

Approach to Absolute Zero

3. 0.3 K to a Few Milli-Kelvin

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In part 2 of this series we dealt with the ^3He cryostat. By pumping on liquid ^3He bath a temperature down to 0.3 K can be attained. To reach still lower temperatures in the milli-Kelvin region one uses a dilution refrigerator. In this part the principles of operation of a dilution refrigerator will be described.

Introduction

The ^3He cryostat was described in part 2 of this series of articles. By reducing the pressure on the ^3He bath one can reach a temperature down to about 0.3K. To reach still lower temperatures Debye and Giauque suggested the adiabatic demagnetization of a paramagnetic salt. This one-shot technique was used till the development of the dilution refrigerator in the late sixties and early seventies. The dilution refrigerator has the advantage that it provides continuous refrigeration down to about 10 milli-Kelvin.

The development of the dilution refrigerator had to await the availability of the rare isotope, ^3He , of helium in sufficiently large quantities. This became possible around the early sixties. As mentioned in part 2, ^3He is a Fermion while the more abundant isotope ^4He is a Boson. Investigations were undertaken to study the mixture of ^3He and ^4He to see the influence of the concentration of ^3He on the superfluid transition in ^4He . These studies led to the development of the dilution refrigerator.

Phase Diagram of a Mixture of ^3He in ^4He

Figure 1 shows the phase diagram of a mixture of ^3He and ^4He as a function of the atomic concentration x of ^3He in the mixture.

The previous articles of this series were:

1. Liquefaction of gases, December 1996.
2. Approach to absolute zero, February 1997.

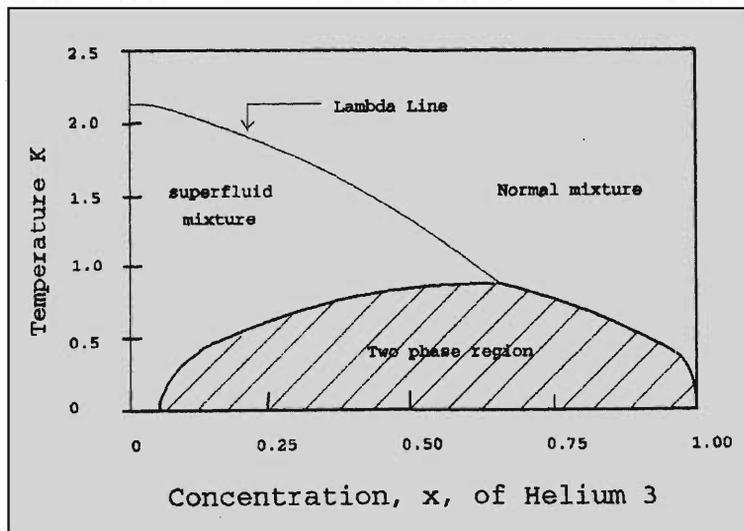


Figure 1 The phase diagram of mixtures of ^3He and ^4He as a function of temperature and concentration x of ^3He .

One sees from this diagram that for a concentration x below 0.67, the mixture first becomes a superfluid as the temperature is lowered. The superfluid transition temperature of the mixture decreases as the concentration of ^3He increases. On further cooling, the mixture separates into two phases, phase A rich in ^3He which floats on phase B rich in ^4He . As the temperature is lowered, phase A becomes almost 100% ^3He . In the phase B, the concentration of ^3He decreases as the temperature is lowered and appears to approach a constant value of 6.5%. If the initial concentration of ^3He is more than 0.67, no superfluid transition is seen. When the mixture is cooled from the normal phase, it reaches the two phase boundary at which point a phase separation into the A and B phases is seen.

