

# 1. ADR General Description

## 1.1. Principle of Magnetic Cooling

A cooling process may be regarded as an entropy reducing process. Since entropy (or degree of disorder) of a system at constant volume or constant pressure decreases with decreased temperature, cooling can be achieved within a medium via any process, which results in the decrease of entropy of that medium. For example, the liquification of gases is achieved by the isothermal reduction of entropy through compression of a volume  $V_1$  at temperature  $T_1$  to a smaller volume  $V_2$  generating heat, which is extracted, followed by adiabatic or isentropic expansion which results in cooling of the gas to below  $T_1$ .

In the magnetic cooling process the disordered collection of magnetic dipoles associated with a particular ion within a medium (paramagnetic material) constitutes such a system described above. 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. Cooling via this process was proposed by Debye<sup>i</sup> in 1926 and Giauque<sup>ii</sup> in 1927. The first practical demonstration was by De Haas, Wiersma and Kramers<sup>iii</sup>, Giauque and MacDougall<sup>iv</sup> in 1933 and Kurti and Simon<sup>v</sup> in 1934. Since those pioneering days numerous text on magnetic refrigeration (the generic term for adiabatic demagnetisation refrigeration) have been published and most low temperature physics books contain a chapter describing it. For further information see references <sup>vi</sup>, <sup>vii</sup> and <sup>viii</sup>.

Certain paramagnetic materials are suitable for use as magnetic refrigerants. The magnetic ions of these materials have an interaction energy,  $\epsilon$ , with their crystalline environment and each other which is smaller than the average thermal energy  $kT$ . In such a situation each magnetic ion is relatively free resulting in a distribution of randomly oriented dipoles, which is  $2J+1$  degenerate, where  $J$  is the angular momentum quantum number. This means that there are  $2J+1$  possible orientations of the ions. This gives an  $R\ln(2J+1)$  per mole contribution to the entropy of the material from the magnetic dipoles, where  $R$  is the gas constant.

As the temperature of the paramagnetic material is reduced the lattice contribution to the entropy of the material reduces and a point is reached where in the magnetic entropy given by  $R\ln(2J+1)$  dominates. As the temperature decreases further the entropy will remain at the value given by  $R\ln(2J+1)$  until the thermal energy approaches the interaction energy  $\epsilon$  at which point spontaneous ordering of the dipoles occurs, due to their own weak magnetic fields, and the entropy falls. When  $\epsilon \sim k\theta$ , where  $\theta$  is the magnetic ordering temperature of the material (or Néel temperature), the entropy drops rapidly. At very low temperature the internal interactions between ions removes the degeneracy and the system resides in a singlet ground state of zero entropy. At a temperature greater than  $\theta$  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  $kT$ . If the internal interaction energy is very low the magnet ions can be considered as free and the entropy ( $S$ ) of a collection of magnetic ions can be given by Equation 1. Figure 1 shows the typical form of the entropy curve as a function of temperature and applied magnetic field.

$$\frac{S(B,T)}{R} = L_n \left[ \frac{\sinh((2J+1)x/2)}{\sinh(x/2)} \right] + \frac{x \cdot \coth(x/2)}{2} - \frac{(2J+1) \cdot x \cdot \coth((2J+1)x/2)}{2} \quad (1)$$

Where

$$x = g\beta B/kT \quad (2)$$

and

$g$  = spectroscopic splitting factor

$\beta$  = Bohr magneton

$B$  = magnetic field (Gauss)

$k$  = Boltzmann constant

$T$  = temperature (K)

