Demagnetization cooling
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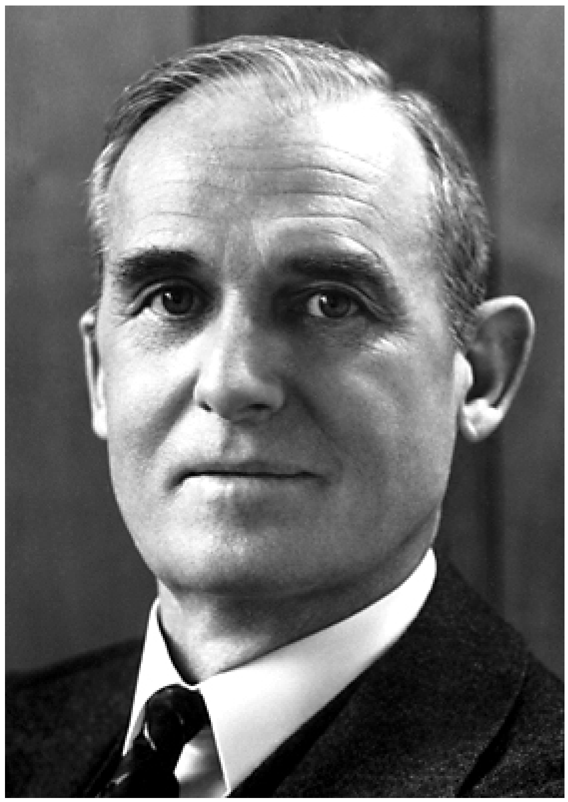
William Francis Giauque (May 12, 1895 – March 28, 1982) was an American chemist and Nobel laureate recognized in 1949 for his studies in the properties of matter at temperatures close to absolute zero. He spent virtually all of his educational and professional career at the University of California, Berkeley.



https://en.wikipedia.org/wiki/William_Giauque

The lowest temperature obtainable with liquid ^{4}He is about 0.7 K, 1 K being the limit for most cryostats. In order to reach appreciably lower temperatures one has to look for some quite different physical system. The requirements for such a system are that its entropy at the starting temperature (1 K) should still be high and that it should depend on an easily manageable parameter, other than the temperature. In 1926, Giaque and Debye independently suggested that certain paramagnetic salts would fulfill these conditions since the electron spins are still completely disordered at helium temperatures, but can be aligned to a high degree of order by the application of a magnetic field of a few thousand Oe. The first cooling experiments with the new method were performed in 1933 and since then have become a standard technique.

1. Approach from the microcanonical ensemble (review)

We consider the electron spin system with two energy levels in the presence of an external magnetic field *B* along the z axis. The spin magnetic moment $\boldsymbol{\mu}$ is given by

$$\boldsymbol{\mu} = -\frac{2\boldsymbol{S}}{\hbar}\mu_B = -\mu_B\boldsymbol{\sigma} ,$$

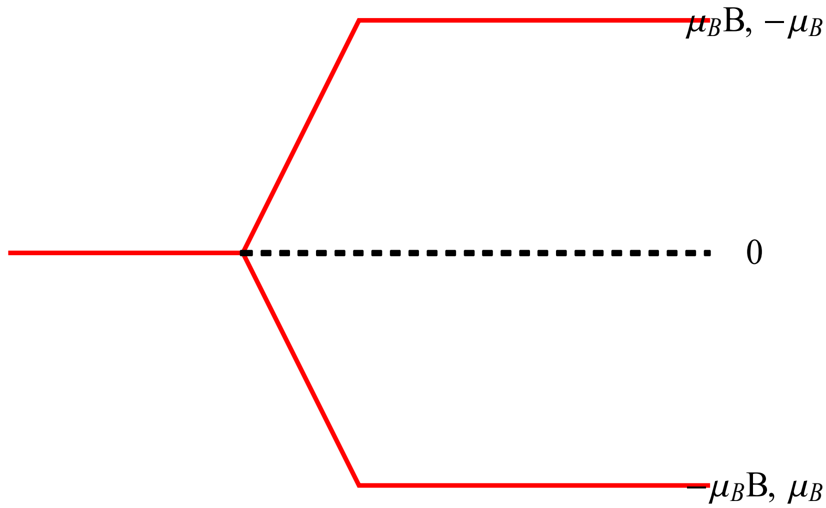
where $\boldsymbol{S}$ $(=\frac{\hbar}{2}\boldsymbol{\sigma})$ is the spin angular momentum, $\mu_B = \frac{e\hbar}{2mc}$ (>) is the Bohr magneton, and the charge of electron is $-e$ ($e>0$). In the presence of the magnetic field along the z axis, we have a Zeeman energy given by

$$\varepsilon = -\boldsymbol{\mu}\cdot\boldsymbol{B} = -(-\mu_B\boldsymbol{\sigma})\cdot\boldsymbol{B} = \mu_B B\,\sigma_z$$

Noting that $\sigma_z|+z\rangle = |+z\rangle$ and $\sigma_z|-z\rangle = -|-z\rangle$ in quantum mechanics, the energy level splits into two levels, $\pm\mu_B B$.

(a) The energy $\mu_B B$ (higher level),
The spin state $|+z\rangle$. The spin magnetic moment is antiparallel to the z-axis $(-\mu_B)$. $|\downarrow\rangle$ state.

(b) The energy $-\mu_B B$ (lower level).
The spin state: $|-z\rangle$. The spin magnetic moment is parallel to the z-axis $(+\mu_B)$; $|\uparrow\rangle$ state.



