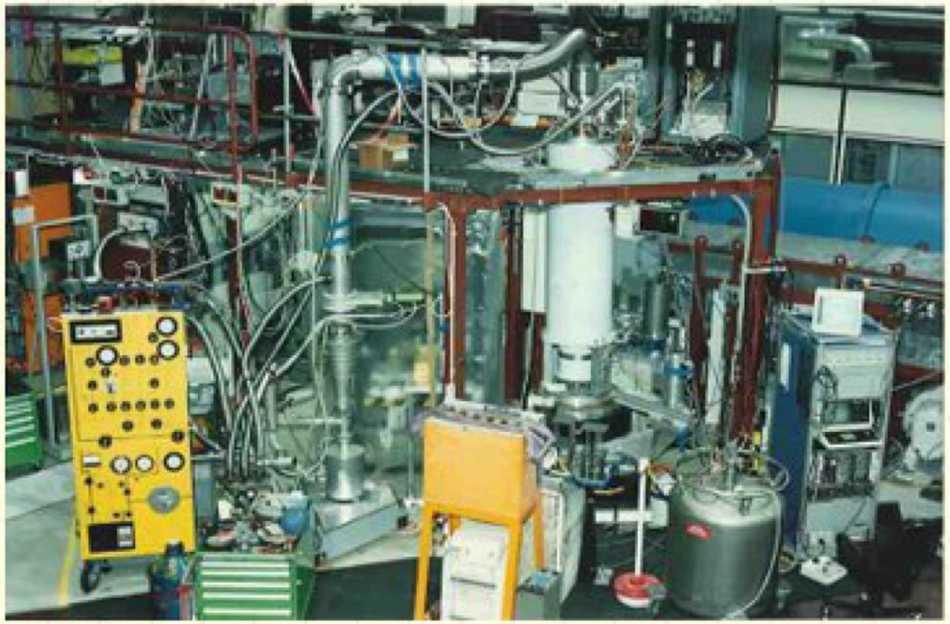
Lowest temperature is limited by $T_c \sim 0.4$ mK