## 1.2. Magnetic Cooler Operation.

The operation of a magnetic refrigerator (ADR) requires the system to be at a temperature in which the lattice entropy does not dominate the paramagnetic material in order that applied magnetic field can reduce the entropy of the material. The process of cooling can be separated in to three stages. The first is isothermal magnetization of the paramagnetic material at a temperature  $T_1$  (Figure 2), transferring the paramagnetic material from point A (Figure 2) to point B. This process generates heat (magnetization energy), which has to be extracted to a heat sink at a temperature of  $T_1$ , via a heat switch. The magnetization energy ( $Q$  Joules per mole) is given by

$$Q = \int TdS \quad (3)$$

and since  $T$  is constant

$$Q = T_1 [S_A - S_B]. \quad (4)$$

Adiabatic demagnetisation forms the second stage in which the magnetic field is reduced to a value  $B_f$  that corresponds to the desired final temperature  $T_f$ . During this stage the entropy of the paramagnetic material remains constant, resulting in cooling as given by equation (5)

$$\frac{\sqrt{B_i^2 + b^2}}{T_i} = \frac{\sqrt{B_f^2 + b^2}}{T_f} \quad (5)$$

Where

$B_i$  = Initial magnetic field

$B_f$  = Final magnetic field

$b$  = Internal magnetic field associated with each magnetic ion

$T_i$  = Initial temperature

$T_f$  = Final temperature

The third stage can be effective in two ways, the first and most thermally efficient is isothermal demagnetisation. This provides stability at  $T_f$  by reducing the magnetic field  $B$  from  $B_i$  to zero at a rate, which counteracts the thermal input from the surrounding environment. The total amount of energy the paramagnetic material can absorbed is given by equation 3 and equation 6 below, since  $T$  is constant,

$$Q = T_2 (S_C - S_D). \quad (6)$$

The duration in seconds of operation at  $T_f$ , called the hold time, is given by

$$\text{Hold time} = \frac{nT_f(S_C - S_D)}{\frac{dQ_{th}}{dt}} \quad (7)$$

Where

$n$  = Number of moles of magnetic ion

$dQ_{th}/dt$  = Total power into the paramagnetic material  
(e.g. parasitic heating).

Once the magnetic field reaches zero the whole process must be repeated (recycled) from stage 1. The second way is for the magnetic field to be reduced to zero, completely demagnetising the paramagnetic salt to a temperature of  $T_{min}$ . A constant temperature above  $T_{min}$  can be achieved by using a stage which is heated via a heater (resistor) which has a weak thermal link to the paramagnetic material. The heater power needs to be reduced as the paramagnetic material warms under parasitic load. Since the temperature of the paramagnetic material is not constant the energy which can be absorbed is given by equation 3 only. This process is less thermodynamically efficient to the isothermal process however it does not require active control of the magnetic field and therefore is simpler in some respect.

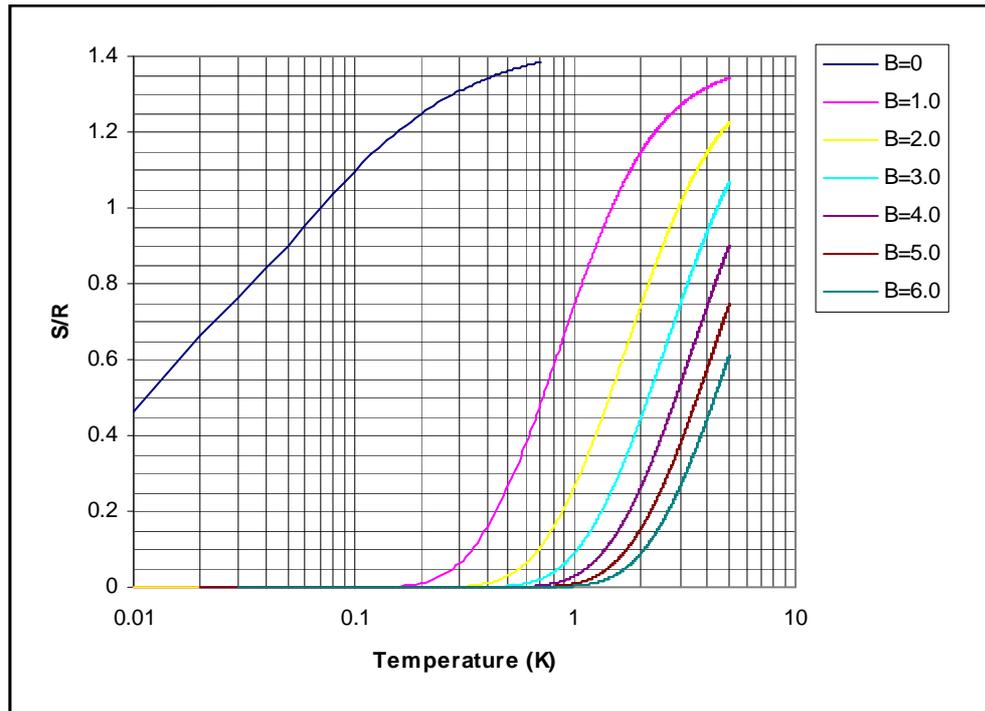


Figure 1 Typical Behaviour of Entropy with Temperature and Magnetic Field.

