Approach to Absolute Zero

4. Below 10 milli-Kelvin

R Srinivasan

R Srinivasan is a Visiting Professor at the Raman Research Institute after retiring as Director of the Inter-University Consortium for DAE Facilities at Indore. Earlier, he was at the Physics Department of IIT, Chennai for many years. He has worked in low temperature physics and superconductivity.

The previous articles of this series were:
1. Liquefaction of gases, December 1996.
2. From 4.22 K to 0.3 K, February 1997.
3. 0.3 K to a few milli-Kelvin, June 1997.

In Part 3 of the series a description of the dilution refrigerator was given. With this one may conveniently attain and maintain a temperature around 10 to 20 milli-Kelvin. In this concluding part of the series a brief discussion is given of the Pomeranchuk cooling and adiabatic nuclear demagnetisation to achieve still lower temperatures.

Introduction

In Part 3 of this series the principles of operation of the dilution refrigerator were described. The dilution refrigerator is used routinely to attain and maintain temperatures around 10 mK. To go to still lower temperatures one has to use one of the two techniques, namely (a) Pomeranchuk cooling and (b) adiabatic nuclear demagnetisation. These will be described briefly below.

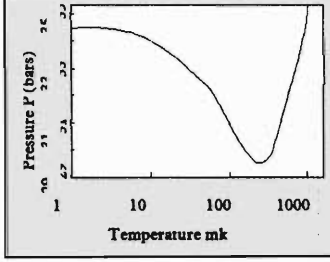
Pomeranchuk Cooling

The melting curve of ^{3}He is shown in *Figure 1*. Below 0.3 K the slope of the melting curve of ^{3}He is negative. The melting temperature decreases with an increase in pressure. The Clausius-Clapeyron equation for the melting line is given by

$$dP/dT = L/\{T(v_L - v_S)\}, \quad (1)$$

where L is the latent heat of melting and v_L and v_S are the specific volumes in the liquid and solid phases. The negative slope of the melting line below 0.3 K in ^{3}He arises because the entropy in the solid phase is larger than the entropy in the liquid phase. So the latent heat of melting is negative, i.e. when the liquid freezes to form a solid, *heat is absorbed.*

Figure 1. Melting curve of ^{3}He. Below 0.3 K slope is negative.



The nuclear spin of ^{3}He is 1/2. There are two possible spin orientations. At sufficiently high temperatures the two orientations are equally probable. The entropy per atom in this case is

$$S = k_B \ln 2 , \qquad (2)$$

k_B is the Boltzmann constant. At the temperatures we are considering ($T < 0.3$ K) the dominant contribution to the entropy comes from spin disorder.

Below 0.3 K solid ^{3}He has a negative latent heat of vaporisation.

According to the third law of thermodynamics, when the temperature tends to absolute zero, this entropy should tend to zero.

Liquid ^{3}He is a Fermi liquid. At absolute zero, each energy level up to the Fermi level will be occupied by a pair of quasi particles with opposite spins. At a finite low temperature (low compared to the Fermi temperature) the contribution to entropy comes from a few quasi-particle excitations above the Fermi level. The entropy is low because of the ordering of the spins in momentum space.

In solid ^{3}He, on the other hand, the atoms are localised at their lattice positions and execute vibrations about these mean positions. Above a temperature of 10 mK, the interaction between the atoms can be neglected and the atoms can be treated as independent particles. Because the particles are localised they become distinguishable and they obey classical statistics. They will show the full spin disorder entropy given by (1) above this temperature. But below this temperature the atoms populate preferentially the lower energy levels of the degenerate ground state split by the exchange interactions between the atoms. This causes a decrease in entropy. *Figure 2* shows the temperature dependence of the entropy of the solid and liquid phases.

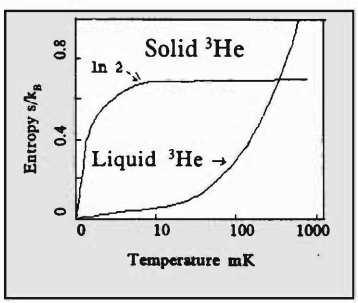


Figure 2. Temperature dependence of entropy per atom, S/k_B of solid and liquid ^{3}He. The entropy of the solid is higher than the liquid. Hence latent heat of melting is negative.

