Introduction to dilution refrigeration (DilFridg.tex)

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1 Introduction

Dilution refrigerators are fantastic machines: starting from 4.2 K they provide continuous cooling to temperatures as low as 2 mK without moving parts in the low-temperature region. Also from the thermodynamic and hydrodynamic point of view they are interesting machines since they are based a unique combination of a Fermi liquid and a superfluid in the ultralow temperature region.

2 Schematic diagrams

Figure 1: Schematic diagram of a dilution refrigerator. The components are described in the text.

Figs.1 and 2 are schematic diagrams of dilution refrigerators. Fig.1 represents the complete system while Fig.2 represents only the low-temperature part. The working fluid is $^3$He which is circulated by pumps at room temperature. In this Section we will only mention the components. The how and why will be discussed in later Sections.

1. The pumps at room temperature bring the pressure of the $^3$He to a value $p_c$ which usually
Figure 2: *Schematic diagram of the low-temperature part of a dilution refrigerator. The components are described in the text.*

is a few hundred millibar. It enters the cryostat and is precooled by the $^4$He bath to 4.2 K.

2. The $^3$He enters the vacuum chamber. There it is further cooled to the temperature $T_{1K}$ of the 1K bath, which is around 1.2-1.5 K. As $p_c$ is larger than the $^3$He vapor pressure $p_v (T_{1K})$ at $T_{1K}$ the $^3$He condenses in the heat exchanger with the 1K bath. The heat of condensation is removed by the 1 K bath.

3. Next the $^3$He enters the main impedance. The value of the flow resistance is chosen in such a way that $p_c > p_v (T_{1K})$ for the design flow rate of the machine.

4. Next the $^3$He exchanges heat with the still at a temperature $T_S$ which is around 0.5 to 0.7 K.

5. Next the $^3$He enters the secondary impedance. The value of the flow resistance is chosen in such a way that the pressure of the $^3$He in the heat exchanger to the still is larger than the local vapor pressure $p_v (T_S)$. This prevents that the $^3$He, which was liquefied in the 1 K
bath, evaporates again in the still heat exchanger. With $T_S = 700$ mK the vapor pressure $p_v$ is about 100 Pa.

6. Next the $^3$He enters a set of counterflow heat exchangers where it is cooled by the $^3$He in the dilute phase which flows upward in the other side of the heat exchangers. Usually the heat exchangers in the high-temperature region are of the tube-in-tube type. In the colder regions they are more complicated.

7. After leaving the coldest heat exchanger the $^3$He enters the mixing chamber. In the mixing chamber the $^3$He passes the phase boundary (is diluted) between the concentrated phase and the dilute phase. The heat needed for the dilution is the cooling power of the refrigerator. The $^3$He leaves the mixing chamber in the dilute phase.

8. On its way up the cold $^3$He in the dilute phase cools the warm flow of $^3$He in the concentrated phase flowing down.

9. The $^3$He enters the still. The liquid in the still contains only 0.7% $^3$He; the rest is $^4$He. Yet the vapor in the still is practically (96%) pure $^3$He. A heating power $\dot{Q}_S$ is supplied to the still to maintain a steady flow of $^3$He through the system.

The condensation of $^3$He takes place at a point where the incoming flow is in thermal contact with the 1 K bath. This is a fairly well-defined point in the system. Knowing the volumes at the concentrated side, including the volume of half the mixing chamber, one knows exactly how much $^3$He is needed to force the phase boundary in the mixing chamber. Given the position of the phase boundary one can also calculate the quantity of $^4$He to guarantee that there is a liquid surface in the still. There is some $^3$He in the dilute side. This has to be added to the quantity of $^3$He already calculated in the concentrated side and the quantity in the room-temperature part of the machine. Now that we know the quantities of $^3$He and $^4$He we can determine the composition of the gas mixture needed for the dilution fridge.