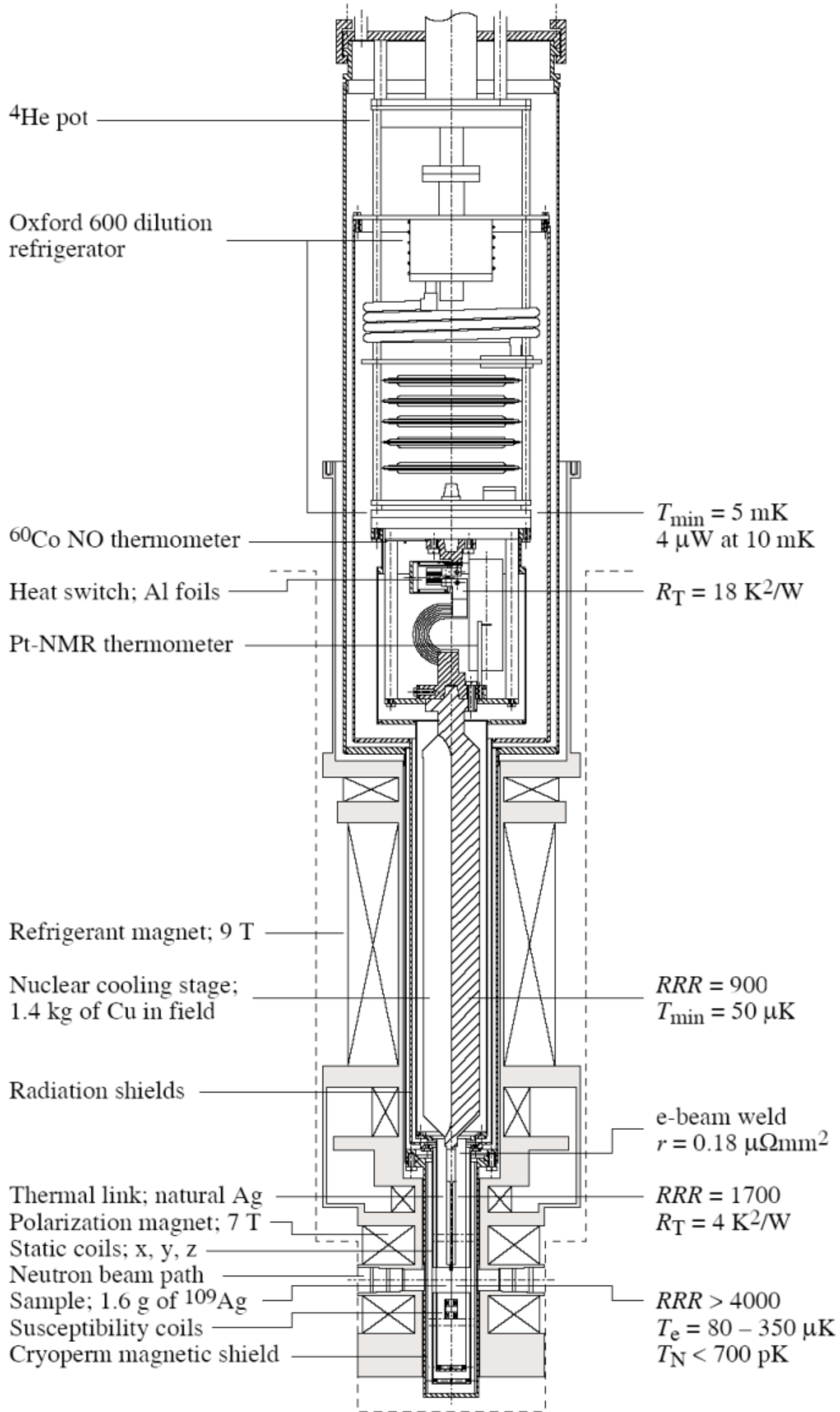
Benefits:	Drawbacks:
moderate initial conditions	material-technical difficulties
high heat capacity	**only modest conductivity**
fast spin-lattice coupling	large internal fields (high T_c)

- Performs well in rough conditions (shaky environment)
- Coming back again with dry dilution refrigerators?

Double nuclear demagnetization

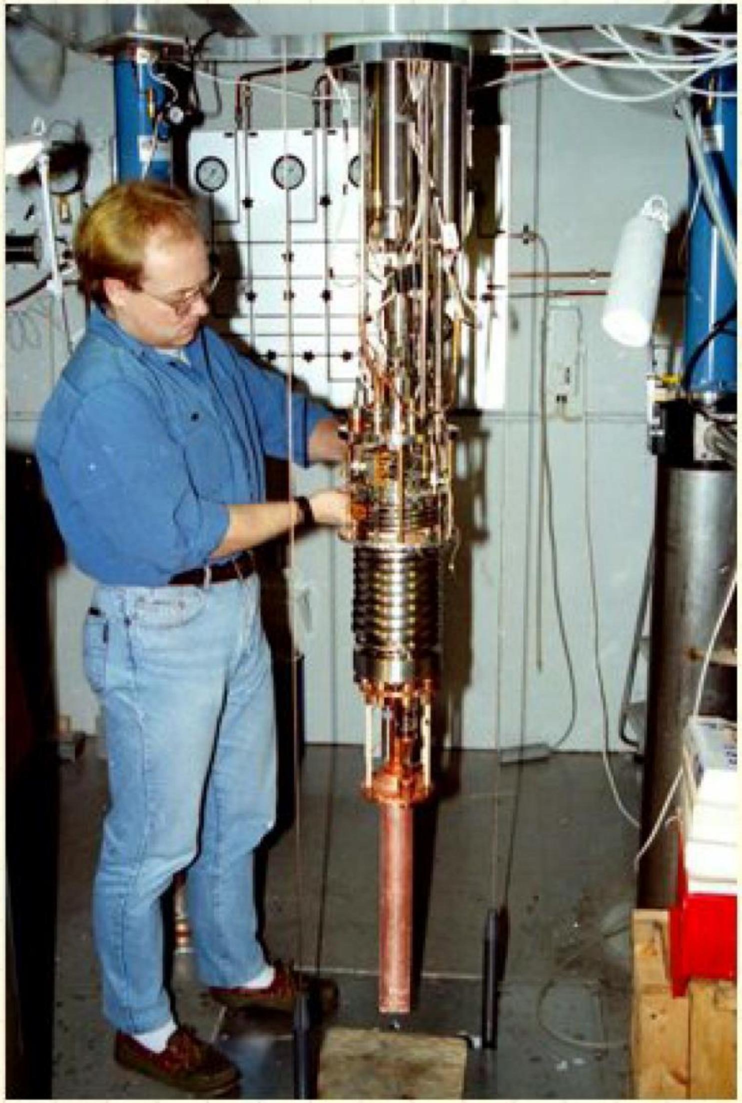
picokelvin installation at HMI in Berlin
(operational from 1992 to 1996)
Neutron diffraction on nuclear
spin ordering in silver

