Continuous cooling in the millikelvin range

F. A. Staas

The cooling of a system to very low temperatures is a means of investigating weak interactions that would be completely swamped by other effects at higher temperatures. In the temperature range of about 1 to 10 K, superconductivity and superfluidity are among the striking effects that arise because of the dominance of certain interactions. There are quite a number of interesting interactions, however, such as magnetic interactions between nuclear spins, that only become dominant in the millikelvin range. As long ago as the thirties the millikelvin range was reached by means of adiabatic demagnetization, but this method has the disadvantage that the cooling process is a 'single-shot' method. Continuous cooling in this range first became feasible with the advent of the 'dilution refrigerator' in the sixties. The dilution refrigerator described in this article is capable of extracting about ten times more heat per second from the environment than earlier types.

Low-temperature research would nowadays scarcely be conceivable without liquid helium as a 'cold bath'. Under normal pressure helium boils at 4.2 K. In 1908, on the very same day that he became the first to liquefy helium, Kamerlingh Onnes obtained a temperature of 1 K by vigorously pumping away the helium vapour above the liquid (the experiment was in fact part of an abortive attempt to solidify the helium).

Temperatures much lower than 1 K cannot be reached in this way, because the cooling capacity then approaches zero. This quantity is determined by the amount of helium vapour that can be removed per second, in other words it depends on the pumping capacity and the vapour pressure. At 1 K the vapour pressure of helium is still 16 Pa (0.12 mm Hg), but at 0.5 K it is no more than 2.1 x 10^{-8} Pa (1.6 x 10^{-5} mm Hg). In 1932 W. H. Keesom obtained a temperature of 0.71 K (vapour pressure 0.5 Pa) with a pumping capacity of 675 l/s. These figures relate to naturally occurring helium, which consists almost entirely of $^4$He. It is possible with the (very expensive) isotope $^3$He, which has a higher vapour pressure at the same temperature (boiling point 3.2 K), to reach a temperature of about 0.3 K by pumping.

The millikelvin range was opened up in the thirties by means of 'adiabatic demagnetization'. This method, which will not be dealt with here, has the disadvantage for the experimenter that the cooling process occurs once only (single-shot experiment). Continuous cooling in the millikelvin range did not in fact become possible until the sixties, with the advent of the dilution refrigerator discussed in this article.

The operation of the dilution refrigerator is based on some special properties of liquid mixtures of $^4$He and $^3$He. These are summarized in the phase diagram given in fig. 1, which shows the temperature $T$ plotted against the molar concentration $X$ of the $^3$He in the mix-
ture. On cooling, pure $^4$He ($X = 0$) changes at the ‘$\lambda$ point’ (2.17 K) from a normal liquid (‘He I’) into a liquid with superfluid properties (‘He II’). Above 0.88 K $^4$He and $^3$He are miscible in all proportions; the $\lambda$-temperature in this region decreases as $X$ increases. Below 0.88 K, however, the liquid separates to form two phases: a ‘dilute mixture’ ($d$), which has a low content of $^3$He, and a ‘concentrated mixture’ ($c$), with a high content of $^3$He.

The concentrated mixture is the lighter of the two and thus floats on the dilute mixture. The concentrated mixture behaves as a normal liquid. In the dilute solution, however, the $^4$He is superfluid, and the $^3$He atoms contained in it are not impeded in their movement; it is as if for these atoms the $^4$He does not exist. The $^4$He thus behaves like a kind of vacuum, and the $^3$He dissolved in it may be regarded as a gas.

This ‘gas’ has an exceptionally high density compared with that of a normal gas at the same temperature. In about 1960 the location of the part of the phase-separation line between 0.88 K and 0.4 K was known experimentally. It showed that at 0.5 K, for example, phase $c$ is in equilibrium with a phase $d$ that contains approximately 20% of $^4$He, i.e. $7 \times 10^{-9}$ mol/cm$^3$. This is more than a thousand times the density of normal $^3$He gas that is in equilibrium with liquid $^3$He at 0.5 K. Since these large quantities can indeed be removed selectively from the dilute solution, very effective cooling should be possible. This is the basic principle of the dilution refrigerator [1]. The heart of the refrigerator is the mixing chamber in fig. 2. When the $^3$He is removed from the dilute solution $d$, the $^3$He in $c$ goes over to $d$. Like evaporation, this mixing process takes up heat, since it is the fastest atoms from $c$ that have gone over to $d$. This heat is extracted from the chamber, which is therefore cooled.

The first dilution refrigerators [2] worked better than expected. This was because the solubility of $^3$He in $^4$He, extrapolated to $T = 0$, does not occur at $X = 0$, as had originally been thought, but at $X = 6.4\%$ [3] (see fig. 1). Unlike the situation with normal vapour, there remains in the dilute solution, even at the lowest temperatures, a considerable quantity of $^3$He which can be removed.

