UCL DEPARTMENT OF SPACE AND CLIMATE PHYSICS
MULLARD SPACE SCIENCE LABORATORY

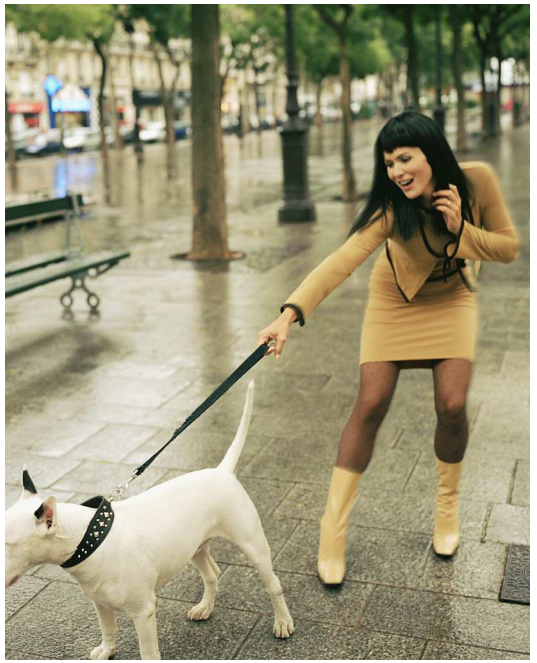
Adiabatic Demagnetization Refrigerator: A Practical Point of View

Ian Hepburn

Cryogenic Physics Group
Mullard Space Science Laboratory
UCL

idh@mssl.ucl.ac.uk

How to Order Entropy



How to Control Entropy

What's in a Name

- ADR
- Magnetic cooling
- Magnetic refrigerator
- Magnetocalorimetric effect

Relationship between magnetic flux density and Magnetic field

$$B = \mu_0 \mu_r H$$

B = Magnetic flux density (Tesla)

H = Magnetic field strength (A m-1)

μ_0 = Permeability of free space

μ_r = Relative permeability of material in magnetic field

Contents

- Suggested texts
- History
- Ideal Theory (briefly)
- Theory in practice
- Paramagnetic materials
- Modelling
- ADR construction

Suggested texts

- Principles and applications of Magnetic Cooling. R.P. Hudson, 1972, ISBN 0 7204 1257 9 (out of print – should be available from national library)
- Experimental Techniques in Low-Temperature Physics. **THIRD EDITION**. Guy K. White, 1979, ISBN 0 19 851381X
- Most LT texts

History

- Cooling to milli-Kelvin temperatures by effecting electron spins was proposed by.
  - Debye 1926.
  - Giauque 1927.
- The first practical demonstration was by.
  - De Haas, Wiersma and Kramers, Giauque and MacDougall 1933.
  - Kurti and Simon 1934.

History

- ~1950s Dilution refrigerators can along and the ADR faded.
- ~1980s/90s Need for milli-kelvin cooling in space drove the resurgence of ADRs due to their gravity independence.
- 2000 – ADRs rising in popularity for quick LT experiments due to their small size, easy of use, ruggedness and quick operation
  - RT to 0.1 K ~ 4 hours (ADR as a LHe dewar insert).

Entropy

- Entropy S can be defined

$$dS = \frac{dQ}{T}$$

- It is a measure of disorder (or order) within a system

Also

$$Q = \int T\,dS \qquad C = T\frac{dS}{dT}$$

Entropy in Relation to Cooling

- A cooling process may be regarded as an entropy reducing process.

- Cooling can be achieved within a medium via any process which results in the decrease of entropy of that medium.

Magnetic Cooling

- Uses the disordered collection of magnetic dipoles associated with a particular ion within a medium (paramagnetic material).

- For such a material the application of a magnetic field causes alignment of the dipoles with the magnetic field and thus a reduction in entropy.

Entropy of Paramagnetic Material

- S=Sm+Slattice

Material Characteristics

- Interaction Energy of magnetic ion given by ε.
- When $kT > \varepsilon$ each magnetic ion is relatively "free" resulting in a distribution of randomly oriented dipoles with 2J+1 degeneracy.
- We have R.Ln(2J+1) per mole contribution to the entropy of the material from the magnetic dipoles.

Temperature Behaviour

- At high temperature $S_{lattice} > S_m$
- As temperature is reduced $S_{lattice} < S_m$

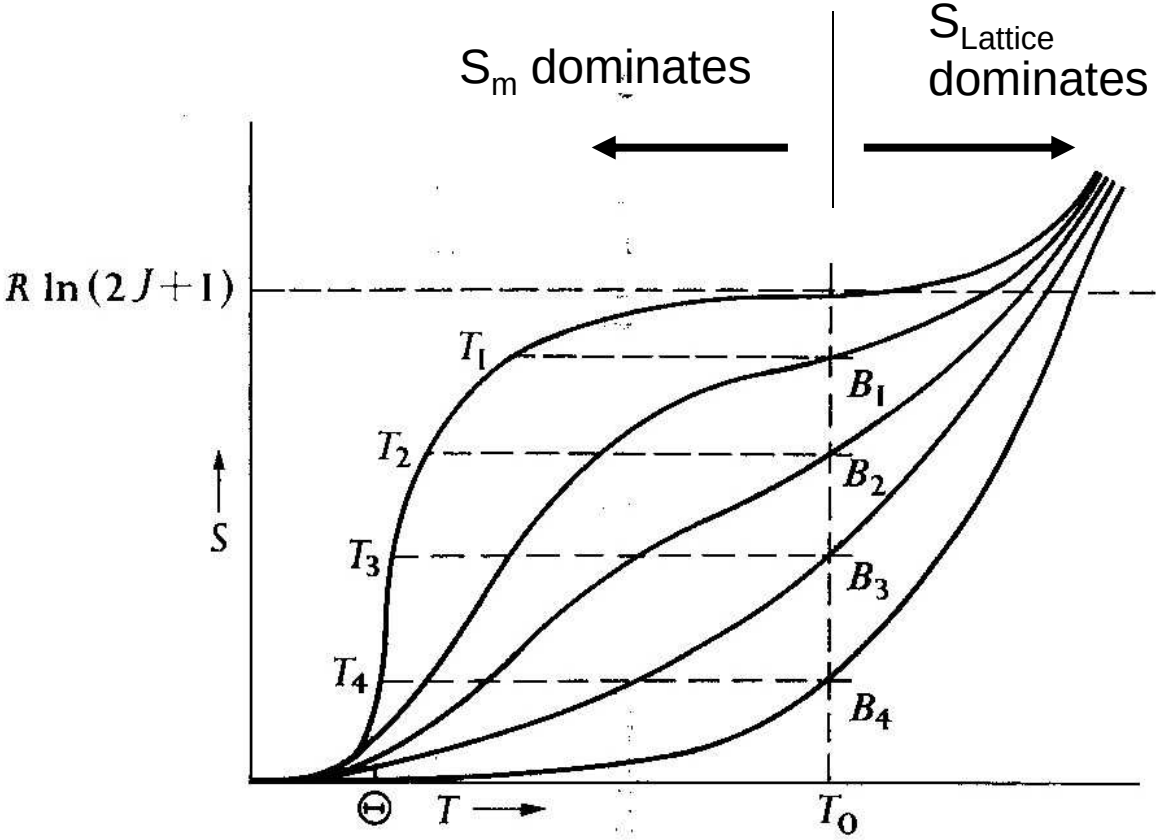
  (magnetic entropy given by RLn(2J+1) dominates)

- As temperature reduces further
  - $\varepsilon \sim k\theta$

    (θ is the magnetic ordering temperature)
  - spontaneous ordering of the dipoles occurs, due to their own weak magnetic fields, and the entropy falls

- At a temperature greater than θ the entropy of the spin system of the magnetic ions can be reduced significantly if the interaction of the dipoles and the applied magnetic field is greater than the thermal excitation given by kT.