If a mixture of liquid and solid ^{3}He is compressed isentropically below 0.3 K, part of the liquid turns into solid absorbing heat

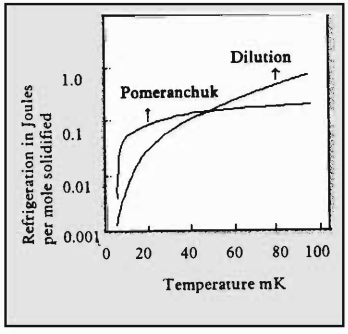


Figure 3. Refrigeration capacity of a Pomeranchuk and a dilution refrigerator as a function of temperature.

and the temperature of the liquid falls. This is the principle of Pomeranchuk cooling. If we start with the mixture at an initial temperature T_i (>10 mK), and cool it to a final temperature T_f, the fraction of the liquid solidified is given by

$$f = \{S_{\text{liq}}(T_i) - S_{\text{liq}}(T_f)\}/\{S_{\text{sol}}(T_f) - S_{\text{liq}}(T_f)\}. \quad (3)$$

The fraction of the liquid solidified is small, of the order of 10 to 20% depending on T_i and T_f. *Figure 3* compares the refrigeration capacity as a function of temperature for a Pomeranchuk refrigerator and a dilution refrigerator. The superiority of Pomeranchuk cooling over the dilution refrigerator below 10 mK is apparent.

If the liquid is compressed isentropically, a part of it turns to a solid with a decrease in temperature of the mixture of solid and liquid ^{3}He.

The disadvantage of the Pomeranchuk method is that one can work only along the melting line. A schematic diagram of a Pomeranchuk cooling cell is shown in Figure 4. Note that by applying pressure on the liquid ^{4}He in the mixing chamber,

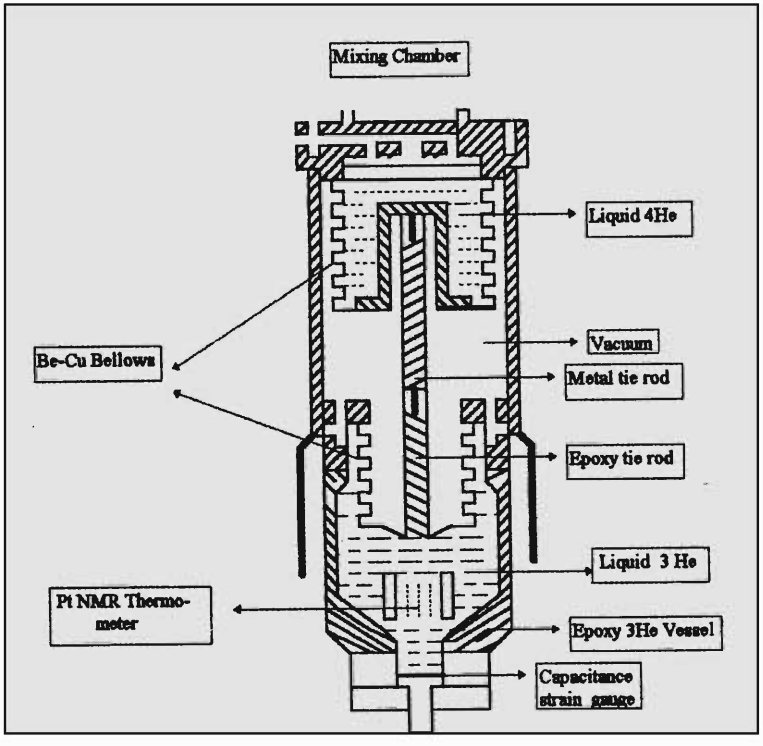


Figure 4. Pomeranchuk cell built at Cornell University. Pressure is applied through two bellows connected with a tie rod. The bellows have different cross-sections leading to a pressure amplification. (From F Pobell, see Suggested Reading).

pressure will be transmitted to the lower liquid ^{3}He cell in which Pomeranchuk cooling takes place. The pressure transmission is achieved by two beryllium-copper bellows connected by a tie rod. By choosing the ratio of the diameters of the two bellows, it will be possible to achieve pressure amplification. Since liquid ^{4}He has a freezing pressure lower than liquid ^{3}He such a pressure amplification is necessary. It was with such a cell that the Cornell group was able to reach a temperature down to 2 mK and discover the superfluid transition in liquid ^{3}He.