Dilution refrigerators are now widely used in laboratories all over the world. In most of them, $^3$He is circulated by means of a pump. The most important of the quantities that determine the cooling capacity is the circulation rate $\eta_3$, i.e. the number of moles of $^3$He passing through a given cross-section per second. There is also a version in which $^3$He is circulated instead of $^3$He [4]; in this type the $^4$He is conducted towards the phase-separation plane instead of $^3$He.

In this article we shall discuss a new type of dilution refrigerator, in which $^3$He and $^4$He both circulate. In this refrigerator we have been able to circumvent some of the typical limitations of the ‘conventional’ type employing $^3$He circulation. The result is a cooling capacity one or two orders of magnitude higher than in the conventional refrigerator. By way of introduction we shall first discuss the conventional refrigerator and its limitations, and then very briefly describe the refrigerator using only $^4$He circulation.

**The conventional dilution refrigerator**

The operation of the conventional dilution refrigerator is illustrated by the diagram in fig. 3. A $^3$He-$^4$He mixture is cooled to about 1.5 K and then cooled further by pumping. The composition is chosen in such a way that phase separation finally takes place in the mixing chamber $M$. A heater wire $H$ may be used to ensure that the temperature of the vaporization chamber (or ‘still’) $V$, which is connected to $M$ via the pipe $P_2$, does not fall below 0.7 K. At this temperature the partial vapour pressure of $^3$He above the mixture is still high enough (9 Pa = $7 \times 10^{-2}$ mm Hg) for the $^3$He to be effectively pumped away. The $^4$He evaporates to a much lower extent; the gas contains 98% of $^3$He. The $^3$He is thus selectively removed from the dilute solution ($d$), so that heat is extracted from the system in $M$ as described in the caption of fig. 2. Also because the concentrated

---

$^3$He (c) entering through the pipe $P_1$ via the heat exchanger $W$ is precooled by the liquid in the pipe $P_2$, the temperature in $M$ falls to 0.1 K or lower. The liquid in $P_2$ is preheated in $W$. The $^3$He evaporating in $V$ is returned at a pressure of $13 \times 10^3$ Pa (10 cm Hg) by an external pump to the chamber $C$, which is kept at 1.5 K by thermal contact with an evacuated helium bath, where the gas condenses. The concentrated $^3$He returns to $P_1$ through the flow resistance $W'$, which takes up the pressure difference of $13 \times 10^3$ Pa between $C$ and $P_1$ and is in thermal contact with $V$. The diagram in fig. 4 indicates the phases through which the liquid passes in going from $C$ to $V$.

The part of the system drawn in fig. 3 is contained in a vacuum. The only connections with the outside world are the pump leads, the thermal contact of $C$ with the helium bath, the heater $H$ and other measurement and control leads.

Fig. 3. A conventional dilution refrigerator. Cooling takes place in the mixing chamber $M$, where $^3$He of the concentrated mixture (c) transfers to the dilute solution (d). The $^4$He passes through the pipe $P_2$ and enters the vaporization chamber (or still) $V$, from which it is pumped away; the vapour consists almost entirely of $^3$He owing to the great difference in the vapour pressures of $^3$He and $^4$He. The $^3$He is compressed to about 10 cm Hg, condenses in the vessel $C$ (1.5 K) and returns to the mixing chamber via the heat exchanger and flow resistance $W'$ and the pipe $P_1$. $W$ heat exchanger. $S$ sample. $H$ heater.

Fig. 4. Phase diagram showing the phases which the liquid passes through between $C$ and $V$ in fig. 3. $M_0$ is the concentrated phase in $M$, $M_d$ the dilute phase.
were also to apply to $^3$He in $^4$He, circulation would eventually become impossible because the pressure of the $^3$He would fall to zero as the temperature in the mixing chamber decreased. In fact, however, the situation is much more favourable: even at $T = 0$ the osmotic pressure of the equilibrium solution of 6.4% $^3$He in $^4$He is still 2300 Pa (17 mm Hg). The pressure has this value because the $^3$He behaves like a Fermi-Dirac gas, as we shall now briefly explain.

In quantum-statistical terms, there are two classes of particles: bosons and fermions. $^4$He belongs to the first, $^3$He to the second. If Bose-Einstein statistics are applicable, each quantum state can be occupied by an arbitrary number of particles, whereas in Fermi-Dirac statistics a quantum state can be occupied by no more than one particle. As an illustration, fig. 7 shows the distribution at $T = 0$ for a number of particles over the quantum states in the momentum space of a free particle in two dimensions. The bosons (fig. 7a) are all in the lowest state (Bose-Einstein condensation), whereas the fermions (fig. 7b) fill a ‘sphere’ in momentum space. The bosons ($^4$He) need not be considered here. We merely note that superfluidity is usually attributed to Bose-Einstein condensation.