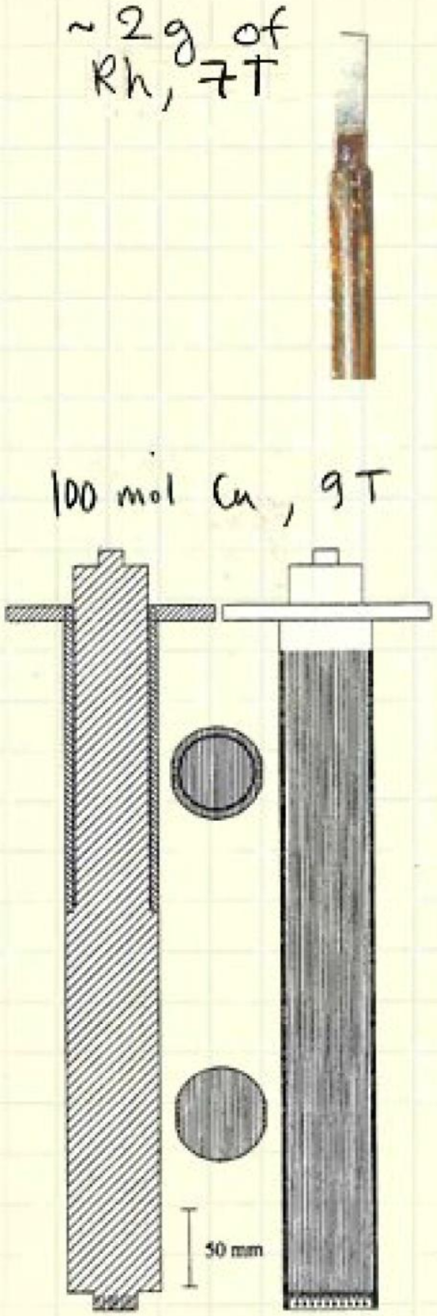


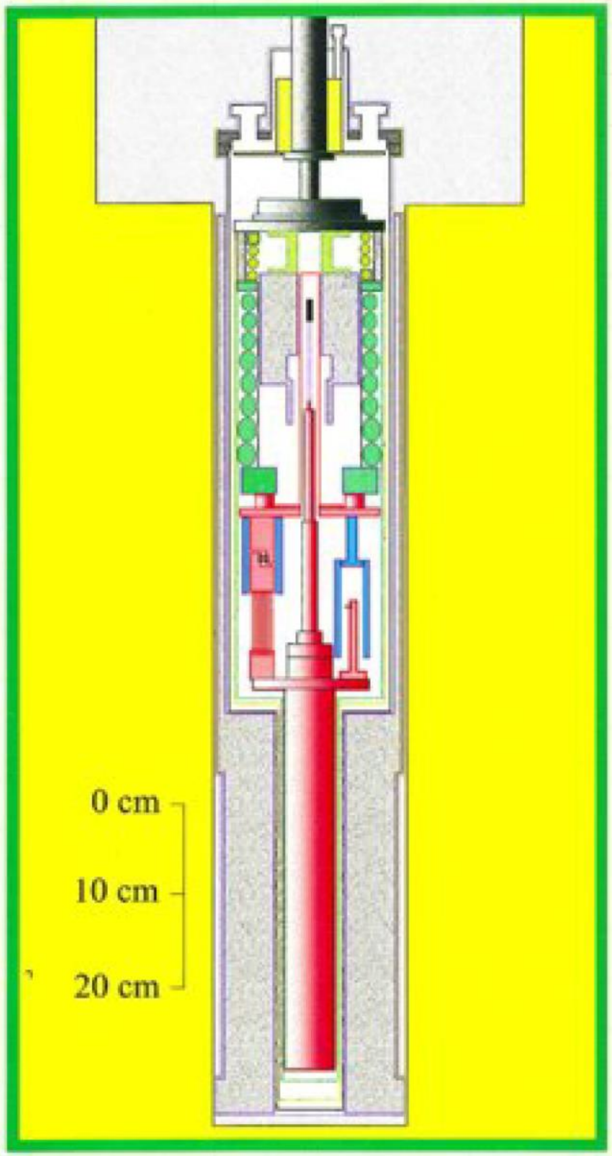


Helsinki cryostat

World record YKI cryostat
LTL, Helsinki: 100 pK in 1999
Nuclear magnetism in Rh, Li,
He mixtures, etc.