In a Pomeranchuk cell a mixture of solid and liquid ^{3}He is compressed by a bellows arrangement.

Adiabatic Nuclear Demagnetisation

The principle of adiabatic demagnetisation of a paramagnetic salt to achieve low temperatures was first proposed by Debye and Giauque in 1926 and experimentally realised in 1933.

A free magnetic atom in a ground state J has $(2J+1)$ magnetic states which are degenerate. Such an atom will contribute an entropy

$$S_M = k_B \ln(2J+1) \qquad (4)$$

which is independent of temperature. However in a paramagnetic crystal containing the magnetic atom, because of the electric field of neighbouring ions at the site of the magnetic atom, the $(2J+1)$ fold degeneracy of the energy level of the atom will be partially removed, with the resulting energy levels differing by small amounts. Because of this crystal field splitting of energy levels, the entropy, which will have a value given by (4) at high temperatures ($k_B T >>$ spacing between the energy levels), will start decreasing as the temperature is reduced.

If a magnetic field B, is applied to the crystal, the energy sub-levels, $M_J = J, (J-1), \ldots\ldots -(J-1), -J$ will suffer Zeeman shifts in energy given by

$$\Delta\varepsilon(M_J) = -g_J \mu_B M_J B \qquad (5)$$

A paramagnetic salt placed in a magnetic field has a lower entropy than the salt in zero field.

Isothermal magnetisation of a paramagnetic salt releases heat. If the salt is now adiabatically demagnetised, its temperature will fall.

Here g_J is a constant called the *Lande splitting factor* and μ_B is the *Bohr magneton* ($eh/4\pi mc$), which has the value 9.27×10^{-24} J/Tesla.

For large B and low temperatures, the lower of these Zeeman shifted levels will be more populated than the higher Zeeman shifted energy levels. This imposes an order on the orientation of the magnetic spins and reduces the entropy (i.e.)

$$S_M(B,T) < S_M(0,T). \qquad (6)$$

This is shown in *Figure 5*.

If now we start with the salt at an initial temperature T_i, in zero magnetic field, and apply a magnetic field *isothermally*, the entropy of the crystal will decrease. The path followed by the crystal in the T-S diagram is along AB and the heat released, $Q = T_i \Delta S$, will be absorbed by the heat reservoir in contact with the crystal. The salt is thermally *isolated* from the heat reservoir and the magnetic field is reduced to B_f *isentropically*. The path followed by the crystal in the T-S diagram is along BC and we note that the temperature will fall. This is the principle of adiabatic demagnetisation of a paramagnetic salt. The lowest temperature that one can attain is set by the magnetic ordering temperature of the atomic magnetic moments in the crystalline salts due to the exchange interaction. The final temperature is given by

$$T_f / T_i = (B_f^2 + b^2)^{0.5} / B_i , \qquad (7)$$

B_i and B_f being the initial and final values of the magnetic field and b the internal field due to the exchange interaction. The salt will warm up from the final temperature T_f absorbing an amount of heat

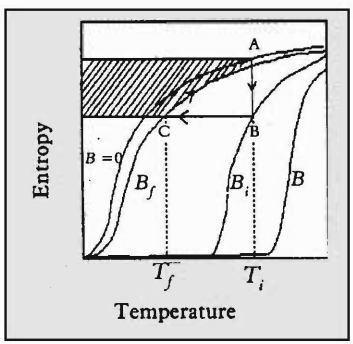


Figure 5. Schematic diagram of the entropy as a function of temperature for a magnetic material in different magnetic fields. Process of adiabatic demagnetisation is represented by the steps A → B → C. Subsequent warm-up is shown along the entropy curve from C by the arrow.

$$Q_a = \int_{T_f}^{\infty} T\,\mathrm{d}S. \qquad (8)$$

This is shown by the shaded area in *Figure 5*. The larger the value of B_f the larger the heat absorbed; but the value of the final temperature will be higher.