Even at $T = 0$, fermions therefore possess momentum and kinetic energy, and hence also exert a pressure at $T = 0$. This pressure is generally given by:

$$P = \frac{3}{2} U,$$

(2)

These were built by A. P. Severijns of these laboratories.
The lattice points are the quantum states. At Fig. 7. Momentum space for a free particle in two dimensions. particles (fermions) still possess an average momentum. This condense in the lowest state in the case of Bose-Einstein statistics where \( X \) occupy a 'sphere' of lowest states \( (a) \). We consider a volume of 1 cm\(^3\) and fill it from the experimental value (2300 Pa) must be attributed to a weak interaction between the \( 3\text{He} \) atoms. Consequently \( e \) is proportional to \( n^{2/3} \). Fig. 8 gives a plot of \( e \) as a function of \( n \). The total energy \( U \) is the sum of all the particle energies. For large \( n \) the quantity \( U \) can be expressed as an integral equal for \( n = n_0 \) to the cross-hatched area in fig. 8. The effective mass of \( 3\text{He} \) atoms in superfluid \( 4\text{He} \) is 2.34 times the true mass. Using this to calculate the integral to a limit \( n_0 \) corresponding to \( X = 6.4\% \), we then find from (2) a pressure of 2900 Pa (22 mm Hg) for the Fermi-Dirac gas. The discrepancy from the experimental value (2300 Pa) must be attributed to a weak interaction between the \( 3\text{He} \) atoms.

Fig. 9 demonstrates once again the practical importance for the dilution refrigerator of the fact that the \( 3\text{He} \) behaves like a Fermi-Dirac gas.

Finally, we shall briefly consider the solubility of \( 3\text{He} \) in \( 4\text{He} \). The volume of liquid \( 3\text{He} \) is 37 cm\(^3\) per mole, that of \( 4\text{He} \) is 28 cm\(^3\) per mole. Thus, \( 3\text{He} \) atoms occupy a greater volume in the liquid than \( 4\text{He} \) atoms. Theoretically this would be expected: the amplitude of the zero-point vibration, which each enclosed particle still performs at \( T = 0 \), is greater for the lighter \( 3\text{He} \) atoms. Fig. 10 pictures the situation for liquid \( 3\text{He} \); the difference in atomic volumes is greatly exaggerated. Owing to the attractive forces (Van der Waals forces) the particles tend to bunch as closely as possible together. A \( 4\text{He} \) atom will not tend to move towards the \( 3\text{He} \), where it would have fewer neighbours at a greater distance (measured from centre to centre). Conversely, a \( 3\text{He} \) atom will prefer to move towards the \( 4\text{He} \), since it will then have more neighbours at a shorter distance. On the other hand, the \( 4\text{He} \) atoms will have to move away from each other to admit a \( 3\text{He} \) atom. As confirmed by theoretical calculations it turns out that energy is gained upon such a transition, i.e. the thermodynamic potential \( \mu_e \) of \( 3\text{He} \) atoms in pure liquid \( 3\text{He} \) in relation to that in pure liquid \( 4\text{He} \) is positive. As the Fermi sphere fills up, however, the energy gain decreases with the increase in the number of \( 3\text{He} \) atoms in the \( 4\text{He} \), and equilibrium is reached at \( X = 6.4\% \). In fig. 8, \( \varepsilon \) may be regarded as the thermodynamic potential \( \mu_d \) of \( 3\text{He} \) atoms in a dilute solution with respect to that in pure \( 4\text{He} \). At equilibrium, \( \mu_d \) is equal to \( \mu_e \).

The energy gain on the transition of a \( 3\text{He} \) atom from pure \( 3\text{He} \) to pure \( 4\text{He} \) is in sharp contrast with the considerable energy loss, due to the Van der Waals forces in the liquid, that occurs on the transition of an atom from the liquid to true vacuum. It is of course the magnitude of this energy loss that accounts for the vapour pressure being so small at low temperature. In \( 4\text{He} \) the Van der Waals forces are stronger than in \( 3\text{He} \) and therefore the vapour pressure of \( 4\text{He} \) is lower at the same temperature, as we have already seen.
Cooling capacity and lowest attainable temperature of dilution refrigerators

Heat exchangers

One of the factors that limits the cooling capacity of the conventional dilution refrigerator is the heat exchanger $W$ in fig. 3. Why should this be?