Nowadays so-called dry dilution refrigerators are available in which the liquid nitrogen and helium baths are replaced by a pulse-tube cooler. A schematic is given in Fig.3. An added advantage is that the 1K bath with its pumping system is no longer needed.
3 Thermodynamics

3.1 1K bath and still

3.1.1 Thermal properties of pure $^3$He

The cooling in the mixing chamber is due to the dilution of $^3$He from the concentrated phase to the dilute phase. In order to understand this cooling process we need to know the thermodynamics of $^3$He-$^4$He mixtures. However, we can understand the function of the 1K bath and the still surprisingly well from the thermodynamic properties of pure $^3$He. Therefore we give here the $H_m - T$ diagram of $^3$He (Fig.4).

3.1.2 The 1K bath

In this section we will first show why a precooling stage, such as the 1K bath is necessary. In order to do this we assume that it would not be there. Consider the system inside the dotted contour of Fig.2. The first law in this case reads

$$0 = \dot{Q} + \dot{n}_3 (H_{m1} - H_{m2})$$  \hspace{1cm} (1)
Figure 4: $H_m - T$ diagram of $^3$He. Lines of constant pressure are given together with the phase diagram.

where the index 1 (2) applies to the entrance (exit) of the $^3$He flow at a molar rate of $n_3$ and $\dot{Q}$ is heating power supplied somewhere to the system. If there is no 1 K bath the temperature of the incoming $^3$He is 4.2 K. With the pressure of 0.5 bar the corresponding molar enthalpy can be obtained from Fig.4 to be $H_{m1}(0.5 \text{ bar}, 4.2 \text{ K}) = 99 \text{ J/mol}$. Now suppose that the dilution refrigerator works properly and that the temperature of the pumped vapor leaves our system at a temperature of 0.7 K. In that case $H_{m2}(0 \text{ bar}, 0.7 \text{ K}) = 35 \text{ J/mol}$. With these numbers Eq.(1) gives

$$\dot{Q} = n_3 (35 - 99) = -64 \frac{\text{J}}{\text{mol}} n_3.$$  \hspace{1cm} (2)

Eq.(2) shows that a positive flow is only possible if the applied heat is negative, so if there is additional cooling. This additional cooling is provided by the 1K bath. The cooling power from the 1K bath should be at least the value given by Eq.(2).

3.1.3 Still

Perhaps it is a bit surprising that a cooling machine like a dilution refrigerator needs heating at some point (in this case the still) to operate properly. The reason for this strange requirement will be explained in this Section.

Due to the presence of the 1 K bath liquid at a temperature of 1.2 K enters the dotted contour in Fig.2 instead of gas at 4.2 K. Fig.4 tells us that $H_{m1} = 6 \text{ J/mol}$. Substituting this value in Eq.(1) gives the value for the heating power

$$\dot{Q}_S = n_3 (35 - 6) = 29 \frac{\text{J}}{\text{mol}} n_3.$$  \hspace{1cm} (3)
This heat usually is supplied at the still, hence the label S. Note that the heating power at the still is calculated without using the mixture properties! The reason is that only pure $^3\text{He}$ is passing the boundaries of our system. The cooling power, that has to be provided by the 1 K bath to change the $^3\text{He}$ from gas at 0.5 bar and 4.2 K to liquid at 0.5 bar and 1.2 K, is

$$Q_{1K} = n_3^* (99 - 6) = 93 \frac{\text{J}}{\text{mol}} n_3^*. \quad (4)$$

In Eq.(2) we saw that 66 J/mol would have been sufficient. This explains why heating power to the still is needed to keep the circulation going. The 1 K bath has too much cooling power. On the other hand the heating power to the still is a very convenient way to set the circulation rate to the desired value.

### 3.2 $^3\text{He}-^4\text{He}$ mixtures

The phase diagram of $^3\text{He}-^4\text{He}$, depicted in Fig.5, gives some important properties as functions of the $^3\text{He}$ concentration $x$. The region between the lines with label $x_d$ and $x_c$ is the region of coexisting phases. The lambda line gives the superfluid transition temperature of the $^4\text{He}$ component. Also the Fermi temperature of the $^3\text{He}$ component is given. Fig.6 shows what happens if a mixture with $x=40\%$ is cooled from 2 K to some very low temperature.

1. At the starting point the mixture is homogeneous and both components behave as normal fluids.
2. At about 1.5 K the $^4$He component becomes superfluid.

3. At 1.2 K the temperature is equal to the Fermi temperature of the mixture. Apart from this nothing special happens at this temperature.