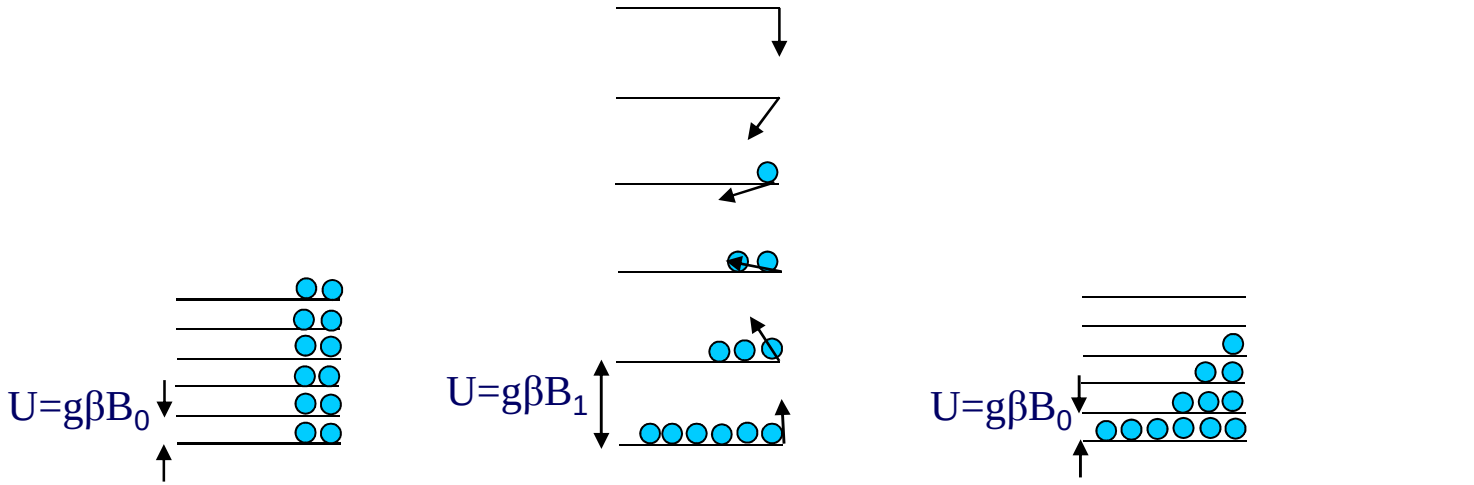
Entropy/Temperature/magnetic field plot for a paramagnetic material



How is cooling achieved?

- Boltzmann distribution factor exp(U/kT), U= gβB

$B=B_0$, $T= T_0$ | $B=B_1$, $T=T_0$ | $B=B_0$, $T=T_1$



a) Dipoles have no preferred orientation

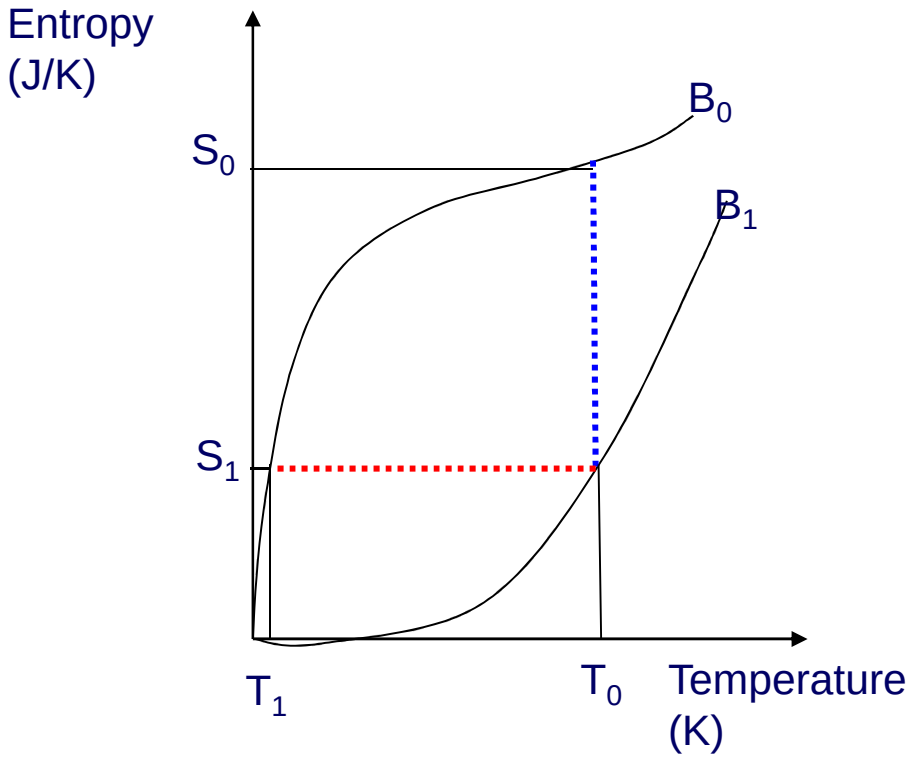
b) Dipoles align in the direction of the applied field.

c) Dipoles remain ordered as external field is removed.