2. Entropy *S*

The entropy S is obtained as

$$S = \frac{E - F}{T} = k_B N\{\ln(2\cosh(\beta\mu_B B)] - \beta\mu_B B \tanh(\beta\mu_B B)\}$$

or

$$\frac{S}{k_B N} = \ln[2\cosh(\frac{\mu_B B}{k_B T})] - \frac{\mu_B B}{k_B T}\tanh(\frac{\mu_B B}{k_B T}).$$

The entropy S is expressed by a scaling function of B/T. This is an essential point to this system.
We introduce the characteristic temperature T_0 and magnetic field B_0 as

$$\mu_B B_0 = k_B T_0$$

Then we have

$$\begin{aligned}\frac{S}{k_B N} &= \ln[2\cosh(\frac{\mu_B B}{k_B T})] - \frac{\mu_B B}{k_B T}\tanh(\frac{\mu_B B}{k_B T}) \\ &= \ln[2\cosh(\frac{b}{t})] - \frac{b}{t}\tanh(\frac{b}{t})\end{aligned}$$

or simplicity we use the form

$$f(\frac{t}{b}) = \ln[2\cosh(\frac{\mu_B B}{k_B T})] - \frac{\mu_B B}{k_B T}\tanh(\frac{\mu_B B}{k_B T})$$

$$= \ln[2\cosh(\frac{b}{t})] - \frac{b}{t}\tanh(\frac{b}{t})$$

where

$$b = \frac{B}{B_0}, \qquad t = \frac{T}{T_0}$$

$$\frac{\mu_B B}{k_B T} = \frac{\mu_B B_0 \frac{B}{B_0}}{k_B T_0 \frac{T}{T_0}} = \frac{b}{t}$$

We make a plot of $\frac{S}{k_B N}$ as function of *t*, where *b* is changed as a parameter. In the limit of $t \to \infty$, the entropy reached

$$\frac{S}{k_B N} = \ln(2s+1) = \ln 2 = 0.693147.$$

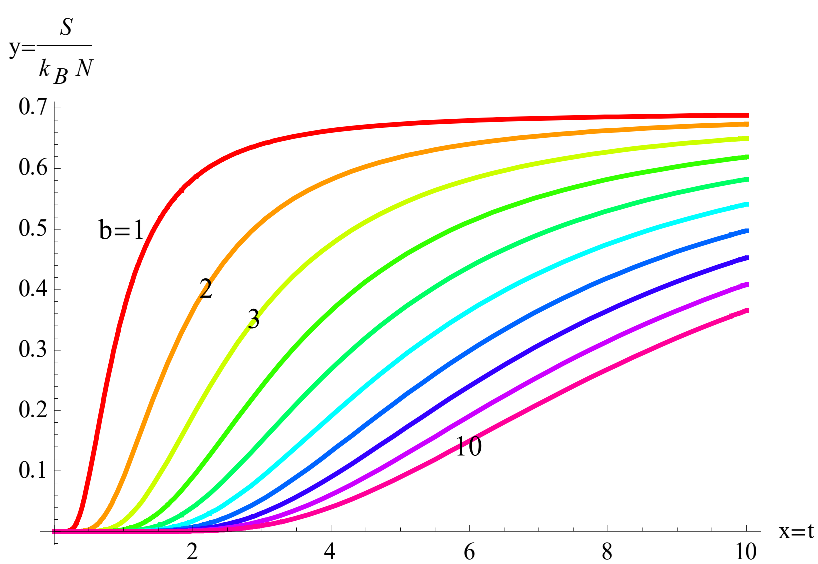


Fig. Plot of $\frac{S}{k_B N}$ as a function of a reduced temperature t (= T/T_0), where the reduced magnetic field b (= B/B_0) is changed as a parameter. Note that $\mu_B B_0 = k_B T_0$. The highest value of y is ln2 = 0.693147.

3. Proof of $\frac{S}{k_B N} \to 0$ in the limit of $t \to 0$

Noting that $e^{b/t} >> 1$ and $e^{-b/t} << 1$, we can approximate the expression of entropy as

$$\begin{aligned}
\frac{S}{k_B N} &= \ln[2\cosh(\frac{b}{t})] - \frac{b}{t}\tanh(\frac{b}{t}) \\
&= \ln(e^{b/t} + e^{-b/t}) - \frac{b}{t}\left(\frac{e^{b/t} - e^{-b/t}}{e^{b/t} + e^{-b/t}}\right) \\
&= \ln[e^{b/t}(1 + e^{-2b/t})] - \frac{b}{t}\left(\frac{1 - e^{-2b/t}}{1 + e^{-2b/t}}\right) \\
&\approx \frac{b}{t} + e^{-2b/t} - \frac{b}{t}\left(1 - 2e^{-2b/t}\right) \\
&\approx \frac{2b}{t} e^{-2b/t}
\end{aligned}$$

where we use the approximation

$$\ln(1+x) \approx x, \qquad \frac{1-x}{1+x} \approx 1 - 2x \qquad \text{for } 0 < x << 1$$

In the limit of $t \to 0$, we have

$$\lim_{t \to 0} S = 0$$

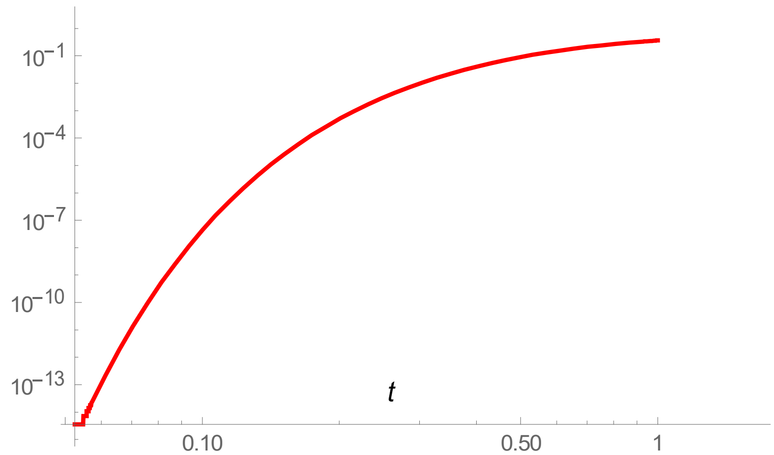


Fig. The entropy of $S/(Nk_B)$ as a function of t. b = 1. The system settles into ground state; the multiplicity becomes 1: and the entropy goes to zero.