A finite concentration of ^3He in phase B as the temperature approaches absolute zero might appear to be a violation of the third law of thermodynamics according to which the entropy of phases A and B should tend to zero as the temperature tends to absolute zero. If the mixture obeyed classical statistics, then a zero entropy will be achieved at absolute zero only if the mixture separates into two pure phases, one consisting only of ^4He and the other only of ^3He . However this does not happen in mixtures of ^3He and ^4He . This is because in the dilute phase one may treat the ^3He atoms as forming an interacting gas of Fermions

A mixture of ^3He and ^4He , cooled to a temperature below 0.1 K shows phase separation into a concentrated phase of ^3He floating on a phase dilute in ^3He . Even at absolute zero of temperature the dilute phase will contain 6.5% of ^3He .



For any given number density of the Fermions, the maximum kinetic energy to which the levels are completely filled at absolute zero of temperature is called the Fermi level.

in a neutral background of the ^4He condensate. The ^3He atoms obey quantum statistics of a degenerate Fermi gas (in which the occupancy of each state is close to one or zero). Since ^3He obeys Fermi-Dirac statistics, at absolute zero of temperature, the state with all energy levels filled up to the Fermi energy represents the ground state of the system with zero entropy. Thus there is no violation of the third law of thermodynamics.

Why is there a finite concentration of ^3He in phase B? This can be understood as follows. Due to the Heisenberg uncertainty relation, position and momentum of a particle cannot simultaneously assume precise values. If an atom is at rest at a point, it will violate the uncertainty principle. Even at the absolute zero of temperature there will be a zero point motion. A ^3He atom is lighter than a ^4He atom. It has a larger zero point motion than the heavier ^4He atom. Therefore a ^3He atom can come closer to a ^4He atom than to another ^3He atom. The Van der Waals interaction energy, which is attractive, (and therefore negative), increases in magnitude as the distance between two atoms decreases. So a ^3He atom will have a higher binding energy in the dilute phase B than in the concentrated phase A. In Fermi-Dirac statistics, only one particle can occupy a given quantum state. A quantum state is not only characterised in this case by the kinetic energy of the atom, but also by the spin of the atom. Since kinetic energy is independent of spin, and the nuclear spin of ^3He is $1/2$, a given kinetic energy state can be occupied by two ^3He atoms of opposite spin. If we add more and more ^3He atoms to the dilute phase, the ^3He atoms will have to go into states with larger and larger kinetic energy. For any given number density of the Fermions, the maximum kinetic energy to which the levels are completely filled at absolute zero of temperature is called the Fermi level, E_F . One can define an equivalent temperature T_F by the relation

$$kT_F = E_F$$

Here k is the Boltzmann constant. The chemical potential is the energy required to remove an atom from a given phase. As more

and more ^3He atoms are added to the dilute phase its chemical potential decreases and ultimately one reaches a limiting concentration at which the chemical potential of a ^3He atom in phase B is the same as the chemical potential of a ^3He atom in the concentrated phase A. When the chemical potential in the concentrated phase becomes equal to the chemical potential in the dilute phase, the two phases will be in equilibrium. This happens when the dilute phase has a concentration of 6.5% ^3He .

The chemical potential is the energy required to remove an atom from a given phase.

Heat of Mixing and Cooling Power of a Dilution Refrigerator

At temperatures below 0.5 K, the ^4He liquid, which is in the Bose condensed state, has a negligible specific heat. On the other hand the ^3He atoms in the dilute phase form an interacting Fermi gas. For such a gas, as we have seen above, one can define a Fermi temperature T_F which depends on the $1/3$ power of the number of Fermions per unit volume. The specific heat per atom of such a gas is given by

$$c = (\pi^2/2) (T/T_F) k \quad \text{for } T \ll T_F$$

and

$$c = (5/2) k \quad \text{for } T \gg T_F$$

At temperatures below 0.1 K the specific heat of liquid ^3He is linearly dependent on temperature, while, at a temperature above 1 K, it is independent of temperature. Since our interest is in temperatures below 0.1 K the first expression is valid. For the dilute phase ($x = 0.065$) the specific heat is given by