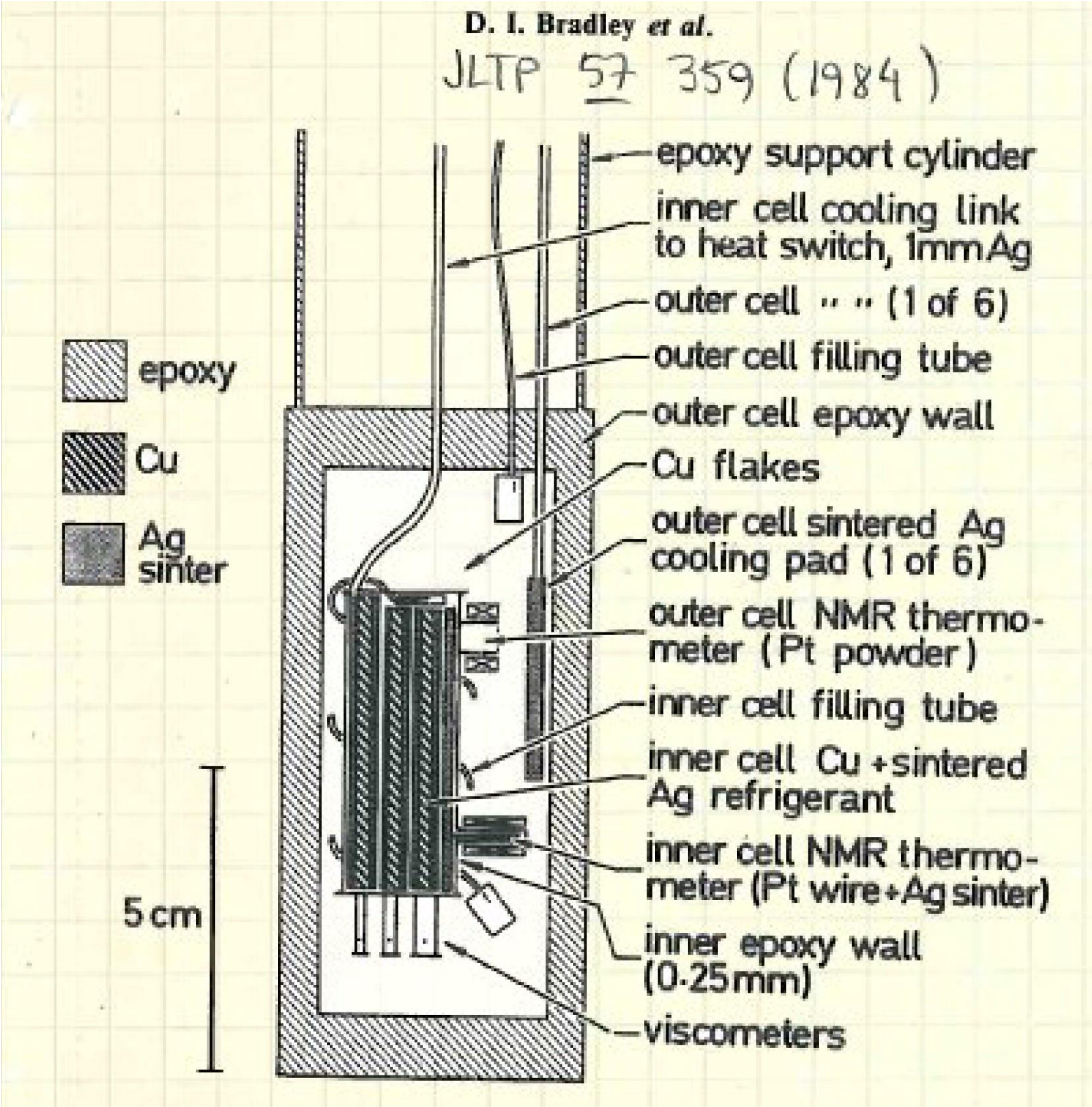






Refrigerating helium

Lancaster design: **NS inside helium** cell to overcome enormous Kapitza resistance between liquid He & metal coolant



Low-temperature records