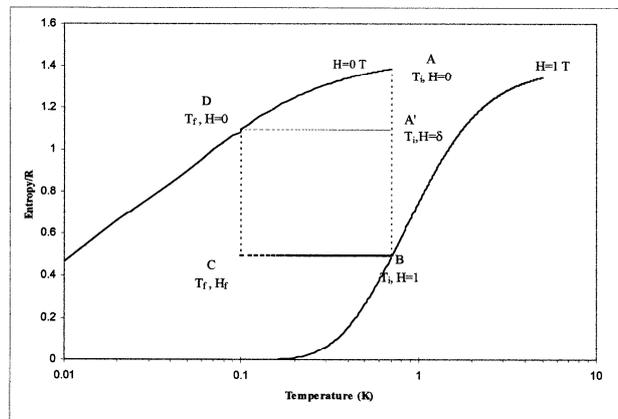


Figure 2 Typical Magnetic Cycle of an ADR.

### 1.3. ADR forms

#### Adiabatic Demagnetisation Refrigerator Configurations

An adiabatic demagnetisation refrigerator (ADR) is essentially composed of the following three items:

1. A paramagnetic material. This is suspended via low thermal conductivity materials within an enclosure at the bath temperature, usually pumped liquid helium in laboratory systems. The paramagnetic material is usually integrated with a stage in order to give access to the temperature obtained on demagnetising the paramagnetic material.

2. A magnet. This may either be a permanent or superconducting magnet. The latter is usually used due to their ease of operation and compactness
3. A heat switch. This is used to make and break a high thermal conductivity path between the paramagnetic material and the liquid helium bath. This is used in order to cool the paramagnetic material to the starting temperature and to extract the heat of magnetization. The form of this switch may be mechanical, gaseous or superconducting<sup>viii</sup>.

A schematic of these components which make up what can be called a “classical” ADR is shown in Figure 3. Variations on the classical form arise due to requirements to a) increase the low temperature hold time by reducing the parasitic heat leak caused by the supports (two stage ADR, Figure 4) and b) increasing the bath temperature (Double ADR, Figure 5).

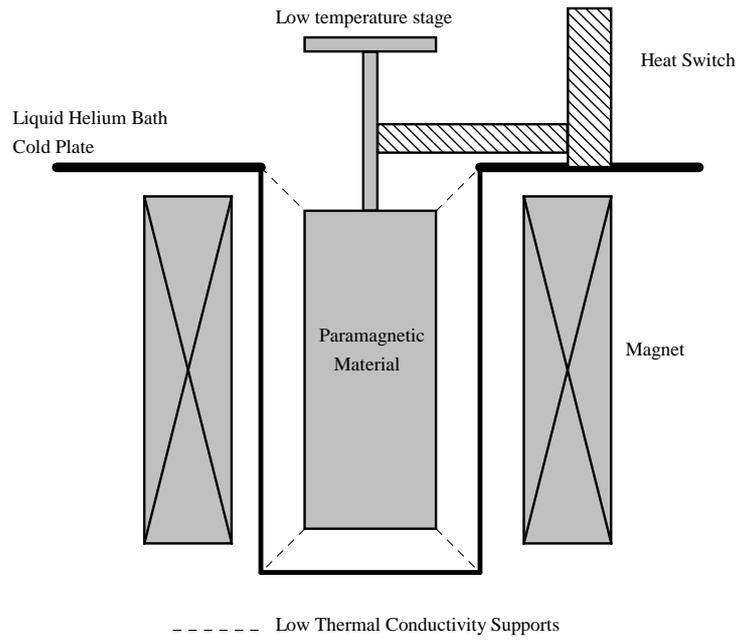
A two stage ADR (Figure 4) uses a second (intermediate) paramagnetic (B) material to intercept the heat leak from the bath to the low temperature paramagnetic material (A). The intermediate material typically comprises a higher temperature material which has a high heat capacity, e.g. GGG. The operation of the ADR is the same as the conventional form except the intermediate paramagnetic material will demagnetise to a temperature in between that of the bath and the low temperature material.