- $T_1 = T_0 (B_0/B_1)$

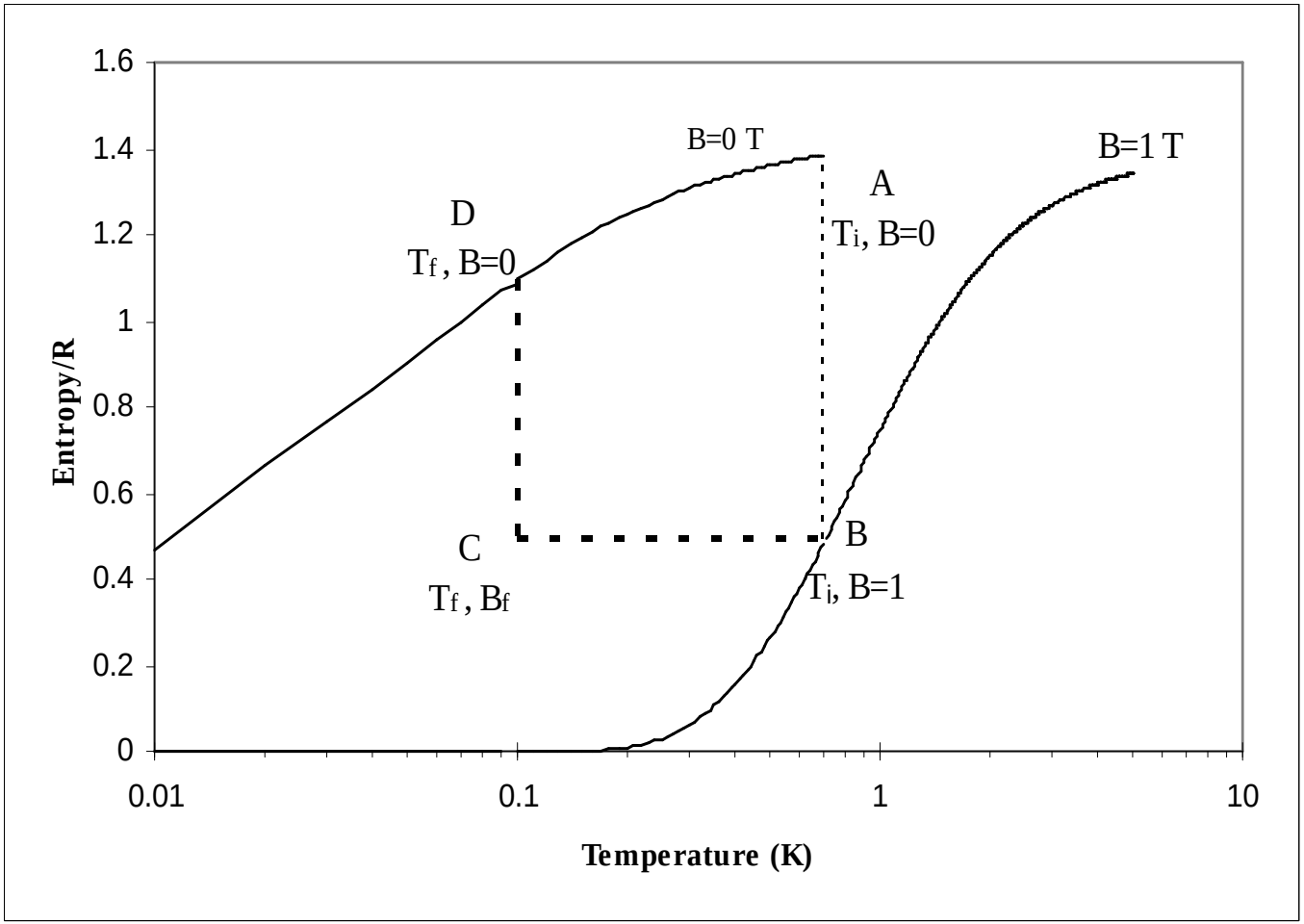
Magnetic Cooling Process

- Process of isothermal magnetisation and adiabatic demagnetisation
- Uses the re-ordering of magnetic dipole moments
- Millikelvin temperatures are achievable

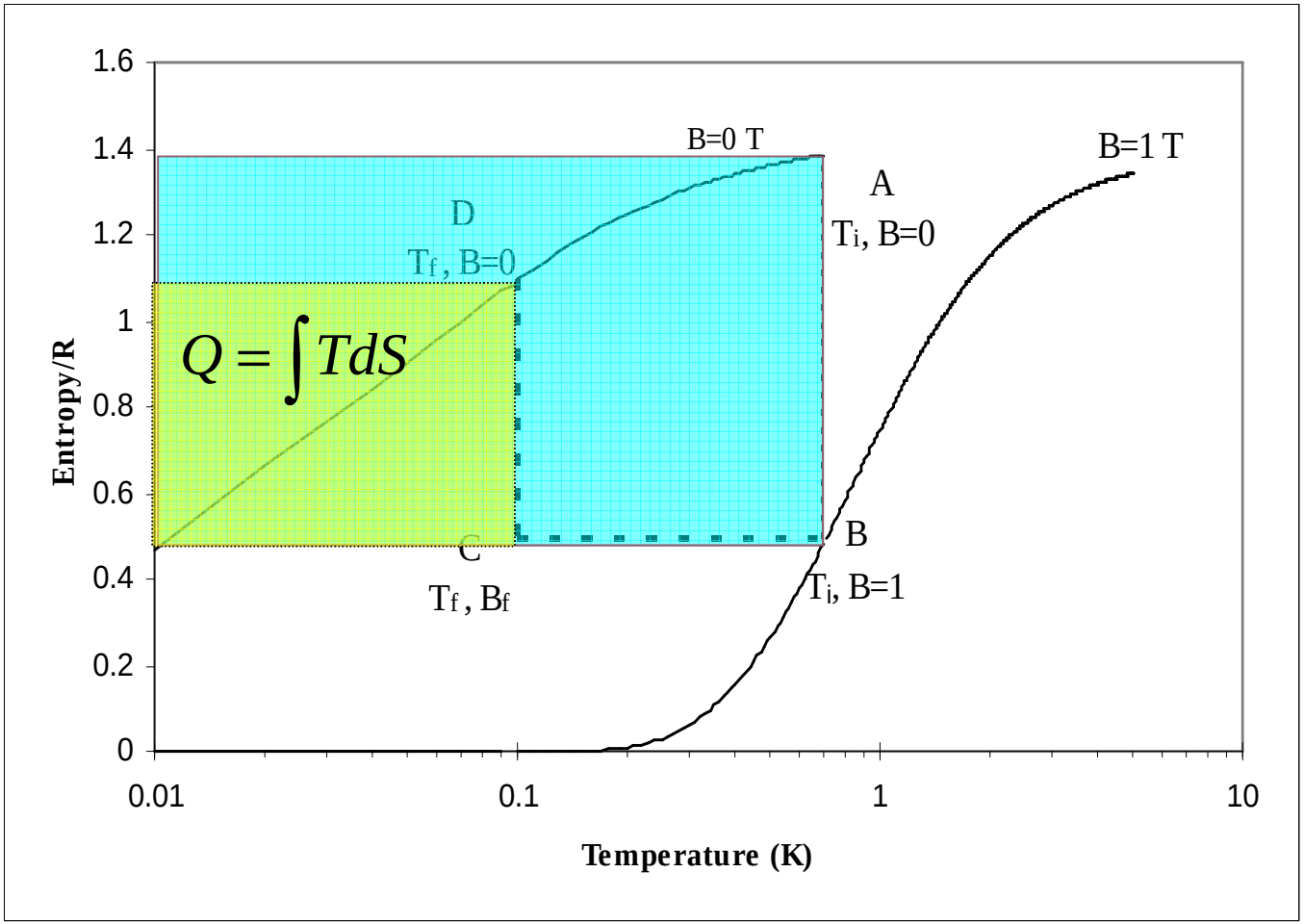


Isothermal magnetisation and isentropic demagnetisation between constant magnetic field entropy-temperature curves