4. At about 750 mK the mixture starts to decompose. At the top of the container a small layer of liquid shows up which has a high helium $^3$He concentration of 80%.

5. If the temperature is lowered further the volume of the upper phase, the concentrated phase, increases and the $^3$He concentration increases as well. The volume and $^3$He concentration of the lower phase, the dilute phase, both decrease. E.g. at 500 mK $x_d = 22\%$ and $x_c = 90\%$.

6. At 0 K $x_d = 6.6\%$ and $x_c = 100\%$.

![Figure 6: Sequence of events if a mixture of 40% is cooled from 2 K to very low temperatures.](image)

In the dilute channel of the dilution refrigerator $\nu_s = 0$ in good approximation. In that case the superfluid hydrodynamics results in the well-known relation

$$p + \rho_0 gz - \Pi - p_f = \text{constant.} \quad (5)$$

Eq.(5), applied to the dilute channel between the mixing chamber and the still, results in

$$p_M + \rho_0 gz_M - \Pi_M - p_{fM} = p_S + \rho_0 gz_S - \Pi_S - p_{fS} \quad (6)$$

where the index M applies to the mixing chamber and the index S to the still. Usually the pressure variations can be neglected. This means that, in a well-designed machine, the flow
resistance of the dilute channel is small. The gravitation and fountain pressure at these low temperatures can neglected as well. In that case relation (6) simplifies to

\[ \Pi (T_M, x_M) = \Pi (T_S, x_S). \] (7)

In other words: in the dilute channel the osmotic pressure is constant. At low temperatures the osmotic pressure in the mixing chamber is fairly temperature independent and fixed to a value close to 2200 Pa (see Fig.7). In that case Eq.(7) is a relation of the temperature and concentration in the still. The temperature and concentration dependence in the dilute channel can be obtained from the isotones in Fig.7. Typically \( T_S = 700 \) mK. As a result of Eq.(7) the \(^3\text{He}\) concentration in the still \( x_S \) is about 0.7%.

### 3.3 The mixing chamber

The cooling power \( \dot{Q}_M \) of the mixing chamber, at a mixing chamber temperature \( T_M \), can be understood on the basis of the second law of thermodynamics. If the mixing is reversible we have in the steady state

\[ 0 = \frac{\dot{Q}_M}{T_M} + \bar{n}_3 \left[ S_3 (T_M, x_c) - S_3 (T_M, x_s) \right]. \] (8)

We must require that the mixing is isothermal since otherwise the mixing is irreversible and an entropy production term would have to be included in the second law. In Eq.(8) \( \bar{n}_3 \) is the \(^3\text{He}\) circulation rate, \( S_3 (T_M, x_c) \) is the molar entropy of the \(^3\text{He}\) entering the mixing chamber; \( S_3 \) is the entropy of one mol \(^3\text{He}\) in the dilute phase, and \( x_s \) is the saturated \(^3\text{He}\) concentration of the dilute phase, which is also a function of \( T \). At low enough temperatures \( x_c = 1 \) and \( S_3 (T_M, x_c) = S_c (T_M) \), the molar entropy of pure \(^3\text{He}\). From Eq.(8) the cooling power is

\[ \dot{Q}_M = \bar{n}_3 T_M \left[ S_3 (T_M, x_s) - S_c (T_M) \right]. \] (9)

The \(^3\text{He}\) in the concentrated phase and in the dilute phase behave as ideal Fermi gases with the molar heat capacity at constant volume given by

\[ C_V = \frac{\pi^2}{2} R \frac{T}{T_F (x)} \] (10)

with \( R \) the molar ideal gas constant and \( T_F \) the Fermi temperature. Based on Eq.(10) the molar entropy is also given by

\[ S_F = \frac{\pi^2}{2} R \frac{T}{T_F (x)}. \] (11)