4. Isentropic demagnetizion

The principle of magnetically cooling a sample is as follows. The paramagnet is first cooled to a low starting temperature. The magnetic cooling then proceeds via two steps.

Suppose that the spin system is kept at temperature T_1 in the presence of magnetic field B_1. The system is insulated ($\Delta S = 0$) and the field removed, the system follows the constant entropy path AB, ending up at the temperature T_2 (isentropic process). If B_Δ is the effective field that corresponds to the local interactions, the final temperature T_2 reached in an isentropic demagnetization process is

$$\frac{T_2}{B_\Delta} = \frac{T_1}{B_1}.$$

since the entropy is a function of only B/T.

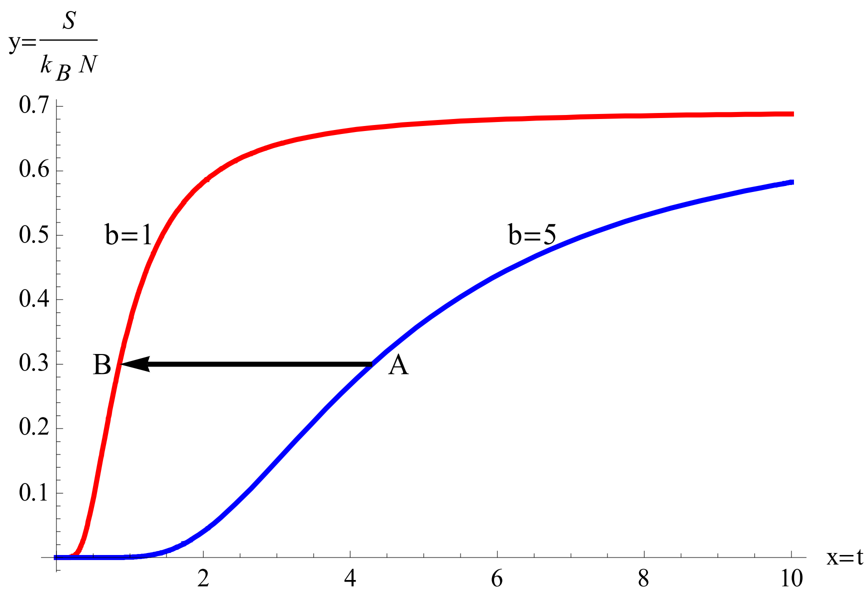
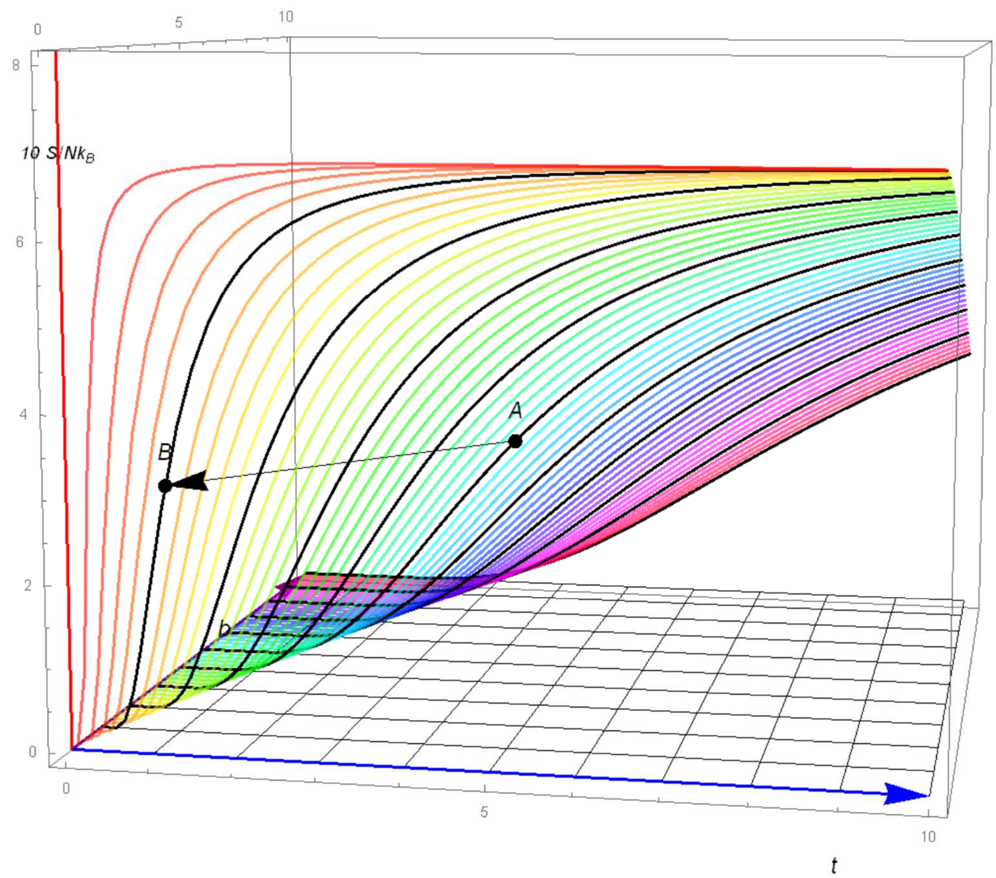
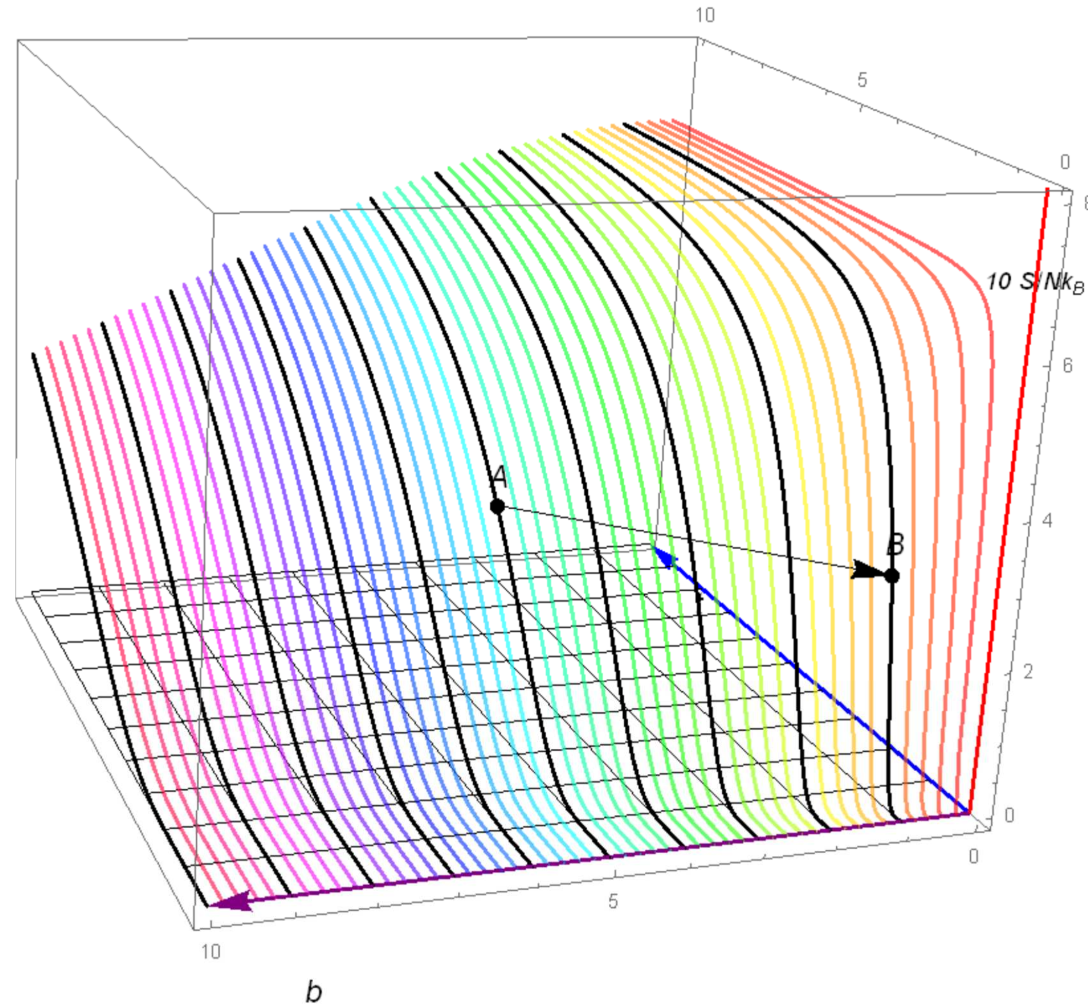