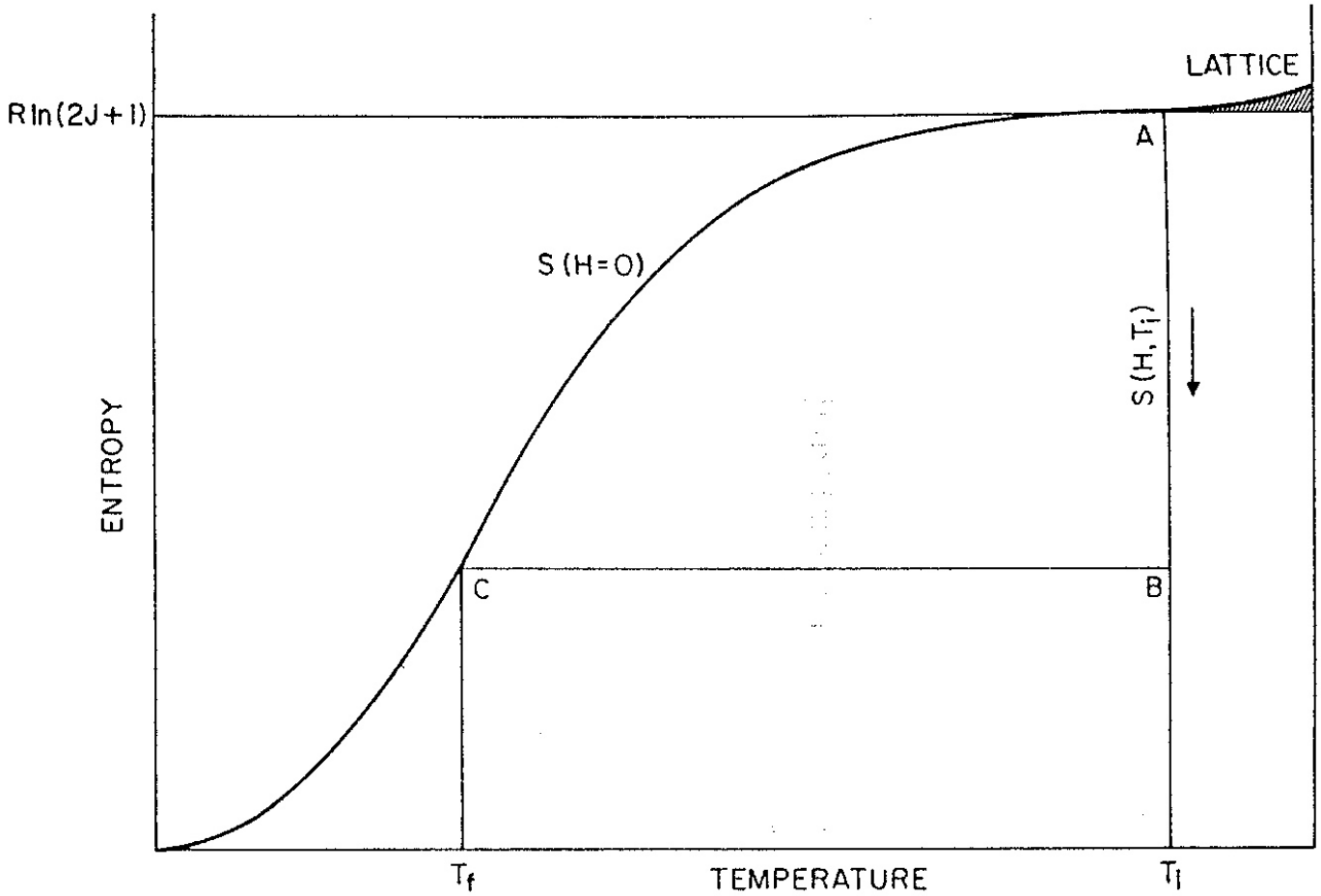
Magnetic Cycle



Energy in Magnetic Cycle



Ideal Theory



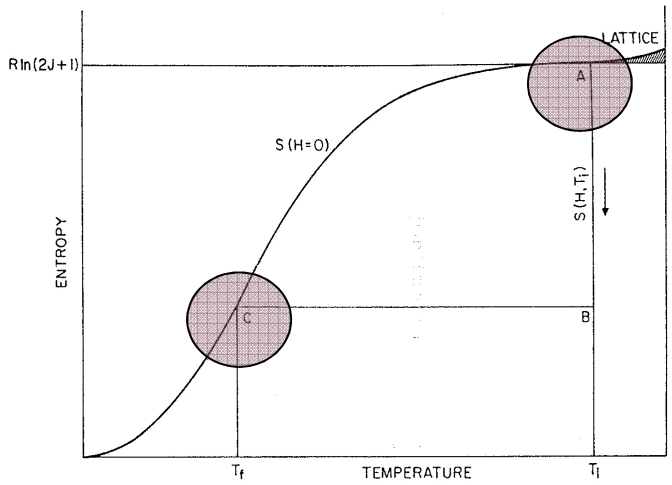
Ideal Theory: point A & C

- The following equation can be derived.

$$\frac{S(T)}{R} = Ln(2J+1) - \frac{b}{2T^2} + \frac{aT^3}{3}$$

Interactions term

Lattice term



Breaks down at low T due to the T^{-2} term. As $T \rightarrow 0$, $T^{-2} \rightarrow \infty$

At low T The Schottky-Effect anomaly in the heat capacity occurs and stops $S/R \rightarrow -\infty$

Ideal Theory: point B

- For a collection of free ions the following can be derived

Free Ion approximation Equation

$$\frac{S(H,T)}{R} = Ln\left[\frac{\sinh(2J+1)\frac{x}{2}}{\sinh\frac{x}{2}}\right] + \frac{x}{2}\coth\frac{x}{2} - \frac{(2J+1)x}{2}\coth\frac{(2J+1)x}{2}$$

Where

$$x = \frac{g\beta H}{kT}$$

J = Total angular momentum quantum number

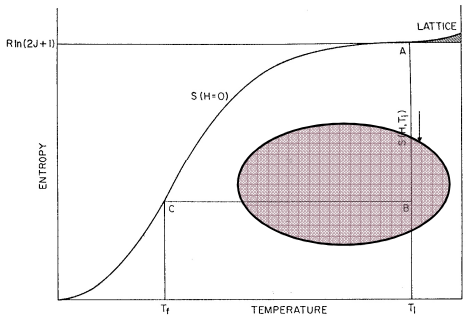
g= spectroscopic splitting factor

β= Bohr magneton

H= Magnetic field intensity

k= Boltzmann constant

T= Temperature



Extending Ideal Theory: point B

- Free ion approximation is a simple approximation.
  - Assumes Ion is free.
  - Does not take into account crystal effects
  - Main effect to be considered is Crystal Field Splitting

Ideal Theory Incorporating Crystal Field Splitting: point B

- Weak magnetic field case. Where gβH and δ are small compared to kT

$$\frac{S}{R} = Ln(2J+1) - \frac{5g^2\beta^2H^2}{8k^2T^2} - \frac{\delta^2}{8k^2T^2}$$

- Strong magnetic field case. Where gβH is very much greater then δ

$$\frac{S}{R} = Ln2(\cosh\frac{3x}{2} + \cosh\frac{x}{2}) + \frac{T\frac{\partial}{\partial T}(\cosh\frac{3x}{2} + \cosh\frac{x}{2})}{(\cosh\frac{3x}{2} + \cosh\frac{x}{2})} - \frac{1}{8}\left(\frac{\delta}{kT}\right)^2\left[\frac{\sec h^2 x}{3} + \frac{\sec h^2\frac{x}{2}}{6}\right]$$