Record-low temperatures by operating two nuclear demagnetization stages in cascade

Lowest nuclear spin temperatures:

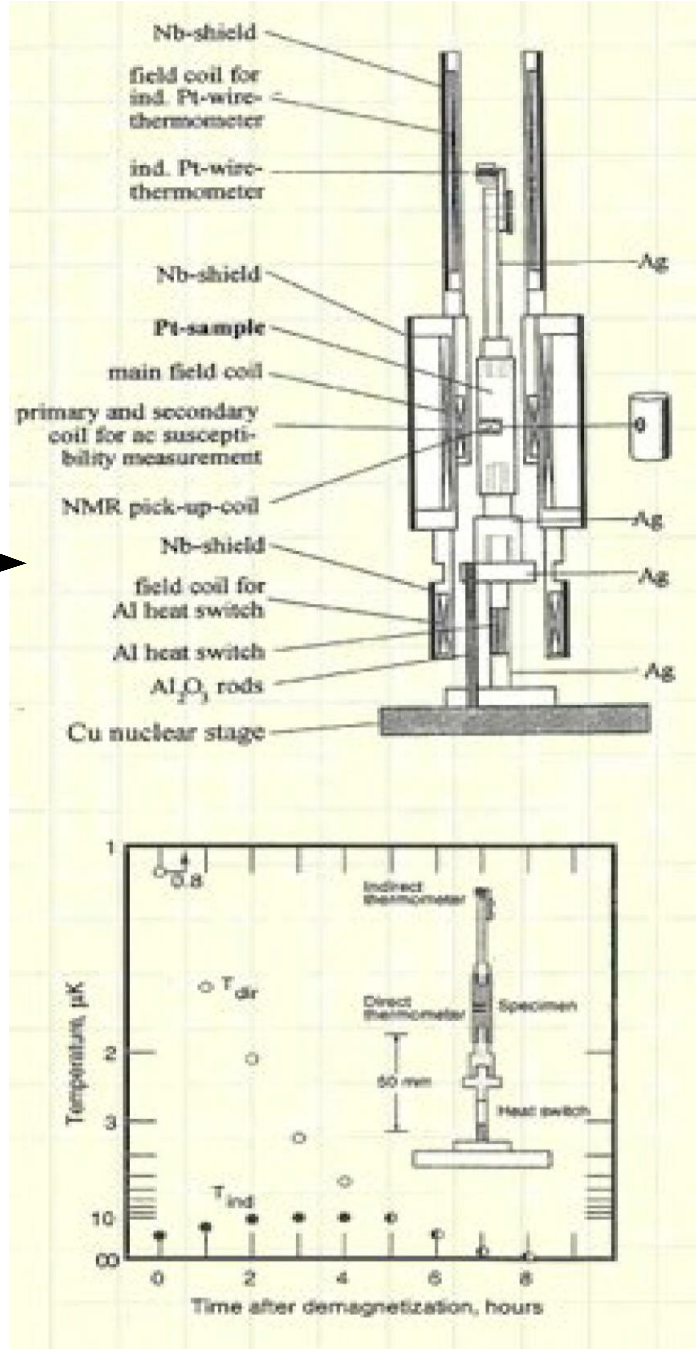
– silver ~ 500 pK (Helsinki 1991)