$$C_d = 106 T \text{ (J/mol } ^3\text{He K)}.$$

For the concentrated ($x = 1$) phase the specific heat is given by

$$C_c = 22 T \text{ (J/mol } ^3\text{He K)}.$$

The smaller value for the concentrated phase is because of the larger value of T_F . The entropy S is given by



$$S = \int_0^T (C/T) dT$$

The difference in enthalpy of the dilute phase and the concentrated phase is (since the chemical potential $\mu = H - TS$ is the same in both phases)

$$\Delta H = H_d(T) - H_c(T) = 84 T^2 \text{ Joule/mole } ^3\text{He.}$$

When ^3He atoms pass from the concentrated to the dilute phase they absorb energy. If the system is thermally isolated this absorption will cause cooling.

Thus the enthalpy in the dilute phase is more than the enthalpy in the concentrated phase. This difference in enthalpy is called the heat of mixing. This is analogous to the enthalpy being more in vapour phase than in the liquid phase of a gas. When we convert in one second n moles of a liquid to its vapour phase by pumping on it under conditions in which the system is thermally isolated, the liquid cools. In a similar fashion if n moles of ^3He are transferred from the concentrated to the dilute phase, when the system is thermally isolated, the system will cool.

In both cases the cooling power is given by

$$Q = n \Delta H$$

For the liquid to vapour transition ΔH is the latent heat of vaporization, which is weakly dependent on temperature. n is proportional to

$$n \propto S p(T)$$

where S is the pumping speed of the pump in litres/second and $p(T)$ is the vapour pressure of the liquid as a function of temperature. Because $p(T)$ decreases exponentially with decreasing temperature, the cooling power achievable by pumping on a liquid bath decreases exponentially as the temperature is decreased. On the other hand for the dilution



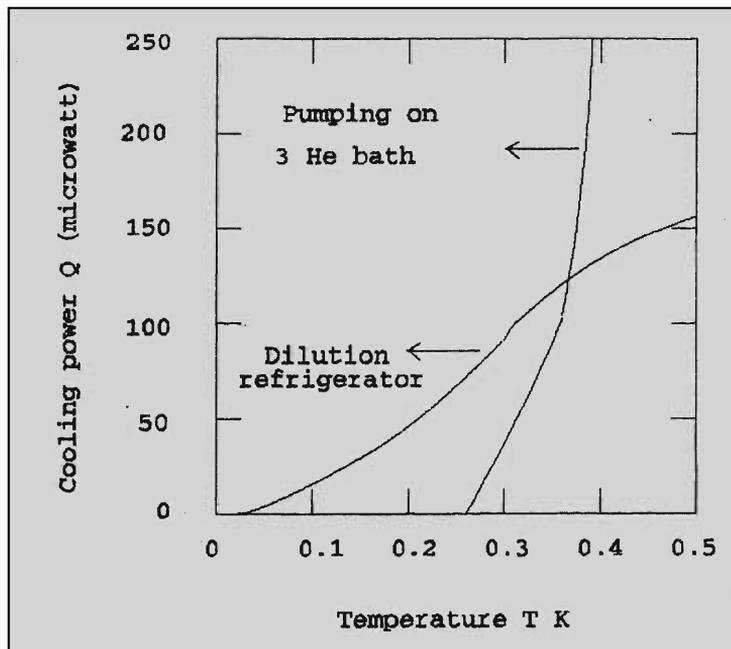


Figure 2 Cooling power as a function of temperature of a dilution refrigerator and a pumped liquid ^3He bath.

refrigerator, which involves the flow of ^3He from the concentrated to the dilute phase across the phase boundary, ΔH is proportional to T^2 . Since, even at absolute zero, there is a finite concentration of ^3He (of 6.5%) in the dilute solution, the number of moles of ^3He crossing the boundary will not decrease significantly as the temperature is reduced. The cooling power therefore decreases as T^2 . Figure 2 shows the cooling power of a dilution refrigerator in comparison to that with a pumped ^3He bath. One sees that the dilution refrigerator provides several microwatts of power down to a few millikelvin.