Fig. Point A (t_A = 4.29726, y_A = 0.3) on the line with $\frac{B_A}{B_0} = 1$. Point B (t_B= 0.859452, , y_A = 0.3) on the line with $\frac{B_B}{B_0} = 5$. The path AB is the isentropic process (y = 0.3). Note that $\frac{t_A}{B_A} = \frac{t_B}{B_B}$.



10 S/Nk_B
A
B
b
t



5. Isentropic demagnetization (electron magnetic moment)

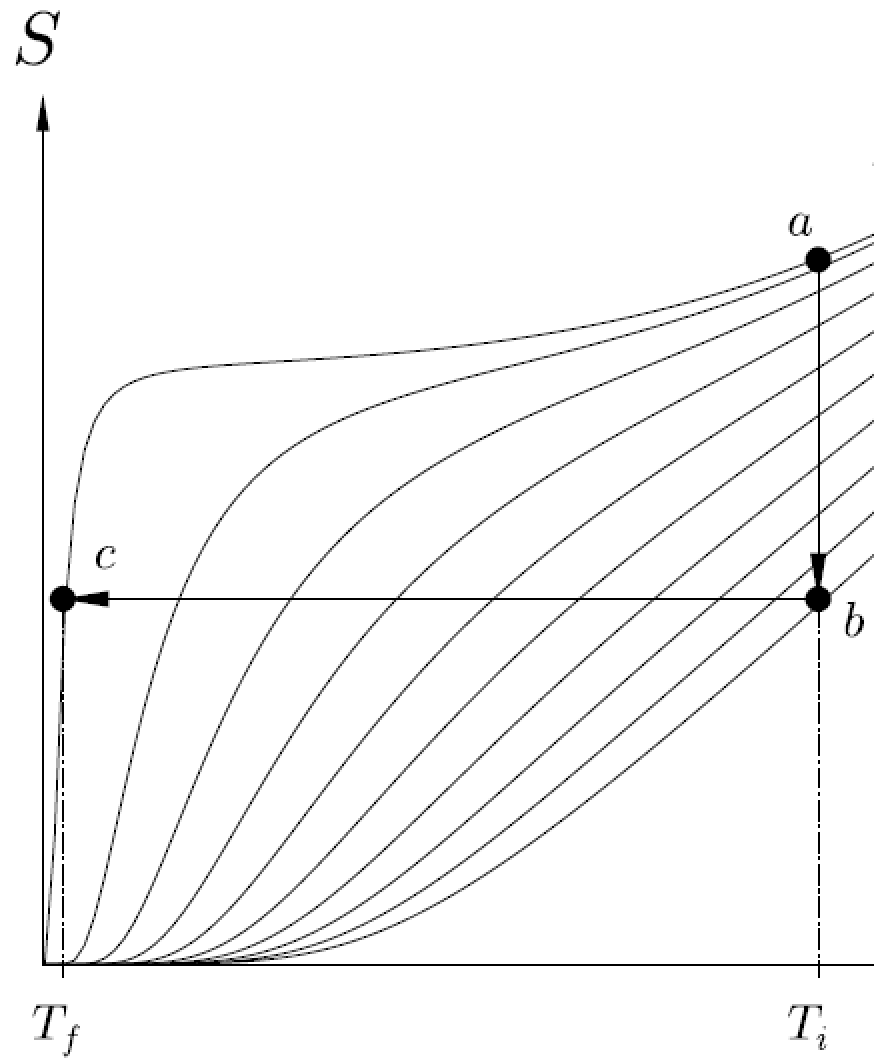


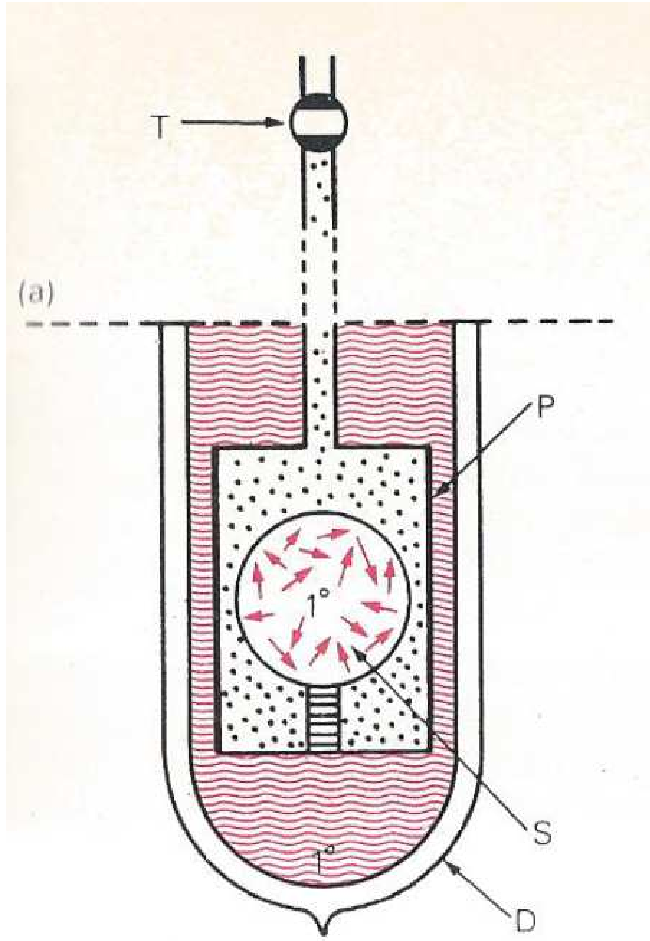
Fig. Entropy vs temperature with the magnetic field being parameters.: $B = B_\Delta$ (at the point *a*) and $B = B_1$ (at the point *b*). $T_i = T_1$. $T_f = T_2$.

The entropy of a paramagnetic salt as a function of temperature for several different applied magnetic fields between zero and some maximum value which we will call B_b. Magnetic cooling of a paramagnetic salt from temperature T_i to T_f is accomplished as indicated in two steps: first, isothermal magnetization from *a* to *b* by increasing the magnetic field from 0 to B_b at constant temperature T_i; second, adiabatic demagnetization from *b* to *c*. The $S(T)$ curves have been calculated assuming spin S = 1/2. A term $\propto T^3$ has been added to these curves to simulate the entropy of the lattice vibrations. The curve for B = 0 is actually for small, but nonzero, B to simulate the effect of a small field. (Blundell and Blundell)

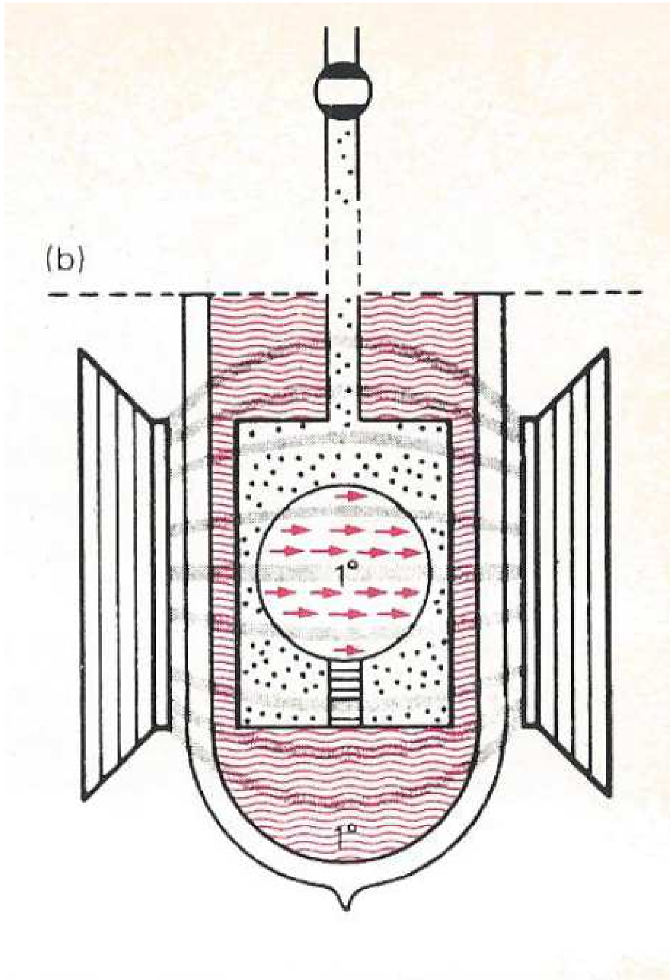
K. Mendelssohn, Cryophysics (Interscience, 1960).