The cooling takes place because the $^3$He atoms in the dilute solution possess more energy than those in the concentrated mixture. Furthermore, the $^3$He in the dilute solution in the pipe $P_2$ takes up additional heat from the concentrated $^3$He in $P_1$ because it expands ($P_{\text{osm}}$ is constant, $T$ increases and $X$ decreases; see fig. 9). The internal energies in the two phases are known from measurements of specific heats. Below 0.7 K the energy transported downwards through $P_1$ by the concentrated solution per mole of $^3$He is $\alpha T^2$, and the energy transported upwards by the diluted solution in $P_2$ is given by $\beta T^2$ per mole of $^3$He, where $\alpha$ is equal to 12 J/mol K$^2$ and $\beta$ to 94 J/mol K$^2$.

The energy-flow balance over one of the closed surfaces $a$, $b$ or $c$ in fig. 11 is therefore given by:

$$Q = n_3(\beta - \alpha)T_M^2 = n_3\beta T_M^2.$$  \hspace{1cm} (3)

$Q$ is the heat absorbed per second from the environment by the mixing chamber, i.e. the cooling capacity. $T_1$ and $T_2$ are the temperatures of the liquids in $P_1$ and $P_2$ at the level of the chosen cross-section.

A complete analysis leads to rather complicated calculations \cite{7}, but the following simple considerations give a qualitative picture of the situation. The temperature in the lower part of $P_2$ is equal to the temperature $T_M$ of the mixing chamber, $T_2(a) = T_M$. Let us now first assume that the heat exchanger is so good that the temperatures at the lower end are identical: $T_1(a) = T_2(a) = T_M$. From the balance over the area (a) it then follows that:

$$Q = n_3(\beta - \alpha)T_M^2 = 82 n_3 T_M^2.$$  \hspace{1cm} (4)

Fig. 9. Left-hand part of the phase diagram, with lines of constant osmotic pressure (solid lines), for a dilute solution which is in equilibrium with concentrated $^3$He at 50 mK and 10 mK. These lines are followed in the pipe $P_2$ of fig. 3 if the pressure drop in the pipe is negligible. If the $^3$He behaved as an ideal gas and not as a Fermi-Dirac gas, the dashed lines would be followed ($X_T$ constant, see eq. 1), and the concentration in the still would have been much smaller, i.e. $X'_1$ and $X'_2$ instead of $X_1$ and $X_2$. The vertical line ($X$ constant) from a concentrated mixture of 0.1 K relates to the new refrigerator (p. 111); this gives a much greater concentration ($X_3$) in the vaporization chamber.

Fig. 10. Simple representation of liquid $^3$He floating on liquid $^4$He. The difference in atomic volumes is greatly exaggerated.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig9}
\caption{Left-hand part of the phase diagram, with lines of constant osmotic pressure (solid lines), for a dilute solution which is in equilibrium with concentrated $^3$He at 50 mK and 10 mK. These lines are followed in the pipe $P_2$ of fig. 3 if the pressure drop in the pipe is negligible. If the $^3$He behaved as an ideal gas and not as a Fermi-Dirac gas, the dashed lines would be followed ($X_T$ constant, see eq. 1), and the concentration in the still would have been much smaller, i.e. $X'_1$ and $X'_2$ instead of $X_1$ and $X_2$. The vertical line ($X$ constant) from a concentrated mixture of 0.1 K relates to the new refrigerator (p. 111); this gives a much greater concentration ($X_3$) in the vaporization chamber.}
\end{figure}

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig10}
\caption{Simple representation of liquid $^3$He floating on liquid $^4$He. The difference in atomic volumes is greatly exaggerated.}
\end{figure}

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig11}
\caption{Lower part of the conventional dilution refrigerator, with closed surfaces $a$, $b$ and $c$ over which the energy balance is taken. $M$, $W$, $S$, $P_1$, $P_2$ are as in fig. 3.}
\end{figure}

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig12}
\caption{Upper part of the conventional dilution refrigerator, with closed surfaces $a$, $b$, $c$ and $d$ over which the energy balance is taken. $M$, $W$, $S$, $P_1$, $P_2$ are as in fig. 3.}
\end{figure}
Eq. (4) gives the cooling capacity $Q$ at a given $T_M$, or the lowest attainable temperature $T_M$ for a given heat load $Q$. The equation shows that in the first case the cooling capacity increases, and in the second case the attainable temperature decreases for increasing values of $n_3$.