Because the dilute phase contains almost a constant fraction of 6.5% of ^3He and the heat of mixing is proportional to T^2 the cooling power of a dilution refrigerator falls as T^2 .

Fractional Distillation of ^3He

In the mixing chamber the concentrated phase is floating on the dilute phase. In order to achieve refrigeration one has to remove ^3He from the dilute phase. Then ^3He will cross from the concentrated phase to the dilute phase and this will cause cooling. This is done by having a distillation still connected to the bottom of the mixing chamber. This still will be at a

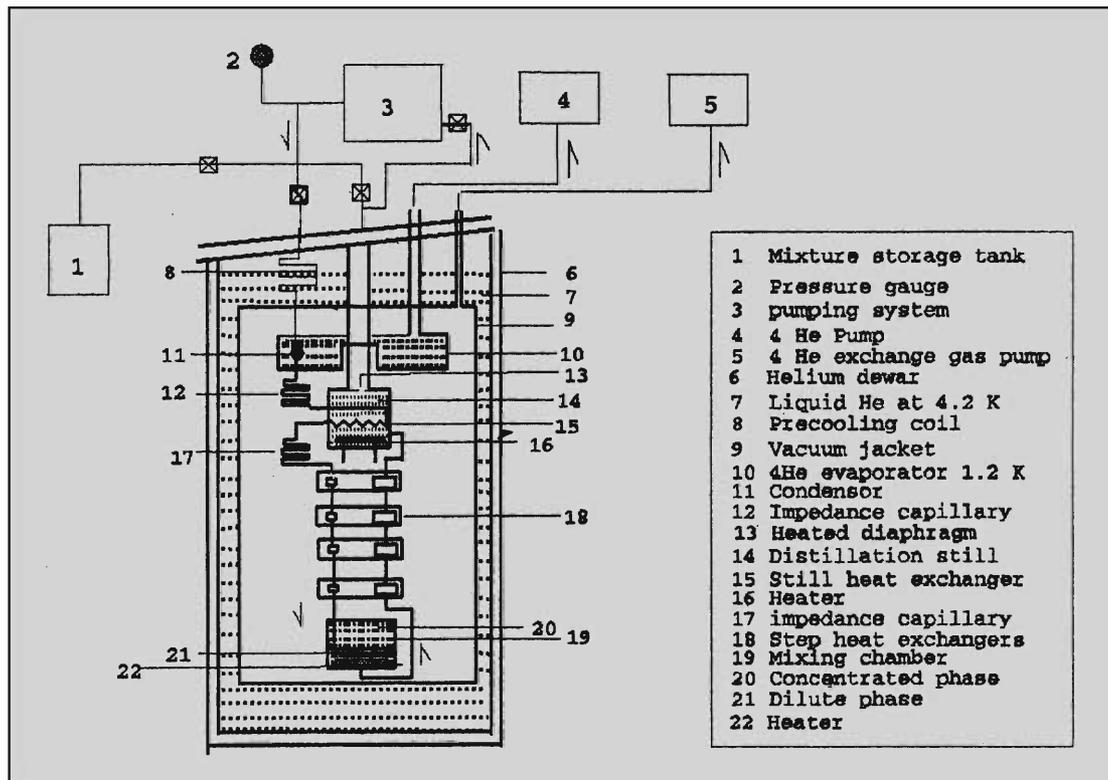
The ^3He atoms move from the mixing chamber to the distillation still by osmotic pressure difference. In the still the ^3He atoms are selectively evaporated by the process of fractional distillation.

temperature around 0.65 K. At this temperature the vapour pressure of ^3He is about a hundred times more than the vapour pressure of ^4He . So when heat is supplied to the still to distil the mixture, the vapour consists almost of pure ^3He atoms. Because of this fractional distillation of ^3He from the still, the concentration x in the liquid mixture in the still is below 0.1%.