The principle of paramagnetic cooling is illustrated by the idealized entropy-temperature diagram of a typical salt. At 1 K there is a small lattice entropy S_L left, but the main contribution is due to the electron spins, which still obey the Curie's law. Deviation from this law occur only at about 0.1 K or lower where the entropy decreases because of spontaneous alignment of electron spin due to interaction. The curve S_0 represents the entropy vs temperature in zero magnetic field. Application of a strong magnetic field at 1K results in orientation of the spins and this ordering process is equivalent to a decrease in entropy. For a field of 1T (= 10 kOe) will reduce the entropy at 1 K to S_H a fraction of its original value. Magnetization of the spins, liberates heat which has to be conducted away if the process is carried out isothermally. On the other hand, adiabatic demagnetization is an isentropic process and when the magnetic field is removed without permitting the salt will cool along a line of constant entropy. The final temperature T_f reached corresponds to that point in the *S*-*T* diagram at which the entropy in zero field attains the value of S_H at 1 K.

((**Step-1**)) The thermal contact at T_1 is provided by He gas in the presence of the residual magnetic field B_Δ (typically crystal magnetic field)



((**Step-2**)) ((**Isothermal process**)) The field is applied from B_Δ to B_1 at $T = T_1$ with the specimen in good thermal contact with the surrounding giving the isothermal path *ab* (sample is surrounded with He gas)



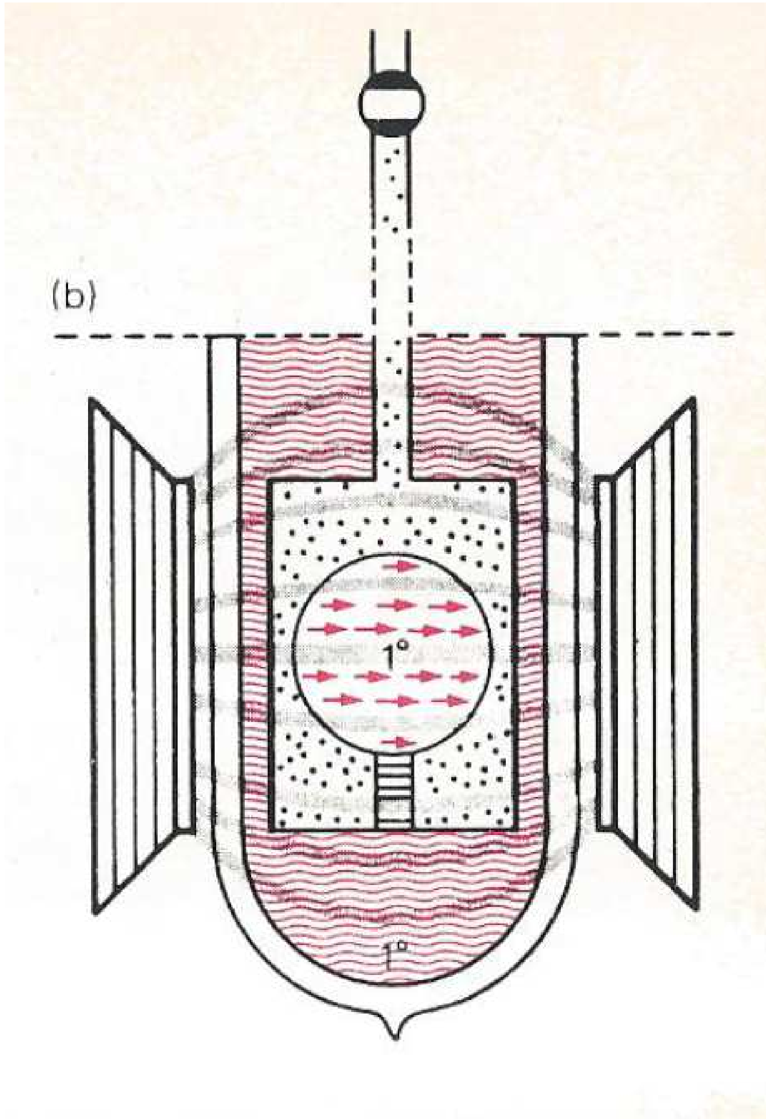
The sample is at the point a (T_1 = 1 K and $B_\Delta = 100\,\mathrm{Oe}$),

$$\frac{\mu_B B_\Delta}{k_B T_1} = 6.71713\times 10^{-3}, \qquad \text{or} \qquad \frac{k_B T_1}{\mu_B B_\Delta} = 148.873$$