Raising $n_3$ indefinitely, even if it could be done, is however no help if the heat exchanger is not ideal. This is because only a limited quantity of heat can be transported per unit time from $P_1$ to $P_2$. With increasing $n_3$ a value is eventually reached at which it is no longer possible to cool the liquid in $P_1$ from 0.7 K, the starting temperature, to the temperature of the mixing chamber. It then follows at once from eq. (3) that the cooling capacity at a given $n_3$ is smaller than it was before, since $T_1$ is now greater than $T_2$ at the lower end of $W$. Let us now suppose, however, as a second extreme case, that the heat leakage $Q$ is zero. Then from eq. (3), for each cross-section between $a$ and $c$:

$$T_2 = \sqrt{\alpha/\beta} T_1 = 0.35 T_1.$$  \hspace{1cm} (5)

The lowest attainable temperature is now determined by the lowest value of $T_1(a)$ that can be reached with the heat exchanger:

$$T_M = T_2(a) = 0.35 T_1(a).$$  \hspace{1cm} (6)

Taken together this yields the following picture. If we let the circulation rate $n_3$ increase from zero, the heat exchanger is initially adequate, and eq. (4) is approximately valid. This implies that at a given $Q$ the temperature $T_M$ decreases. At a later stage, however, the heat exchanger is no longer able to cope. $T_1(a)$ then begins to rise. Since at the same time $Q$ has then become relatively small, eq. (6) is approximately valid, so that $T_M$ also starts to rise. There is therefore an optimum value for $n_3$, which is higher the better the heat exchanger.

In a good heat exchanger the two liquid flows are separated by only a thin foil with the largest possible surface area. Heat exchangers of this type serve their purpose well down to about 100 mK. As the temperature decreases further, however, they rapidly deteriorate because of the marked increase in the Kapitza resistance. This is the resistance to the heat flow that arises at the interface between two dissimilar media, and is a consequence of the difference in the velocity of sound in the two media (due to differences in density and stiffness). Owing to this acoustic mismatch the majority of the phonons — i.e. the waves that are responsible for the heat transport — are reflected from the surface. In a heat exchanger for liquid helium the Kapitza resistance is about $10^4$ times as large as the thermal resistance of the metal of the wall itself. The steep rise with falling temperature is attributable to a decrease in the phonon density. The heat flow $Q_K$ through the interface can be represented by

$$Q_K \propto T_1^4 - T_2^4.$$  \hspace{1cm} (7)

If there is any significant difference between $T_1$ and $T_2$, as in the case where the heat leakage is negligible (see eq. 5), then $Q_K$ is almost completely determined by $T_1$. Equation (7) shows that $Q_K$ is strongly dependent on $T_1$.

The increase in the Kapitza resistance is a serious obstacle to the attainment of temperatures below 10 mK. An added difficulty is that the viscosity of liquid $^3$He behaves very differently from that of the superfluid: it increases with falling temperature. Because of the large surface area necessary in the heat exchanger, at low $T$ and high $n_3$ this can easily lead to considerable viscous heat generation in $P_1$, which counteracts the cooling.

Largely because of the improvements in heat exchangers there are now conventional dilution refrigerators available that can reach a $T_1(a)$ of about 30 mK, and therefore a $T_M$ of about 10 mK. In the refrigerator shown in fig. 5 the part of the heat exchanger at the lowest temperature is divided into four sections $W'_4$; the partition in each of these, separating the liquids, is a folded sheet of foil with a surface area of 350 cm$^2$.

The optimum value of $n_3$ determined by the heat exchanger amounts to about 10 $\mu$mol/s in small refrigerators, and to about 100 $\mu$mol/s in large ones. This corresponds to cooling capacities at 100 mK of the order of 10 $\mu$W and 100 $\mu$W respectively, as may be deduced from eq. (4). In the new type of dilution refrigerator to be discussed shortly, the heat exchanger problem has been completely bypassed. As a result it is advantageous to use higher rates of circulation. With an $n_3$ of about 1 $\mathrm{mmol/s}$ we have achieved a cooling capacity at 100 mK of the order of 1 mW.

**Other factors**

In the still of the conventional dilution refrigerator the concentration of the $^3$He in the liquid, and consequently the vapour pressure of the $^3$He above the liquid, is fairly low (see fig. 9). This implies that to reach a large $n_3$ a high pumping capacity is required. The very high circulation rate of 1 $\mathrm{mmol/s}$ used in the new refrigerator would necessitate a very expensive pump installation in a conventional machine. As we shall show, this is not the case in the new refrigerator because we have succeeded in applying a special method of forced circulation of the $^3$He in the dilute solution, increasing the vapour pressure in the vaporization chamber by about 30 times.