For the concentrated phase

\[ S_c (T) = \frac{\pi^2}{2} R \frac{T}{T_F (1)} \] (12)

where \( T_F (1) \) is the Fermi temperature of pure \(^3\text{He}\). For the entropy of the dilute phase we can write

\[ S_3 (T_M, x_s) = S_F (T_M, x_s) = \frac{\pi^2}{2} R \frac{T_M}{T_F (x_s)} \] (13)
where $T_F(x_s)$ is the Fermi temperature of the dilute phase. So Eq.(9) can be written as
\[ \dot{Q}_M = n_3 \pi^2 \frac{\pi^2}{2} R T_M^2 \left( \frac{1}{T_F(x_s)} - \frac{1}{T_F(1)} \right). \]  
Eq.(14) clearly shows that the cooling power of the mixing chamber is due to the difference in Fermi temperature between the dilute and the concentrated phase. The Fermi temperature is given by
\[ T_F = \frac{1}{8.25} \frac{\hbar^2}{m^*_3 N_A \left( \frac{V_m(x)}{3} \right)^{2/3}} \]  
with $\hbar$ Boltzmann’s constant, $V_m$ the volume of the mixture containing one mol of $^3\text{He}$. In this expression $m^*_3$ is the effective mass of the $^3\text{He}$. In the dilute phase $m^*_3 = 2.46m_3$ and in the concentrated phase $m^*_3 = 2.8m_3$, with $m_3$ the mass of the bare $^3\text{He}$ atom. The main difference between $T_F(x_s)$ and $T_F(1)$ is due to the difference in the molar volumes. At low temperatures $T_F(x_s) = 393$ mK, $T_F(1) = 1.8$ K, $V_3(x_s) = 426$ cm$^3$ and $V_3(1) = 37$ cm$^3$. So the cooling power of a dilution refrigerator is really based on the increase of molar volume (dilution) of the $^3\text{He}$.

3.4 The osmotic enthalpy

In equilibrium the chemical potential of $^3\text{He}$ and $^4\text{He}$ in the two phases must be the same. So
\[ \mu_3(T, x_s) = \mu_c(T). \]  
Using this relation Eq.(9) can also be written as
\[ \frac{\dot{Q}_M}{n_3} = [\mu_3(T_M, x_s) + T_M S_3(T_M, x_s)] - [\mu_c(T_M) + T_M S_c(T_M)]. \]  
The concentrated phase is pure $^3\text{He}$ so the chemical potential is equivalent to the Gibbs free energy per mole
\[ \mu_c = G_c = H_c - T S_c. \]  
So we can write Eq.(17) as
\[ \frac{\dot{Q}_M}{n_3} = [\mu_3(T_M, x_s) + T_M S_F(T_M, x_s)] - H_c. \]  
Now we would like to write the cooling power in the general form of the first law of thermodynamics using enthalpies
\[ \dot{Q}_M = \frac{^*_3}{n_3} (H_3 - H_c). \]  
This can be done if we introduce
\[ H_3(T, x) = \mu_3(T, x) + T S_F(T, x). \]  
This important quantity is called the osmotic enthalpy. In $^3\text{He}^4\text{He}$ mixtures it plays the same role as the enthalpy in pure fluids.
Fig. 7 gives important properties of mixtures at low $^3$He concentrations and temperatures below about 150 mK. The solid lines, labeled *isotone*, are lines of constant osmotic pressure $\Pi$; the dotted lines, labeled *isenthalp*, are lines of constant osmotic enthalpy $H_3$, and the line, labeled $x_s$, gives the saturation concentration of the dilute phase $x_s$. Note that the vertical axes in Fig.7 is $T^2$ as these quantities tend to vary with $T^2$.

![Figure 7: $T^2 - x$ diagram giving the lines of constant osmotic pressure $\Pi$ (solid lines, values in Pa); lines of constant osmotic enthalpy $H_3$ (dotted lines values in J/mol); and the saturation concentration of the dilute phase $x_s$. The scale on the right gives the temperature in K.](image)

We distinguish four important special cases:

1. The concentration is constant and equal to the saturated concentration at absolute zero (vertical line in Fig.7)
   \[ x_0 = 0.066. \] (22)
   In that case the osmotic enthalpy can be approximated by the relation
   \[ H_{3x} (T, x_0) = 84T^2. \] (23)
   In this relation, and in some of the following relations, the enthalpy is expressed in J/mol.
and the temperature in kelvin.\(^1\)