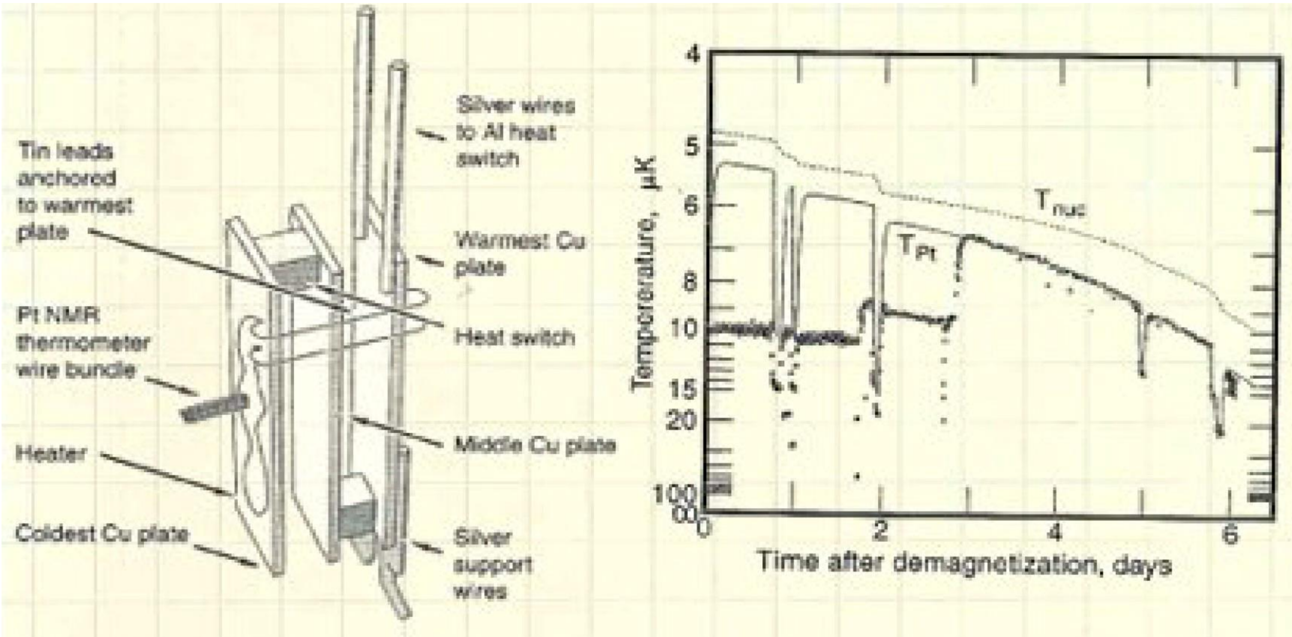
– rhodium ~ 100 pK (Helsinki 1999)

Lowest conduction electron temperatures:

– platinum ~ 2 µK (Bayreuth 1996)

– copper ~ 5 µK (Lancaster 1999)