Finally, we should mention the following difficulty in the conventional refrigerator. At the top of the pipe $P_2$
the temperature is higher and therefore lower (see fig. 9), which means that the liquid at the top is heavier than at the bottom. Convection can thus easily be caused by gravity, resulting in a higher \(^3\)He concentration near the top. This means that the \(^3\)He 'gas' is not able to expand as much, and thus takes up less heat from the inflowing concentrated \(^3\)He. In the conventional cooler shown in fig. 5, convection is avoided by arranging the four sections \(WL\) of the heat exchanger in such a way that in these sections the \(^3\)He in the dilute phase flows downwards. As will be seen from the following, the convection problem is not encountered in the new dilution refrigerator.

Cooling with \(^4\)He circulation

The principle of cooling by \(^4\)He circulation, referred to as 'superfluid injection', is illustrated in fig. 12. Two chambers, \(M_1\) and \(M_2\), connected by a relatively wide tube, are filled with liquid helium mixed in such a ratio that when \(M_2\) is cooled a phase separation takes place in it. Owing to the density difference between the two phases, \(M_1\) is then automatically filled with concentrated \(^3\)He. Superfluid \(^4\)He is now continuously injected into \(M_1\) through the superleak \(S_1\). A new interface is thus produced between the superfluid and the concentrated \(^3\)He. Here the mixing takes place that gives the required cooling. The dilute solution continuously produced in this way overflows and 'falls' through the interconnecting pipe into \(M_2\). Whether this does in fact take place in the form of 'droplets' as suggested in fig. 12, we do not know, but it will be convenient here to speak of droplets.

In the Kamerlingh-Onnes laboratory at Leiden a cooling system has been made \(^{[4]}\) in which the superfluid \(^4\)He is extracted from \(M_2\) through a superleak \(S_2\) and returned to \(S_1\). The \(^3\)He supplied by the droplets cannot pass through \(S_2\), and has to return through the phase-separating interface in \(M_2\) to the concentrated mixture \(c\). In this segregation process ('condensation' in the liquid-vapour model) heat is generated which is removed by means of thermal contact between \(M_2\) and an evacuated \(^3\)He bath; in this way \(M_2\) is kept at 0.4 K.

The refrigerator thus obtained has two intrinsic advantages: there is no convection problem and it gives almost ideal heat exchange because of the direct thermal contact between the two liquids. A temperature of 10 mK has been reached in \(M_1\), and the circulation rate and cooling capacity are about the same as those of a large conventional refrigerator. Any further increase in the circulation rate, however, is counteracted by Kapitza-resistance problems in the removal of the heat generated in \(M_2\) to the \(^3\)He bath.

On the other hand, the liquid in such a refrigerator can be put under pressure, since there is no free liquid surface. At about 33 bars the \(^3\)He solidifies, and on a further increase of pressure the solid \(^3\)He is cooled further — through an effect that will not be dealt with here — although the cooling is not continuous. In this way a temperature of 3 mK has been reached.

The new refrigerator with \(^3\)He and \(^4\)He circulation

The dilution refrigerator that we have developed \(^{[8]}\) also uses superfluid injection, with the intrinsic advantages mentioned above. Instead of segregation, however, there is mixing in the lower chamber since this itself, acting in turn as a mixing chamber, forms part of a conventional dilution refrigerator. In this way the temperature in \(M_2\) can be kept at 0.1 K, with no Kapitza-resistance problems.

Fig. 13 shows a diagram of the system. \(M_1\) and \(M_2\) are the two chambers of the refrigerator. The conventional arrangement incorporating \(M_2\) differs from that

---

See also: G. Frossati, G. Schumacher and D. Thoulouze, Proc. 14th Int. Conf. on Low Temp. Physics, Otaniemi (Finland) 1975, Vol. 4, p. 13.
in fig. 3 in that the superfluid in the pipe $P_2$ is now not stationary; $P_2$, together with $V$ and $S_2$, now forms the return channel for the superfluid that is pumped through $S_1$ to $M_1$. This permits an enhanced circulation of $^3$He, as will presently be shown. First, however, we shall discuss the $^4$He circulation.

$^4$He circulation

The $^4$He is circulated by means of a fountain pump [9] ($F$ in fig. 13). To explain its operation we shall recapitulate some of the properties of superfluid $^4$He with reference to the 'two-fluid model'. In this model the liquid is taken as being composed of a superfluid component and a normal component. The density $\rho_n$ of the normal component depends on the temperature, as indicated in fig. 14. The entropy is carried entirely by the normal component.

The normal component is unable to pass through a superleak. This gives rise to the difference in level in the situation shown in fig. 15, which closely resembles that of fig. 6. This effect, known as the fountain effect, can in a sense be regarded as osmosis of the normal into the superfluid component: the superfluid is drawn to the side where $\rho_n$ is greatest. (Nevertheless the term osmosis is better avoided here, since each component can be converted into the other, which is not usually the case in osmosis.)