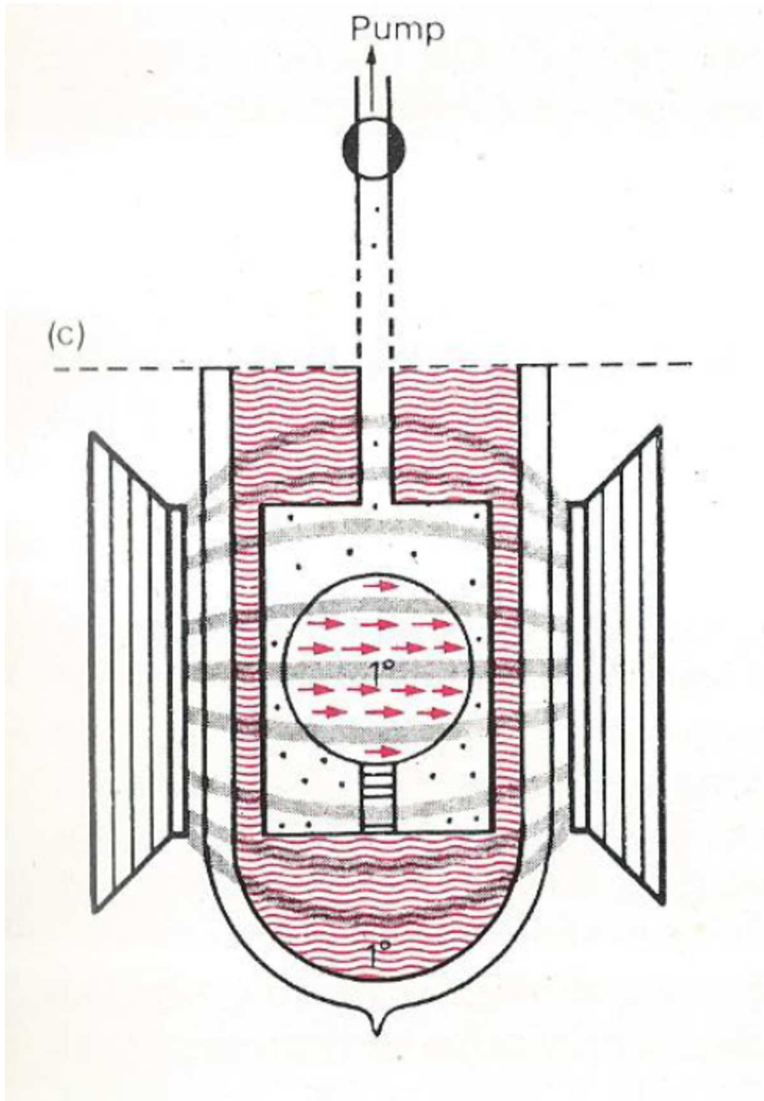
where

$\mu_B = 9.27400915\times 10^{-21}$ emu (-erg/Oe) (Bohr magneton)

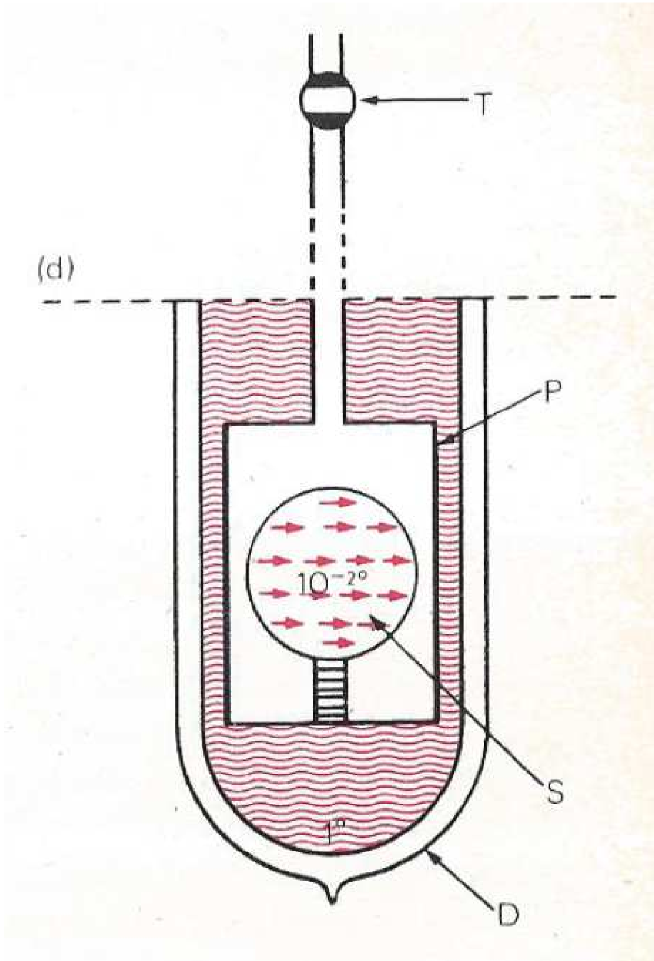
$k_B = 1.3806504\times 10^{-16}$ erg/K (Boltzmann constant)



((**Step-3**)) ((Adiabatic process)) The thermal contact is broken by removing the gas with a vacuum pumping. $\Delta Q = 0$.



((**Step-4**)) ((**Isentropic demagnetization**)) The specimen is then insulated ($\Delta S = 0$). The magnetic field is decreased from $B = B_1$ to $B = B_\Delta$.



The spin entropy is a function of $\frac{T}{B}$.

$$S = S(\frac{T}{B}).$$

The specimen follows the constant entropy path *bc*, ending up to T_2 (isentropic process).

$$\frac{T_2}{B_\Delta} = \frac{T_1}{B}$$

or

$$T_2 = \frac{B_\Delta}{B} T_1 = \frac{100}{10000} 1 = 10^{-2}\ \text{K} = 10\ \text{mK}$$

Using the isentropic demagnetization, the temperature of the sample decreases from 1 K to 10 mK

Note that B_Δ is the effective magnetic field that corresponds to the diverse local interactions among the spins. B_Δ is a small magnetic field, but not equal to zero.