Where δ is the energy splitting. All other symbols are as before

Using these equations

- Free ion approximation okay if large molecule and ions are widely separated (eg CPA).
- Incorporating crystal field splitting
  - Use weak field when $\frac{g\beta H}{k} \leq 0.3$
  - Use strong field when $\frac{g\beta H}{k} > 0.3$

Relationship between initial and final temperatures from ideal theory

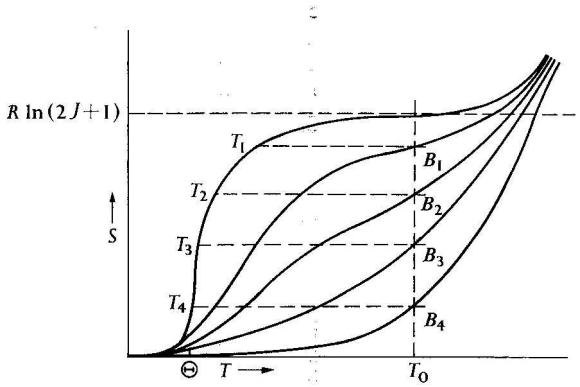
Final temperature on demagnetization is typically given by:

$$T_f = \frac{H_f T_i}{H_i}$$

This violates the third law.

This approximation has omitted the internal magnetic field associated with the ion. Better approximation is:-

$$T_f = \frac{(h^2 + H_f^2)^{\frac{1}{2}}}{(h^2 + H_i^2)^{\frac{1}{2}}}$$



Where

T_f = Final temperature

T_i = Initial temperature

H_f = Final Magnetic field strength

H_i = Initial magnetic field strength

h = magnetic field strength associated with the ion.

Theory in practice

- Ideal theory only concerns the magnetic ion in the molecule.
  - Lattice contribution is taken to be negligible
  - It ignores any associated components.
    - Items attached to the paramagnetic (e.g. Items to be cooled)

Use theory with care – Indicator only

Paramagnetic materials

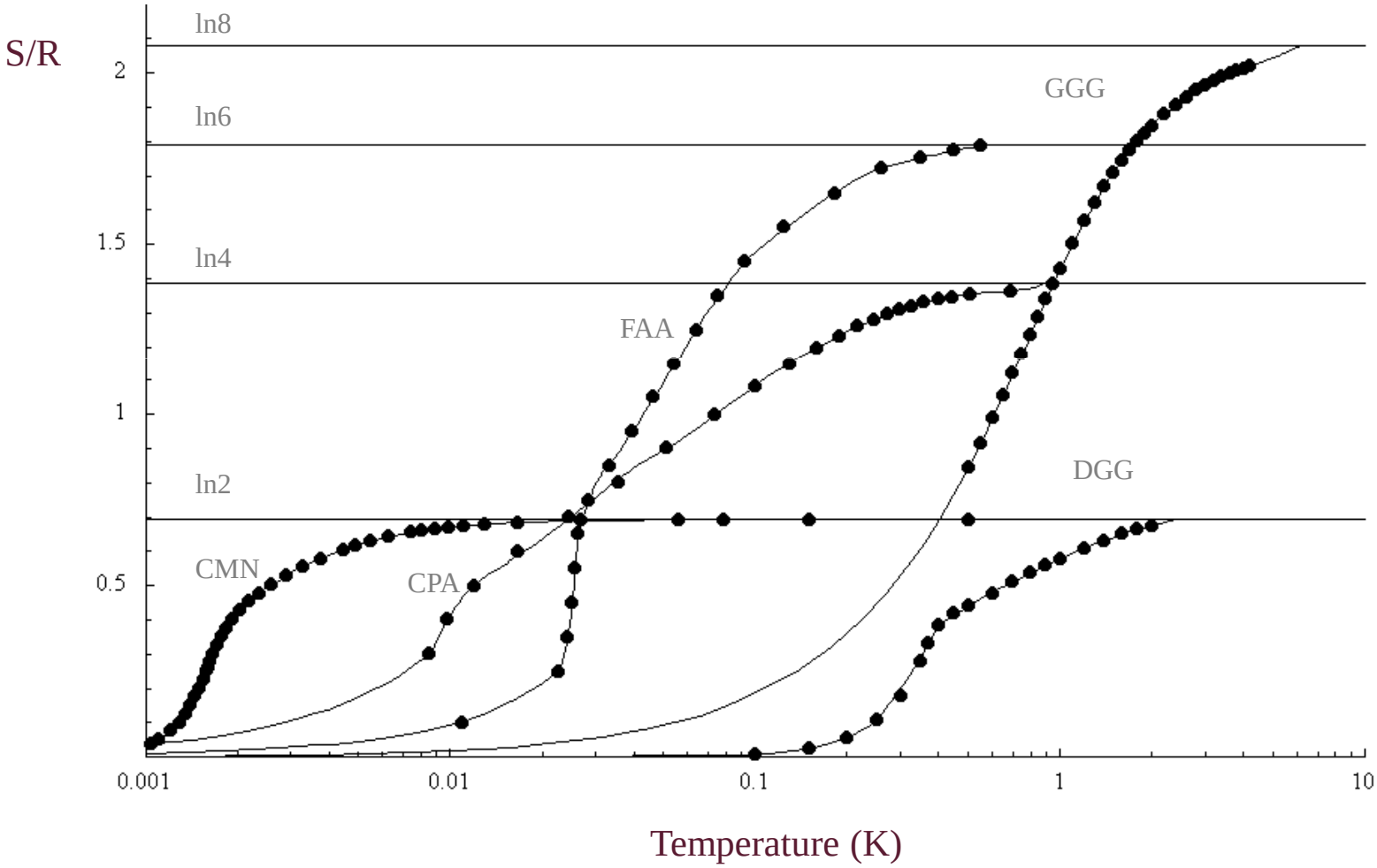
- A wide range of paramagnetics exist.
  - Rare earths
    - Salts
    - Garnets
    - Perovkites (Orthoaluminates)

Paramagnetic Materials

Temperature Range	Material	Formation	J	g	Tn (K)
10- 40 mK	Cerium Magnesium Nitrate (CMN)	$\underline{Ce}_2Mg_3(NO_3)_{12}.24H_2O$	1/2	2	~0.01
40->100 mK	Chromic Potassium Alum (CPA)	$\underline{Cr}K(SO_4)_2.12H_2O$	3/2	2	~0.01
	Cesium Chromic Alum (CCA)	$\underline{Cs}Cr(SO_4)_2.12\ H_2O$	3/2	2	~0.01
	Ferric Ammonium Alum (FAA)	$\underline{Fe}NH_4(SO_4)_2.12H_2O$	5/2	2	~ 0.03
>0.3 K	Manganese Ammonium Sulfate (MAS)	$\underline{Mn}SO_4(NH_4)_2SO_4.6H_2O$	5/2	2	~0.1
>1 K	Dysprosium Gallium Garnet (DGG)	$\underline{Dy}_3Ga_5O_{12}$	1/2	8	~0.4
	ErOA	$\underline{Er}AlO_3$	1/2	9	~0.6
	YbOA	$\underline{Yb}AlO_3$	1/2	7	~0.8
	Gadolinium Gallium Garnet (GGG)	$\underline{Gd}_3Ga_5O_{12}$	7/2	2	~0.8
	Dysprosium Aluminum Garnet (DAG)	$\underline{Dy}_3Al_5O_{12}$	1/2	11	~2.5
	DOA	$\underline{Dy}AlO_3$	1/2	14	~3.5
	GOA	$\underline{Gd}AlO_3$	7/2	2	~3.8