A simple fountain pump is illustrated in fig. 16. Two chambers, $A$ and $B$, are interconnected by the capillary $C$, and connected through the superleaks $S_1$ and $S_2$ with the $^4$He in the vessel $V$. Chamber $B$ is kept at the
temperature $T$ of the bath by heat exchangers; $A$ is kept at a temperature $T + \Delta T$ by means of an additional heat supply.

Since the normal component cannot pass through the superleak, the superleaks $S_1$ and $S_2$ transmit no entropy; the heat $Q$ generated in $A$ is therefore completely removed through $C$ and $B$. This heat transport is equivalent to transport of the normal component, which is formed in $A$ and then converted into a superfluid in $B$. Because of the fountain effect, superfluid is now drawn from $V$ into $A$ via $S_2$. The same effect, however, would cause superfluid to flow from $B$ to $A$ via $C$, if it were not for the fact that the superfluid is dragged along by the normal component that flows in $C$ from $A$ to $B$. This drag effect occurs when the normal component exceeds a critical speed $v_{en}$; the liquid then becomes turbulent. In designing $C$ close tolerances therefore have to be observed. If the capillary is too wide, the heat conduction will be so good that the desired temperature difference $\Delta T$ will not be attainable; if it is too narrow, the normal component encounters such resistance that the velocity and hence also $n_1$ remain too low.

It is only because of this fountain pump that the refrigerator illustrated in fig. 13 can reach the desired $\dot{n}_4$. Above 1 K the fountain-pressure increases rapidly with temperature; the fountain-pressure difference between 1 K and 1.8 K can be as much as 20 000 Pa (150 mm Hg). A pressure as high as this is necessary to make the system work. This is because the fountain pressure has to oppose the osmosis of the $^3$He in $V$, which would try to draw the superfluid through $S_2$ into $V$. Another point, as we shall see, is that the pump is 'loaded' by the $^3$He circulation.

$^3$He circulation

If the $^4$He liquid flowing through a tube exceeds a critical velocity $v_{es}$, the superfluid becomes turbulent. (This is not the same effect as the turbulence in tube $C$ in fig. 16; $v_{es}$ and $v_{en}$ depend in a different way on the tube diameter.) This property is utilized in the refrigerator to increase the circulation of $^3$He. This is done by including restrictions in the pipe $P_2$ in such a way that the liquid flow is increased to a velocity that is a multiple of the critical velocity. As a result, the $^3$He is drawn along by the $^4$He, the velocities of the $^3$He and $^4$He become identical and $\dot{n}_3$ becomes equal to $X/(1 - X)$ times $\dot{n}_4$. The dilute mixture therefore changes very little in composition, as expressed by the vertical line we have drawn in fig. 9. At the temperature of 0.7 K the vapour pressure in $V$ reaches about 270 Pa (2 mm Hg).

Some of the concentrated $^3$He pumped through $P_1$ into $M_2$ (circulation rate $\dot{n}_3$) makes a detour via $M_1$, mixing there and then entering the dilute solution in $M_2$ by way of the droplets (internal circulation $\dot{n}_{31}$); the remainder ($\dot{n}_{32}$) mixes directly in $M_2$. We now have

$$\dot{n}_{31}/\dot{n}_4 = X_2/(1 - X_2),$$
$$\dot{n}_{31}/\dot{n}_4 = X_1/(1 - X_1),$$

and therefore

$$\dot{n}_{32}/\dot{n}_4 = X_2/(1 - X_2) - X_1/(1 - X_1).$$

Here $X_1$ and $X_2$ are the concentrations on the phase-

![Fig. 16. Fountain pump. When $B$ is kept at the temperature $T$ of the helium in vessel $V$ by thermal contact, and $A$ is raised to a temperature $T + \Delta T$, the $^4$He is caused to circulate. $S_1$ and $S_2$ are superleaks.](image)

![Fig. 17. Detail of the phase diagram, with the temperatures $T_1$ and $T_2$ and the concentrations $X_1$ and $X_2$ of the dilute solutions in the mixing chambers $M_1$ and $M_2$ of fig. 13. When $T_1$ and $T_2$ are lower than about 0.1 K, $X_1$ and $X_2$ are approximately equal. No further mixing then takes place in $M_2$, so that this chamber remains at a temperature of about 0.1 K.](image)

Results

Although the new refrigerator, an example of which is shown in fig. 18, is still in an experimental stage, the following very provisional results indicate that it works well. The results were obtained with a model in which the superleak $S_1$ (see fig. 13) had not yet been connected through the heat exchanger $W''$ to the lower part of $P_2$ or with $M_2$, though it did have a heat exchange with the vaporization chamber $V$. This implies a fairly large heat leakage from $M_1$.