Before the invention of the dilution refrigerator, this was the only technique available to reach temperatures below 0.3 K. Starting with an initial temperature of 1.2 K achieved by pumping on a liquid ^{4}He bath, one could achieve temperatures of the order of 0.1 K by adiabatic demagnetisation of a suitable paramagnetic salt. With the discovery of dilution refrigeration, continuous refrigeration down to 10 mK became available. Using this as the starting temperature and choosing a suitable paramagnetic salt one can achieve temperatures down to about 2 mK.

The nucleus of an atom has also a magnetic moment, though its magnitude is nearly a thousand times smaller than the electronic magnetic moment μ_B It was proposed by Gorter in 1934 that adiabatic demagnetisation of nuclear magnetic moments can be used for reaching temperatures in the micro-Kelvin region.

Let us consider the typical example of copper. It has a nuclear spin of 3/2 and so the nuclear spin entropy is k_B ln4 for each nucleus above a temperature of 10 mK. At this temperature the electronic entropy is very small compared to the nuclear entropy. Since the nuclear magnetic moment is very small we must start with a high magnetic field (of the order of 8 T) so that an appreciable population difference between successive magnetic

The nuclear magnetic moments are a thousand times smaller than the electronic magnetic moments. By demagnetising the nuclear moments starting with a ternperature around 10 mK one may reach nuclear spin temperatures in the micro-Kelvin range.

levels can be built up at such low temperatures. Generally one starts with a high temperature (about 20 mK), gradually increases the field to 8 T and cools the copper sample to the initial temperature. The initial cooling is not isothermal. Though this implies a greater amount of heat to be removed at high temperature for a given T_i, it has practical advantages. When the field is isentropically reduced to the final value B_f, the temperature of the nuclear spins decreases to a few micro-Kelvin.

One must remember that we have three systems here: (a) the system of nuclear spins, which reach thermal equilibrium among themselves in a time τ_σ, called the *spin-spin relaxation* time. This is of the order of a few milli-seconds. (b) The second system is that of the conduction electrons. These conduction electrons interact with the nuclear spins through the hyperfine interaction. (c) The third system is the lattice. The electrons and the lattice are tightly coupled through the electron-phonon coupling and so the two systems are at the same temperature. The lattice does not directly couple to the nuclear spins, but only through the electrons. The spin-lattice relaxation time τ_1 is a measure of the rate of exchange of energy between the electron system and the spin system. At low temperatures it varies inversely with the electron temperature and can be several seconds at temperatures below 1 mK. So the system of nuclear spins will be at a lower temperature than the system of electrons (and hence the lattice) after adiabatic nuclear demagnetisation. The temperatures of the two systems will approach each other. The actual rate at which the electron temperature falls will be

While the electrons and lattice rapidly equilibrate thermally, the exchange of energy with the nuclear system is relatively slower. Since the electronic specific heat is much smaller than nuclear specific heat, the electronic system is still cooled fairly rapidly by the nuclear spins.

determined by a time constant τ given by

$$\tau = \tau_1 C_e / C_n \qquad (9)$$

Here C_e and C_n are the electronic and nuclear heat capacities. Since the electronic heat capacity is very small compared to the nuclear heat capacity, the effective relaxation time is very much shorter than the spin lattice relaxation time τ_1. So the electronic system cools rapidly.

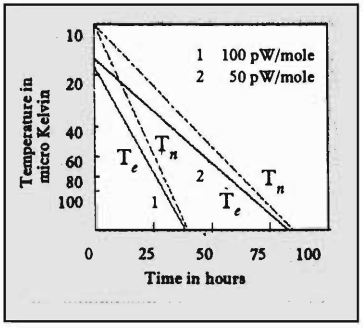


Figure 6. Warm up after adiabatic nuclear demagnetisation as a function of time for the electrons and nuclear spins for two different heat loads.