_ denotes active ion i.e. the ion that provides cooling

Paramagnetic Materials



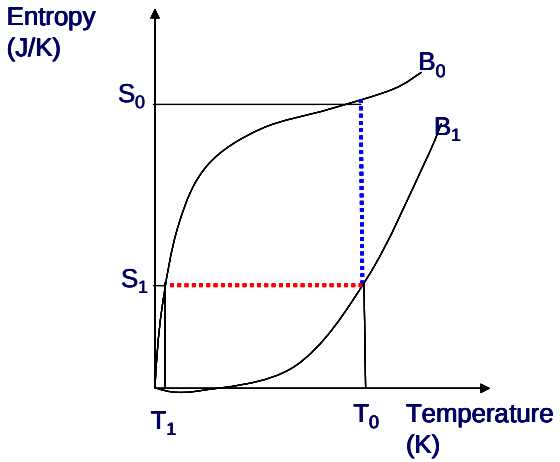
Modelling

- Ideal theory only applies to the active ion in the paramagnetic.
- In a real system the demagnetisation is not adiabatic
- In order to determine how cold our ADR will get and how long it will stay cold we need to understand:-
  - Heat capacity of everything attached to the paramagnetic (this limits final temperature)
    - paramagnetic container
    - Thermal connection to experiment interface
    - The experiment
    - Eddy current heating due to changing magnetic field
  - All heat sources on paramagnetic (determines hold time)
    - Paramagnetic support thermal conductivity (e.g. kevlar)
    - All thermal radiation
    - Thermal conductivity of electrical wires
    - Optical radiation (if used for detectors)
    - Any other connection from another temperature (e.g. capillaries for pressure cells)

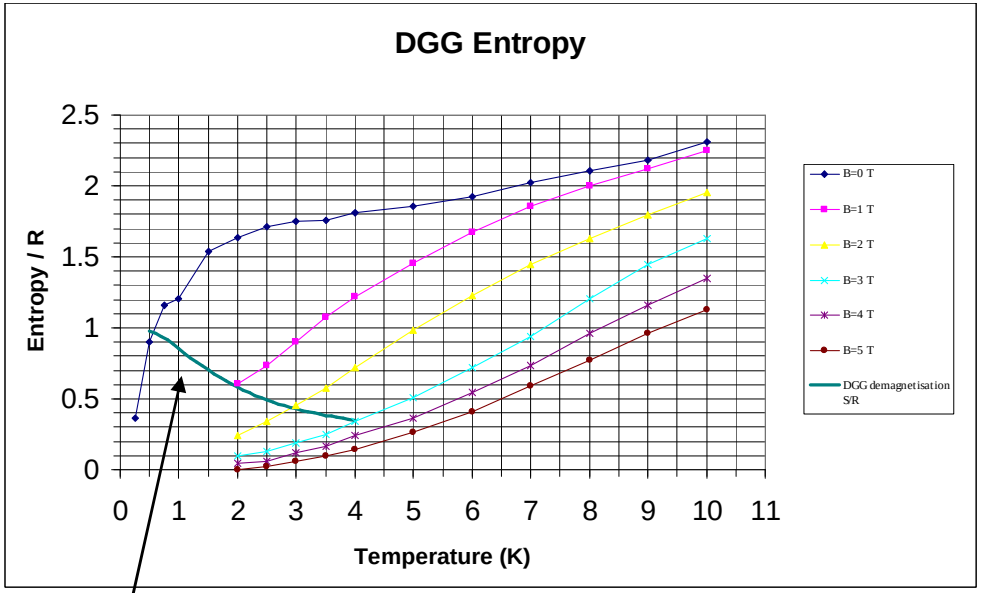
Estimating final temperature

- In order to establish final temperature and hold time we need to model.

Consider cooling a non negligible heat capacity



Ideal heat capacity not considered



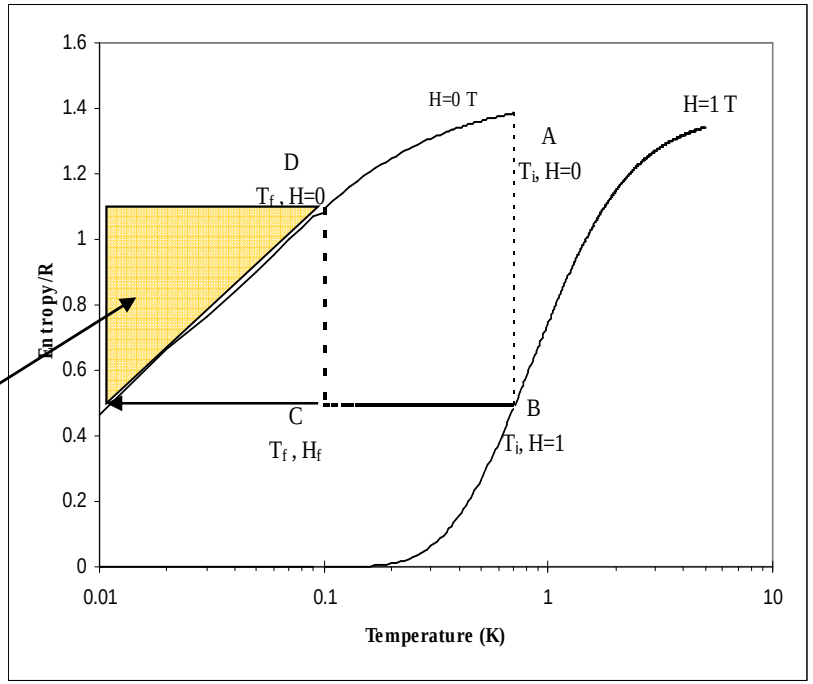
Modelled demagnetisation path. Not adiabatic due to paramagnetic asorbing heat capacity of components attached to it

Providing low temperatures

- Un-regulated
  - Demagnetisation to lowest temperature
  - Allow paramagnetic to warm up