When we set the conventional part of the arrangement into operation we find, as with a conventional dilution refrigerator, that the inlet pressure of the external $^3$He pump is about 100 Pa ($1 \text{ mm Hg}$), corresponding to a circulation rate $l_{32}$ of 1 mmol/s. In a conventional refrigerator the inlet pressure then decreases to 2 Pa, and $l_{32}$ then falls to 10 μmol/s when the temperature of the mixing chamber goes below 0.1 K. In the new refrigerator, however, the fountain pump is brought into operation when the temperature $T_2$ of the lower mixing chamber $M_2$ reaches about 0.35 K. At that moment the temperature $T_1$ of the upper mixing chamber is about 0.7 K. After the fountain pump has started up, $T_2$ first rises to 0.45 K, while $T_1$ falls rapidly. When $T_1$ has reached about 0.45 K, both $T_1$ and $T_2$ decrease further. In this way a temperature $T_1$ of 20 mK can be reached about 10 minutes after the fountain pump has come into operation. The lower mixing chamber takes a longer time (half an hour or an hour) to reach the temperature of 0.1 K. In spite of the heat leakage, the lowest temperature $T_1$ we have reached is about 10 mK.

The cooling capacity can be calculated approximately from the rate at which $T_1$ decreases, since the specific heat of the helium present in $M_1$ is known. A calculation along these lines, at a given fountain-pump speed, gives the value quoted earlier for the cooling capacity: 1 mW at a temperature $T_1$ of 100 mK.

Summary. Since the sixties continuous cooling in the millikelvin range has become possible with 'dilution refrigerators', in which the heat of mixing is extracted from liquid helium present in two phases in a 'mixing chamber': a dilute solution of $^3$He in superfluid $^4$He, with a mixture rich in $^3$He above it. The $^3$He in the dilute solution is extracted through an upper vaporization chamber (still) at a higher temperature, and then returned to the mixing chamber after compression and recondensation. In refrigerators of this type the heat exchangers required between the upward and downward streams of $^3$He are the main obstacle to the cooling process below 0.1 K. It is also difficult to circulate the $^3$He rapidly and to avoid unwanted convection in the upward stream. A new dilution refrigerator is discussed in which these difficulties have been overcome. The circulating $^4$He is injected into a mixing chamber filled with $^3$He; the resultant dilute solution falls into a lower chamber which forms part of a conventional dilution-refrigerator circuit. The heat exchange between the liquids in the pipe connecting the two chambers is almost ideal. The $^4$He carries the $^3$He along with it in restrictions in the pipe, thus enhancing the $^3$He circulation. So far a minimum temperature of about 10 mK has been reached and a cooling capacity of 1 mW at 100 mK, i.e. an order of magnitude higher than that of a conventional dilution refrigerator.
Recent scientific publications

These publications are contributed by staff of laboratories and plants which form part of or cooperate with enterprises of the Philips group of companies, particularly by staff of the following research laboratories:

Philips Research Laboratories, Eindhoven, The Netherlands
Mullard Research Laboratories, Redhill, Surrey, England
Laboratoires d'Electronique et de Physique Appliquée, 3 avenue Descartes, 94450 Limeil-Brévannes, France
Philips GmbH Forschungslaboratorium Aachen, Weißhausstraße, 51 Aachen, Germany
Philips GmbH Forschungslaboratorium Hamburg, Vogt-Kölln-Straße 30, 2000 Hamburg 54, Germany
MBLE Laboratoire de Recherches, 2 avenue Van Becelaere, 1170 Brussels (Boitsfort), Belgium
Philips Laboratories, 345 Scarborough Road, Briarcliff Manor, N.Y. 10510, U.S.A. (by contract with the North American Philips Corp.)

Reprints of most of these publications will be available in the near future. Requests for reprints should be addressed to the respective laboratories (see the code letter) or to Philips Research Laboratories, Eindhoven, The Netherlands.

H. H. Brongersma: Composition and structure analysis of surfaces by ion scattering. Silicates ind. 40, 49-54, 1975 (No. 2).
H. Durand: The Titus system: its characteristics, its performance, its market. Acta Electronica 18, 163-170, 1975 (No. 3). (Also in French.)


F. Meyer & A. Kroes: Surface states at the clean surfaces of cleaved Si(111) and GaAs(110). Surface Sci. 47, 124-131, 1975 (No. 1).


K. Weiss: Kriechvorgänge in einkristallinem NaCl. Z. phys. Chemie neue Folge 94; 273-286, 1975 (No. 4-6).