2. The concentration is equal to the saturated concentration (follow line \(x_s\) in Fig.7). If \(T\) increases \(x\) increases as well. In that case
\[
H_{3s}(T, x_s(T)) = 96T^2. \tag{24}
\]

3. The concentration \(x\) follows a line of constant osmotic pressure (follow isotone of about 2200 Pa in Fig.7). If \(T\) increases \(x\) decreases so that the the osmotic pressure remains constant. In that case
\[
H_{3os}(T, x_{os}(T)) = 54T^2. \tag{25}
\]

4. If (locally) the critical velocity is exceeded, mutual friction sets in and \(H_3\) is constant.

The relation for the molar enthalpy of pure \(^3\)He as a function of temperature is
\[
H_c = 12T^2. \tag{26}
\]

It should be noted that the numbers in the expressions (23)-(26) are only approximate.

In terms of the osmotic enthalpy Eq.(20) can be written as
\[
\dot{Q}_M = \dot{n}_3 [H_3(T_M, x_s) - H_c(T_i)]. \tag{27}
\]

Now it is no longer necessary to limit the expression to reversible mixing. The temperature \(T_i\) is the temperature of the \(^3\)He, entering the mixing chamber in the concentrated phase. With Eqs.(24) and (26) Eq.(27) gives
\[
\dot{Q}_M = \dot{n}_3 (96T_M^2 - 12T_i^2). \tag{28}
\]

If \(\dot{Q}_m = 0\). Using Eqs.(28) gives the famous relation
\[
T_M = \frac{1}{\sqrt{8}}T_i \approx \frac{T_i}{2.8} \tag{29}
\]
which states that there is a fixed ratio between the mixing chamber temperature and the temperature of the incoming \(^3\)He. This property shows the importance of the precooling of the incoming \(^3\)He.

4 Limiting temperature of dilution refrigerators

Consider the exit tube of the mixing chamber. We assume that this is a cylindrical tube with diameter \(D\) and that the flow is laminar. The energy conservation relation for this tube is
\[
\frac{1}{2}C_d \dot{x}^2 \frac{dT^2}{dl} - \frac{\eta_d}{T^2} \frac{128}{\pi D^4} \dot{x}^2 V_{3d}^2 - \frac{\pi}{4} D^2 \frac{d}{dl} \left( \frac{\kappa_d}{T} \frac{dT}{dl} \right) = 0. \tag{30}
\]

\(^1\)The numerical values given here are approximate. If higher accuracy is needed exact numbers should be derived from thermodynamic tables.
The first term represents the enthalpy flow with the parameter $C_d$ derived from the osmotic enthalpy (Eq.(25), $C_d = 2 \times 54$). The second term gives the so-called viscous heating with the viscosity given by

$$ \eta = \frac{\eta_d}{T^2} \quad (31) $$

The third term is the heat conduction with the thermal conductivity given by

$$ \kappa = \frac{\kappa_d}{T} \quad (32) $$

Eq.(30) is a quite complicated differential relation which can only be solved numerically. However, there are some important tricks that can be used to derive essential elements of the relation without having to solve the relation numerically. These "tricks"are:

**4.1 Trick 1**

Introduce a dimensionless length $l$ as follows

$$ l = l_0 \lambda \quad (33) $$

and temperature $\tau$

$$ T = T_0 \tau \quad (34) $$

For the time being $l_0$ and $T_0$ have no meaning other than that we know that they have the dimension of length and temperature respectively. Substitution of (33) and (34) in Eq.(30) gives

$$ \frac{T_0^2}{2l_0 C_d n_3} \frac{d\tau^2}{d\lambda} - \frac{\eta_d}{T_0^2 \tau^2} \frac{128}{\pi D^4} V_3 d n_3^2 - \frac{\kappa_d \pi}{T_0^2} \frac{1}{4} D^2 \frac{d}{d\lambda} \left( \frac{1}{\tau} \frac{d\tau}{d\lambda} \right) = 0. \quad (35) $$