Energy (Q) absorded per mole between T_1 and T_2 is given by

$$Q = \int_{S_1}^{S_2} TdS$$

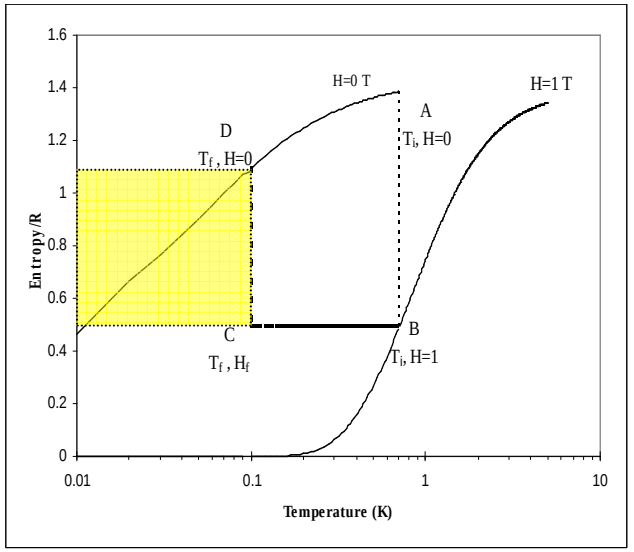


Providing a constant low temperature

Regulated temperature

•Demagnetisation to a value corresponding to the required temperature

•Controlled demagnetisation at a rate to maintain temperature (servo)



Energy absorbed per mole is

$$Q = \int_{S_1}^{S_2} T dS$$

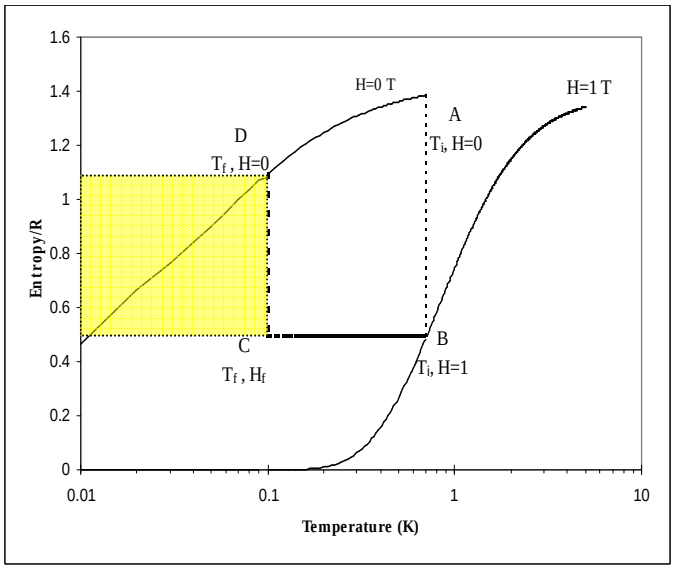
But since T is constant

$$Q = T\Delta S = T(S_2 - S_1)$$

Hold time

- The time at a particular temperature

$$Hold\_Time = \frac{nT_f(S_c - S_{D)}}{\frac{dQ}{dt}}$$



Where:

n = Number of moles of magnetic ion

dQ/dt = Total thermal power into the paramagnetic material

T_f = regulated temperature

S_c = Entropy at point C

S_D = Entropy at point D

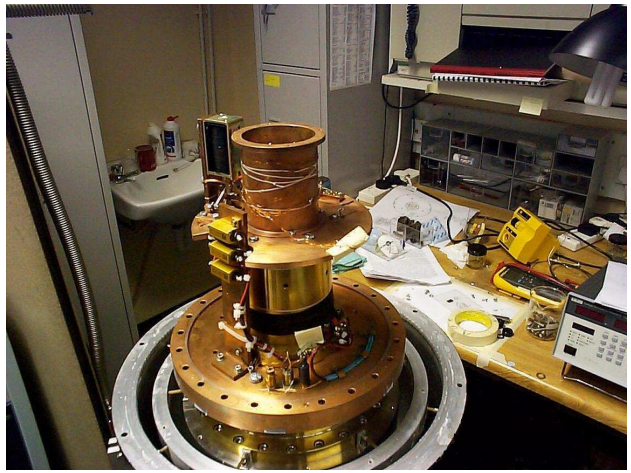
Once all the magnetic field is used up either:

Recycle (i.e. magnetise and then demagnetise

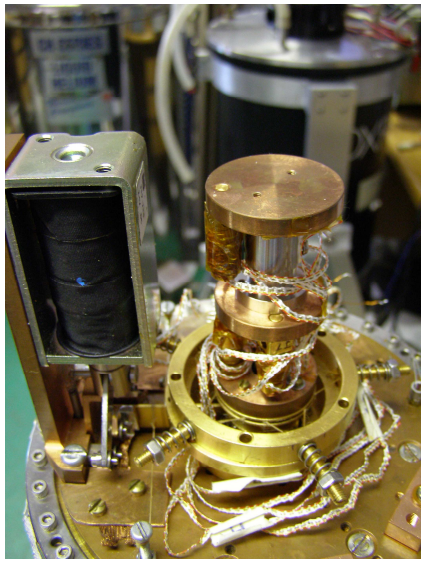
or

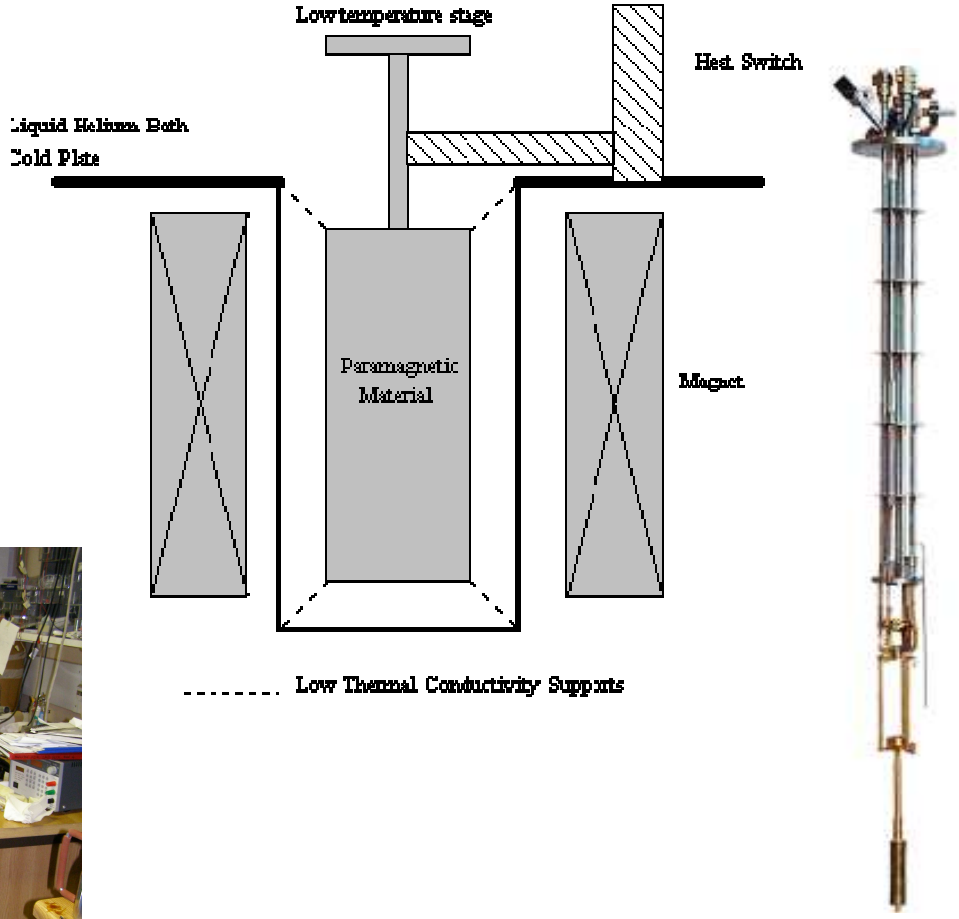
Go to a higher temperature

ADR construction – Basic ADR



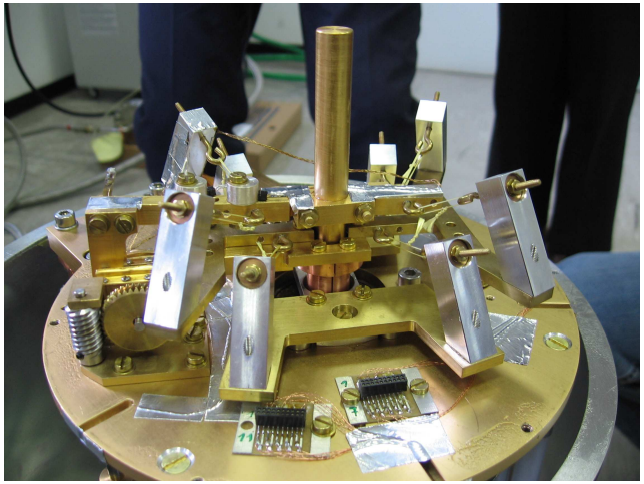
Two of MSSLs Lab ADRs



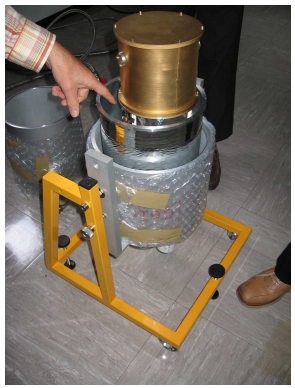


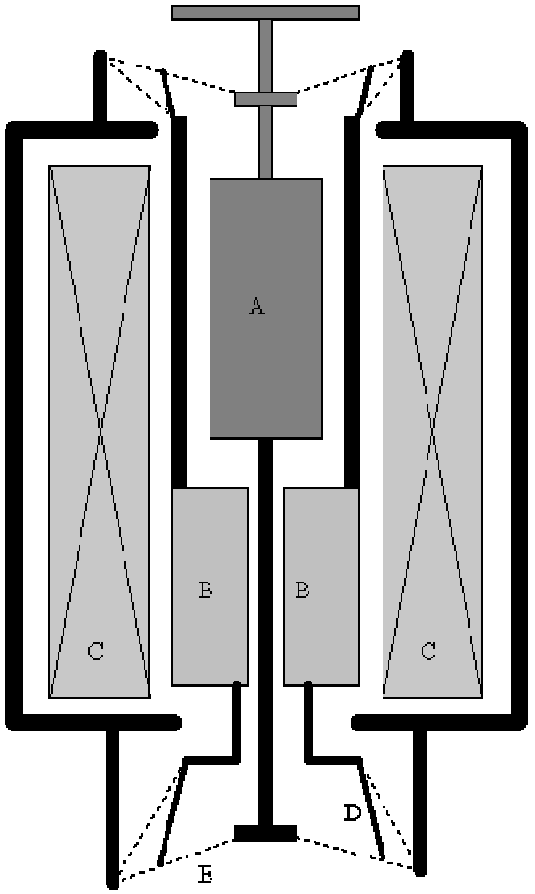
CMR Lab ADR Insert

ADR construction – Two Stage

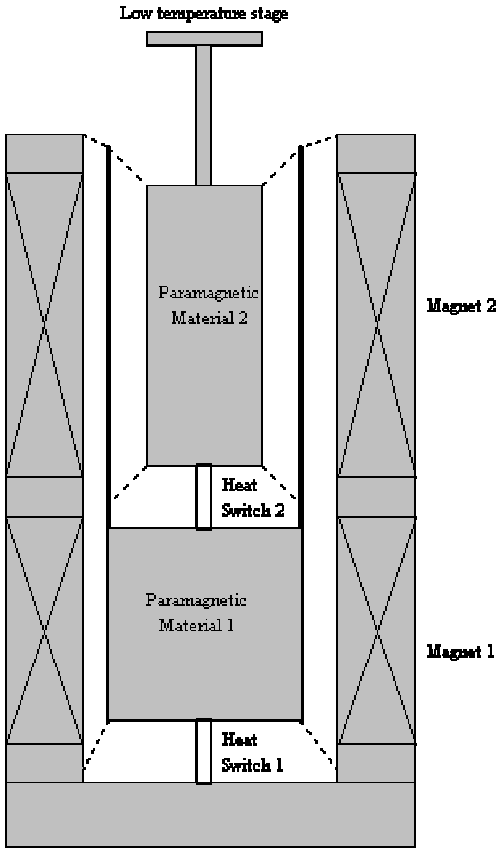


Vericold Lab ADR





ADR construction – Double ADR



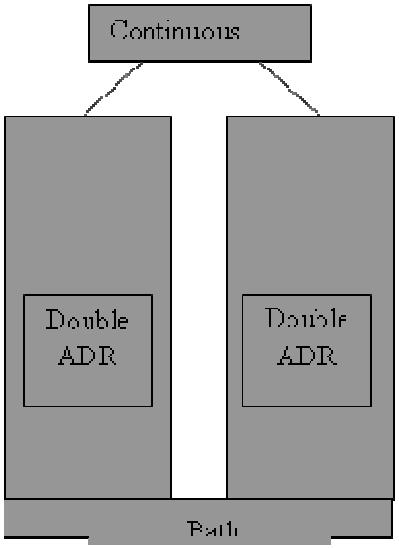
MSSLs double ADR (for space use)

ADR construction - Continuous

Tandem type

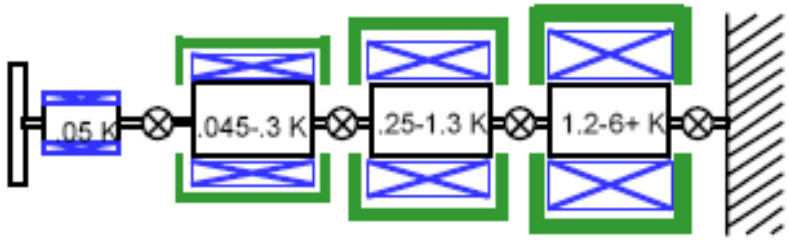
Continuous temperature from bath temperature to milli-kelvin

(under development at MSSL)



Series type

Continuous temperature <100 mK. (paramagnetic material chain dependant)



GSFC Lab based demonstrator

Troubleshooting

Not getting the anticipated temperature or hold time
Typical problems are:

- Trying to cool too big an experiment (to much heat capacity)
- Heat load on to the paramagnetic too high.
  - Too many wires
  - Too high thermal load (e.g. wires, heat switch, radiation).
- Thermometry problems (e.g. thermal contact, self heating)

<u>Usually the paramagnetic does what it was supposed to do</u>

– Don’t blame the Paramagnetic material