The double ADR (Figure 5) is essentially two ADRs in series one configured for high temperatures and the other for low temperatures. The high temperature ADR (paramagnetic material 1) is used to cool the low temperature stage (paramagnetic material 2) prior to the demagnetisation of that stage. This effectively simulates a lower temperature bath for a conventional ADR. The operation sequence is as follows:

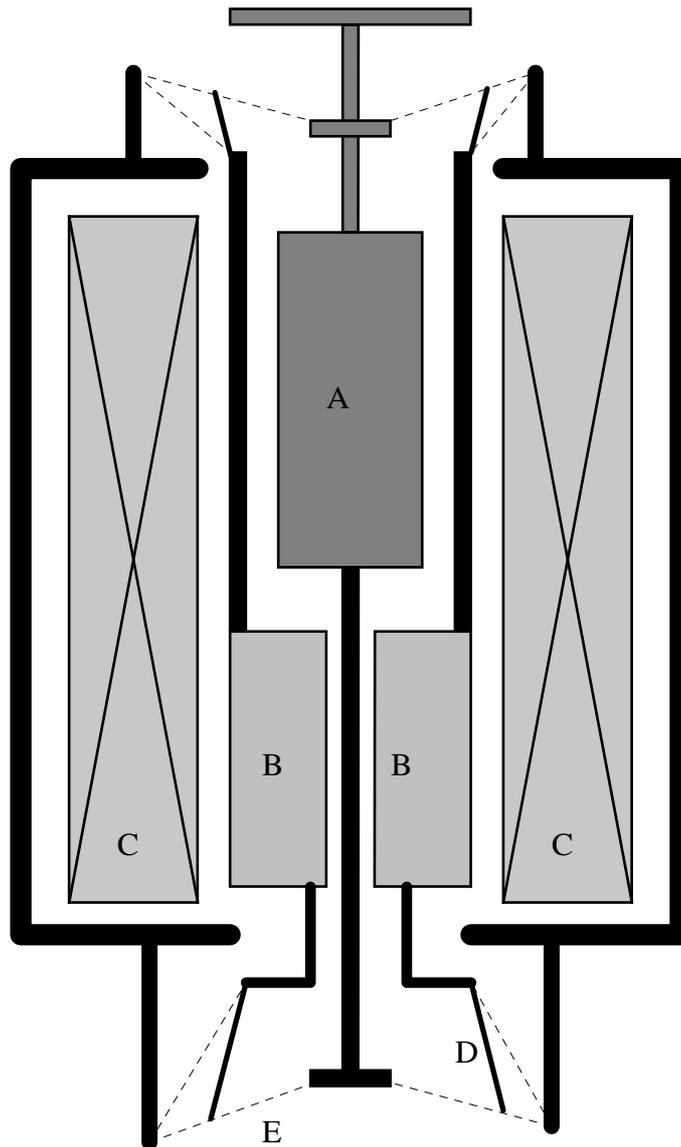
1. Magnetization of both paramagnetic materials with both heat switches closed.
2. Open heat switch 1
3. Demagnetised paramagnetic material 1, thereby reducing the temperature of the magnetized material 2
4. Open heat switch 2
5. Demagnetised paramagnetic material 2
6. Operation of material 2 as conventional ADR, while material 1 slowly warms due to the input power.

By operating two double ADRs in tandem, as shown in Figure 6 a continuous operating cold stage can be provided.

A further variation of the classical ADR is the “hybrid” (Figure 3). This consists of a conventional ADR coupled to a low temperature cryogenic stage provided by He<sup>3</sup>. The He<sup>3</sup> stage is used to dramatically reduce the parasitic heat load and provided a very low starting temperature for the paramagnetic material.