However both the electronic and nuclear systems will warm up as the heat leak into the system increases. *Figure 6* shows a plot of the warm up as a function of time for the electronic and nuclear temperatures of copper for different heat leaks into the system. It is clear from the figure that the larger the heat leak (a) the difference between the electronic and nuclear spin temperatures is more and (b) the two systems warm up more rapidly. In order to maintain the low temperature steady for several hours the heat leak into the system must be less than about 20 picowatts per mole of copper.

Figure 7 shows a copper adiabatic nuclear demagnetisation set up with a superconducting magnet for producing the magnetic field. The copper is in the form of thin wires to reduce eddy current heating in a changing magnetic field. The copper wires are cooled by a dilution refrigerator to an initial temperature of about 10 mK. The superconducting heat switch is a superconducting wire connecting the mixing chamber of the dilution refrigerator and the copper wires. By applying a magnetic field this wire can be driven normal. In the normal state it has a high thermal conductivity. Once the initial magnetic field B_i is applied, one waits for a sufficiently long time till the copper wires cool to the initial temperature T_i by exchanging heat with the mixing chamber through the heat switch. Then the magnetic field in the heat switch is turned off. The heat switch wire becomes superconducting. In this state it is a poor thermal conductor. The magnetic field B on the copper wires is slowly

A lattice temperature of 12 μK has been reached in copper using adiabatic nuclear demagnetisation.

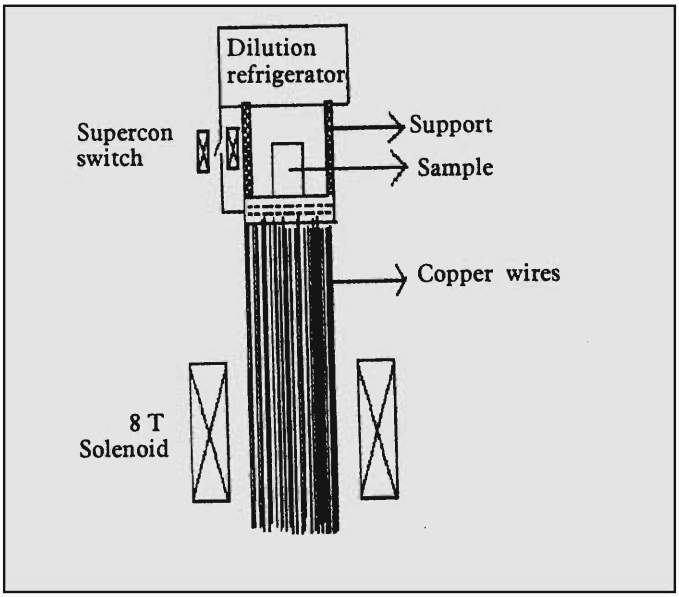


Figure 7. A schematic diagram of an adiabatic nuclear demagnetisation set-up using copper wires. (Adapted from F Pobell, see Suggested Reading).

reduced to the final value. The specimen on which experiments have to be done is placed away from the magnetic field.

The scope of this article does not permit a more detailed discussion of adiabatic nuclear demagnetisation. It is sufficient to state that using 2 moles of copper a lattice temperature of 12 μK was achieved.

Conclusion

It should be mentioned here that in July 1995 a cloud of about 2000 atoms of Rubidium was cooled to a temperature of 20 nK (1 nK = 10^{-9} K) in the vapour state by an entirely novel cooling process using laser and evaporative cooling. This will be the closest to absolute zero anyone has reached so far.

In this series of articles an attempt was made to present the efforts spanning nearly seven decades to reach as close to absolute zero as possible. It was not easy to present the principles at a level which would be easily understood by undergraduate students without sacrificing scientific rigour. It is hoped that at least a few readers were benefitted.

Suggested Reading

- O V Lounasmaa. *Experimental Principles and Methods below 1K.* Academic Press. London, 1974.
- F Pobell. *Matter and methods at low temperatures.* Springer Verlag. Berlin, 1992.

Address for Correspondence
R Srinivasan
Raman Research Institute
C V Raman Avenue
Sadashivanagar
Bangalore 560 080, India
email: rsv@rri.ernet.in