6. Comparison between nuclear demagnetization and spin demagnetization

The nuclear magnetic moment μ_N is much smaller than the electron magnetic moment.

$$\frac{\mu_N}{\mu_B} = \frac{m_p}{m_e} = \frac{1}{1836.15}$$

$$\mu_N = 5.050783699 \times 10^{-24} \text{ emu (-erg/Oe).}$$

$$\frac{\mu_N}{k_B} = 3.65826 \times 10^{-8} \text{ (K/Oe)}$$

which is much smaller than $\frac{\mu_B}{k_B}$,

$$\frac{\mu_B}{k_B} = 6.71713 \times 10^{-3} \text{ (K/Oe)}$$

(a) Nuclear spin de magnetization

When

$$\mu_N B_0 = k_B T_0$$

the characteristic temperature is evaluated as

$$T_0 = \frac{\mu_N}{k_B} B_0 = 3.65826 \times 10^{-8} B_0 .$$

When $B_0 = 1$ kOe, we have $T_0 = 3.65826 \times 10^{-5}$ K $= 36.5826$ μK

(b) Electron spin demagnetization

$$\mu_B B_0 = k_B T_0$$

or

$$T_0 = \frac{\mu_B}{k_B} B_0 = 6.71713 \times 10^{-3} B_0$$

When $B_0 = 1$ kOe, we have $T_0 = 6.71713$ mK

7. Nuclear demagnetization (experiment)

((**Example**))

$B_i = 50$ kOe. $T_i = 0.01$ K

$$\frac{\mu_N B_i}{k_B T_i} = \frac{3.65826 \times 10^{-8}}{0.01} \times (5.0 \times 10^4) = 0.1.82913.$$

or

$$\frac{k_B T_i}{\mu_N B_i} = 0.5465$$

In Cu, typically we have $B_\Delta = 3.1$ Oe. Then we get the isentropic nuclear demagnetization;

$$\frac{B_i}{T_i} = \frac{B_\Delta}{T_f}$$

$$T_f = \frac{B_\Delta}{B_i} T_i = \frac{3.1}{50 \times 10^3} 0.01 = 0.62\ \mu\text{K}.$$

REFERENCES

K. Mendelssohn The Quest for Absolute Zero: the meaning of low temperature physics, second edition with S.I. units (Taylor & Francis, 1977).

K. Mendelssohn, Cryophysics (Interscience, 1960).

APPENDIX

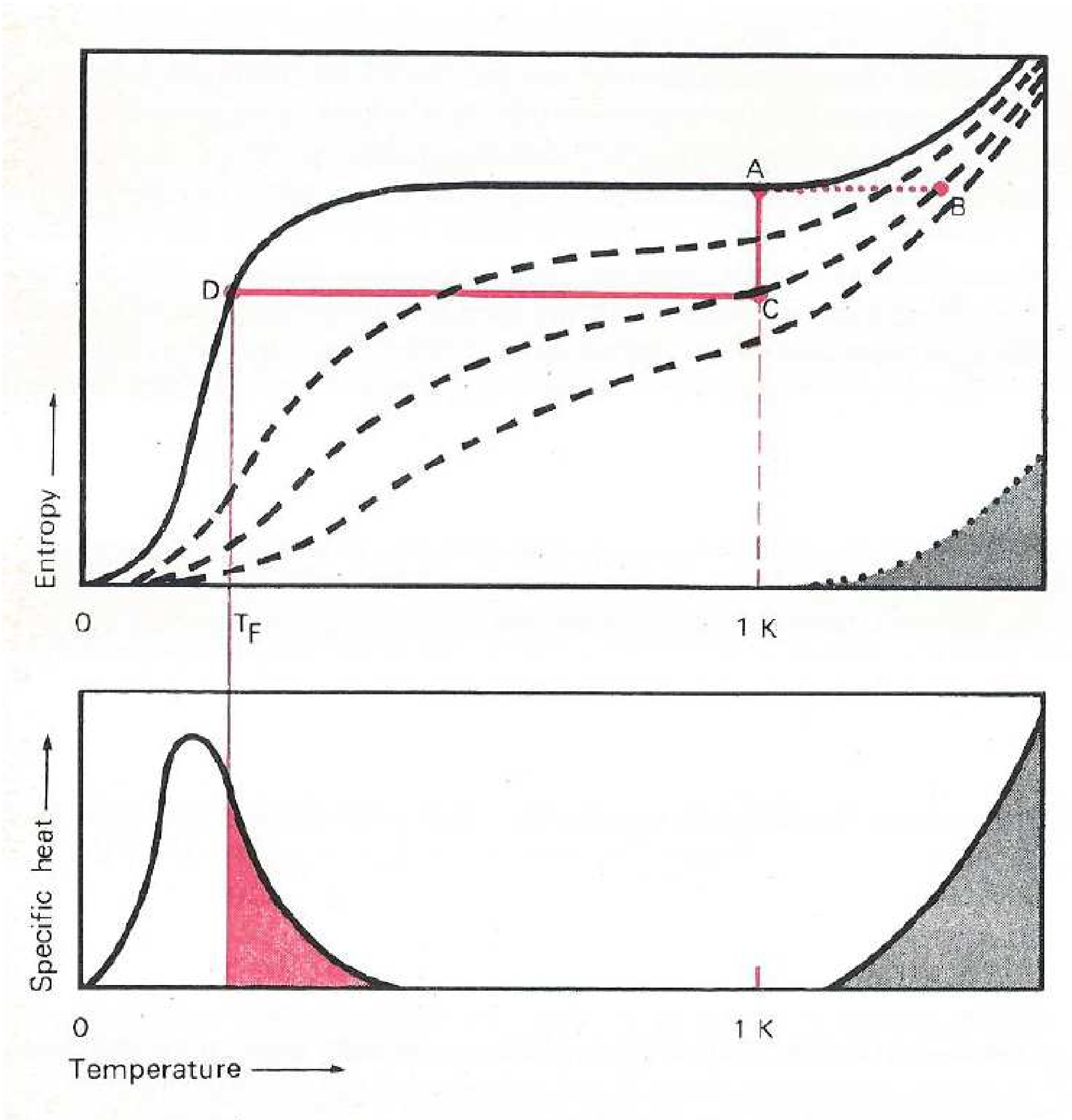


Fig. The entropy diagram of a paramagnetic salt shows how very low temperatures can be reached by the magnetic cooling method.