We clean up this expression a little bit by dividing it by the prefactor of the first term so that we get

$$ \frac{d\tau^2}{d\lambda} - 256 \frac{\eta_d}{C_d \pi D^4} V_3 d n_3^2 l_0 \frac{1}{T_0^2 \tau^2} - \frac{2 \kappa_d \pi}{C_d n_3} \frac{1}{4} D^2 \frac{1}{T_0^2 l_0} \frac{d}{d\lambda} \left( \frac{1}{\tau} \frac{d\tau}{d\lambda} \right) = 0. \quad (36) $$

**4.2 Trick 2**

Chose $l_0$ and $T_0$ in such a way that the prefactors of the two other terms become equal to one. So put

$$ 256 \frac{\eta_d}{C_d \pi D^4} V_3 d n_3^2 l_0 \frac{1}{T_0^2} = 1 \quad (37) $$

and

$$ \frac{2 \kappa_d \pi}{C_d n_3} \frac{1}{4} D^2 \frac{1}{T_0^2 l_0} = 1. \quad (38) $$

This leaves a differential equation

$$ \frac{d\tau^2}{d\lambda} - \frac{1}{\tau^2} - \frac{d}{d\lambda} \left( \frac{1}{\tau} \frac{d\tau}{d\lambda} \right) = 0. \quad (39) $$
4.3 Results

Eliminating $l_0$ from Eqs.(37) and (38) gives a relation for the characteristic temperature

$$ T_0 = \left( 128 \eta_d \kappa_d \right)^{1/2} \frac{V_{3d}}{C_d} \left( \frac{1}{D^{1/3}} \right). $$ (40)

Note that the characteristic temperature is independent of the flow rate. For a certain temperature we can determine the characteristic diameter

$$ D_0 = \left( 128 \eta_d \kappa_d \right)^{1/2} \frac{V_{3d}}{C_d} \frac{1}{T_0^{3}}. $$ (41)

Substitution of Eq.(40) in (37) gives a relation for the characteristic length

$$ l_0 = 64 \pi \frac{\eta_d \kappa_d^2 V_{3d}^2}{C_d^3} \frac{1}{\eta_3 T_0^8}. $$ (42)

With the numerical values

$$ C_d = 108 \frac{J}{\text{mol} \cdot \text{K}^2} $$ (43)

and

$$ V_{3d} = 426 \times 10^{-6} \frac{\text{m}^3}{\text{mol}} $$ (44)

and

$$ \eta_d = 5 \times 10^{-8} \frac{\text{Pas} \cdot \text{K}^2}{\text{mol}} $$ (45)

and

$$ \kappa_d = 3 \times 10^{-4} \frac{\text{W}}{\text{m}} $$ (46)

in Eq.(40) we get in engineering terms

$$ \frac{T_0}{\text{mK}} = 5.57 \left( \frac{\text{mm}}{D} \right)^{1/3} $$ (47)

and with Eqs.(41) and (42)

$$ \frac{D_0}{\text{mm}} = \left( 5.57 \frac{\text{mmK}}{T_0} \right)^3 $$ (48)

and

$$ \frac{l_0}{\text{m}} = 130 \frac{\text{mmol/s}}{\eta_3} \left( \frac{\text{mK}}{T_0} \right)^8. $$ (49)

It can be shown that the lowest temperature that can be reached with dilution refrigerators is

$$ T_{\text{lim}} = 0.7 T_0. $$ (50)

Combining Eqs.(50) and (47) it becomes clear that lower temperatures can be reached with wider tubes. However, this is only true if the length of the tube is also longer than $l_0$. For
For \( D = 1 \text{ mm} \) the characteristic temperature is 5.57 mK. For \( T_0 = 2 \text{ mK} \) we get \( D_0 = 21 \text{ mm} \) and with \( n_3 = 1 \text{ mmol/s} \) \( l_0 = 507 \text{ mm} \). This tube (approximately 2 cm diameter and half a meter long) is quite big but manageable. If we lower the temperature with a factor 2 the characteristic diameter increases with a factor \( 2^3 = 8 \), and the length with \( 2^8 = 256 \), and the volume with a factor 16,384. There is no fundamental limit for the lowest temperature of dilution refrigerators: the bigger the machine the lower the temperature will be. However, below about 2 mK, there is the alternative of nuclear refrigeration, which turns out to be a more practical solution than just making huge dilution refrigerators.