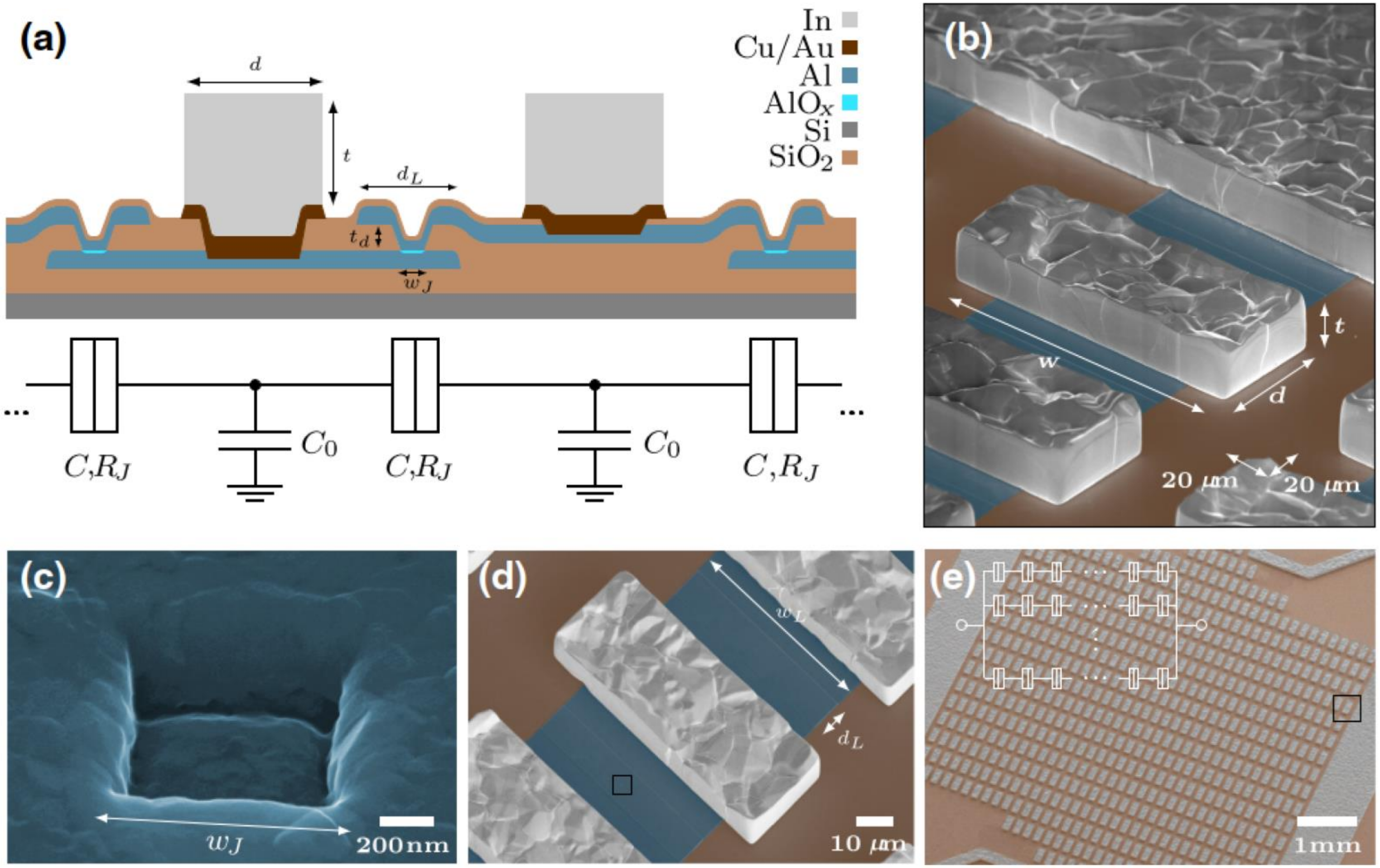


FIG. 1. (a) Cross section of the metallic islands and the Al-AlO_x-Al tunnel junctions with a resistance of R_J. The capacitance C is set by the overlap area $d_L \times w_L$, C_0 is the stray capacitance. The electrons are cooled by the electrodeposited In blocks. (b) False-color scanning electron micrograph of a single In fin. (c) A single tunnel junction between adjacent islands (d). The black square in (d) depicts the area in (c). (e) Overview of the full array with 35 × 15 islands. The black square depicts the area in (b).

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PHYS. REV. APPLIED **12,** 011005 (2019)