**Figure 3 Schematic of a “classical” Single stage ADR and hybrid ADR**  
 (For hybrid the helium bath is replaced with a helium 3 refrigerator at 0.3 K)



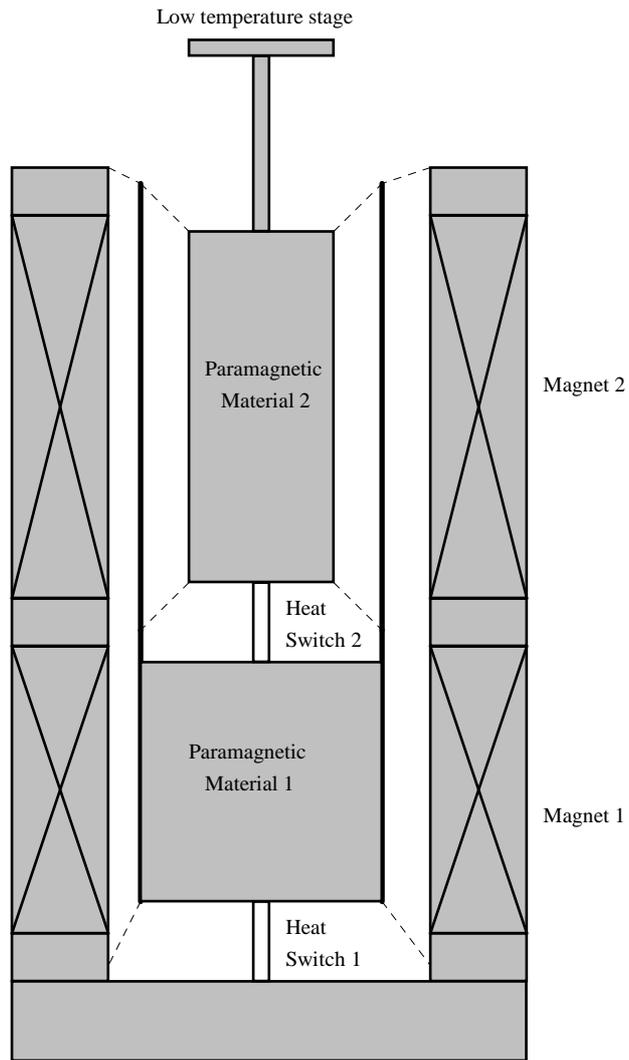
**Figure 4 Schematic of a two stage ADR**

(Heat Switch not shown)

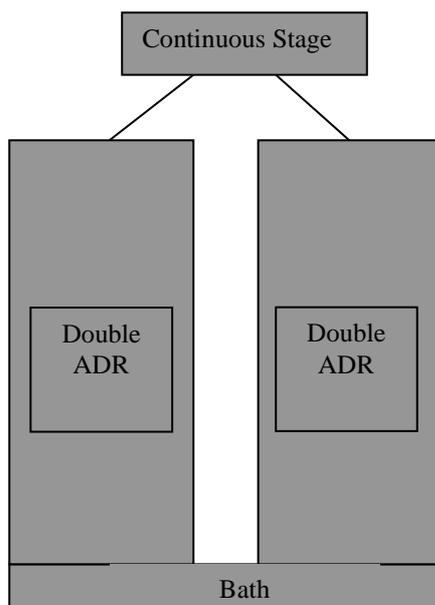
A= Low temperature paramagnetic material

B= High temperature paramagnetic material

C= Magnet, D=High thermal link, E=Kevlar cords



**Figure 5 Schematic of Double ADR**



**Figure 6 Continuous ADR**

## References

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- <sup>v</sup> Kurti N. and Simon, F. E. (1934). *Nature (London)* **133**, 907
- <sup>vi</sup> Hudson, R.P. "Principles and Applications of Magnetic Cooling" 1972 North-Holland series in Low Temperature Physics Vol 2. ISBN 0 7204 1257 9
- <sup>vii</sup> Lounasmaa, O. V., "Experimental Principles and Methods below 1K", 1974, Academic Press, Inc. ISBN 0-12-455950-6
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