How is the ^3He driven from the mixing chamber to the still? Since the concentration x of ^3He and the temperature are different in the dilute phase in the mixing chamber and in the still, there is an osmotic pressure difference between the two. The osmotic pressure in the mixture depends on the product of the concentration and temperature. If the temperature of the mixing chamber is taken to be 10 mK and the temperature of the still to be 0.65 K, then the osmotic pressure difference is zero when the concentration of the liquid mixture in the still is 0.1%. Because ^3He is being continuously removed from the still by fractional distillation, the concentration of ^3He in the dilute mixture in the still falls below the value of 0.1%. Then the dilute phase in the mixing chamber is at a higher osmotic pressure than the mixture in the still. This pressure difference drives the ^3He from the mixing chamber to the still. Thus continuous refrigeration is provided by the circulation of ^3He .

A Schematic Diagram of a Dilution Refrigerator

Figure 3 shows a schematic diagram of a ^3He - ^4He dilution refrigerator. The mixture of ^3He and ^4He from a storage tank (1) is passed through a precooling coil (8) immersed in a liquid ^4He bath (7) at 4.2 K. The cold mixture then condenses in the condenser (11) kept in a pumped liquid ^4He bath at 1.2 K. The condensed mixture then passes through a capillary impedance (12) into a heat exchanger (15) immersed in the dilute mixture in the distillation still (14). The temperature of this still is around 0.65 K. The cold liquid mixture now passes through another capillary impedance tube (17) into a series of four step heat exchangers. The mixture finally reaches the temperature of



the mixing chamber (19) and enters the mixing chamber in a phase separated state, with the concentrated phase (20) of almost pure liquid ^3He floating on the dilute phase (21) (6.5% ^3He). From here the ^3He is pumped by osmotic pressure to the distillation still through the step heat exchangers. The heater (16) in the still helps to fractionally distil ^3He from the liquid mixture in the still. The evaporating ^3He vapour is then pumped by the pumping system (3) and fed back into the pre-cooling coil (8) thus forming a closed loop. The pumping system (4) is used to pump on the liquid ^4He bath (7) to lower its temperature. The pump (5) is used for filling or removing ^4He exchange gas in the vacuum jacket. The system of valves shown allows one to pump the mixture of ^3He and ^4He back into the storage vessel (1) at the conclusion of an experiment. The experimental system can be attached to the bottom of the mixing chamber. The temperature of the mixing chamber can be varied using the heater (22).

Figure 3 A schematic diagram of a dilution refrigerator (adapted from Pobell's book.).

The design of the step heat exchangers is crucial to attain very low temperatures.

The design of the step heat exchanger is crucial. At such low temperatures there is a boundary resistance between the liquid mixture and the wall of the heat exchanging tube. This resistance to heat transfer is called Kapitza resistance. This resistance increases rapidly as the temperature is reduced. To achieve good heat transfer at such low temperatures, overcoming the Kapitza resistance requires a large heat transfer area. The step heat exchanger is made out of a copper block in which two holes are drilled for the flow of the warm and cold liquids. Each channel is filled with compressed and sintered metal powder to increase the surface area for heat transfer.

The dilution refrigerator has become the workhorse for investigations down to about 10 mK. In India a dilution refrigerator with which experiments can be done down to about 20 mK is available at the Tata Institute of Fundamental Research in Mumbai.

The dilution refrigerator in conjunction with the Pomeranchuk cell made it possible to cool sufficiently large quantities of liquid ^3He down to 2 mK leading to the discovery of superfluidity in this liquid. Adiabatic nuclear demagnetization coupled with the dilution refrigerator makes it possible to reach temperatures in the sub-milli Kelvin region. These two techniques will be briefly dealt with in the concluding part 4 of this series article.

Suggested Reading

- ◆ O V Lounasmaa. *Experimental Principles and Methods below 1 K*. Academic Press. London, 1974.
- ◆ D S Betts. *An Introduction to Millikelvin Technology*. Cambridge University Press. Cambridge, 1989.
- ◆ F Pobell. *Matter and Methods at Low Temperatures*. Springer Verlag. Berlin, 1992.

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