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A He\textsuperscript{3}–He\textsuperscript{4} Dilution Refrigerator

by

Donald Otis Pederson

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A $^3\text{He}-^4\text{He}$ Dilution Refrigerator

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ABSTRACT

A $^3\text{He}-^4\text{He}$ dilution refrigerator has been designed and built. A general description of the dilution refrigerator and its operation are given. The principal differences from other refrigerators are the design of the still which includes a $^4\text{He}$ film flow limiting evaporator and the use of high purity silver in the heat exchangers. The refrigerator has been run and has achieved temperatures less than 55 mK for several hours. These temperatures were obtained on the outside of the mixer where any practical application of the cooling power will occur. Instabilities in the operation of the refrigerator have restricted the analysis of the refrigerator and its use. There are indications that the $^4\text{He}$ film flow limiter has some effectiveness. Suggestions are made that might solve the instabilities so that the refrigerator can be studied and operated routinely.
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I. INTRODUCTION

I.1. Historical Survey

In 1951, H. London proposed a new cooling process which makes use of the entropy of mixing of $\text{He}^3$ and $\text{He}^4$ at low temperatures. He suggested that a reversible adiabatic dilution by a factor of one thousand would cause the temperature to fall from 1 K to 10 mK. He pointed out that the dilution of either $\text{He}^3$ or $\text{He}^4$ by the other component is analogous to the adiabatic expansion of a gas.

Following the liquifaction of $\text{He}^4$ in 1908 by Kamerlingh Onnes, $\text{He}^4$ was successfully used in refrigerators by reducing the vapor pressure. By using a diffusion pump, temperatures of about 0.9 K are readily obtainable. The lowest temperature obtained using liquid $\text{He}^4$ (L$\text{He}^4$ hereafter) has been about 0.71 K, achieved with either very high speed diffusion pumps or by pumping the L$\text{He}^4$ through a small hole. There are two problems in achieving temperatures less than 1 K in L$\text{He}^4$. The vapor pressure decreases rapidly below 1 K. At 0.71 K the vapor pressure in only 2.7 μ (see Figure 6). The second difficulty is the presence of the superfluid $\text{He}^4$ film. This film flows up the walls of the $\text{He}^4$ refrigerator until it reaches some higher temperature and evaporates. If the pumps are not sufficiently large, part of the evaporated $\text{He}^4$ film will flow as a gas back to the L$\text{He}^4$ bath thus increasing the heat leak. In order to achieve low temperatures, either very large pumps must be used to remove the vapor from both the L$\text{He}^4$ bath and the $\text{He}^4$ film, or a constriction must be used to limit the flow of the $\text{He}^4$ film and the subsequent diffusion back to the L$\text{He}^4$ bath.
The superfluid film is also a problem in the dilution refrigerator (see Section II.2).

He\textsuperscript{3} was first liquified in 1948 by Sydoriak, Grilly, and Hammel.\textsuperscript{5} In the years following the liquifaction of He\textsuperscript{3} and London's suggestion, low temperature techniques developed in the area of He\textsuperscript{3} refrigerators rather than dilution refrigerators. By pumping on LHe\textsuperscript{3} with a diffusion pump in a properly designed refrigerator, temperatures on the order of 0.20 K are obtainable.\textsuperscript{6} LHe\textsuperscript{3} does not have a superfluid transition in this temperature range so that the problem of the superfluid film is not encountered. The vapor pressure of LHe\textsuperscript{3} is also much higher than the vapor pressure of He\textsuperscript{4}. At 0.71 K, the vapor pressure of He\textsuperscript{3} is about 1.5 x 10\textsuperscript{3} μ (see Figure 6).

A third common method of obtaining low temperatures is adiabatic demagnetization.\textsuperscript{7} Wheatley describes adiabatic demagnetization refrigerators below 20 mK\textsuperscript{8} and the use of cerium magnesium nitrate (CMN hereafter) to obtain temperatures of 2 mK.\textsuperscript{9} The use of CMN in adiabatic demagnetization refrigerators is important to the design of a dilution refrigerator because the thermal problems of mK adiabatic demagnetization and the use of CMN as a magnetic thermometer are common to both methods.

In 1960, London and co-workers\textsuperscript{10} again suggested the dilution of He\textsuperscript{3} by adding He\textsuperscript{4} through a superleak and they also noted that a continuous system seemed feasible. Finally in 1962, London and co-workers\textsuperscript{11} indicated several possible methods of recirculating the He\textsuperscript{3} in order to provide continuous dilution. Both of London's designs use separate He\textsuperscript{3} refrigerators to recondense the gas from the still and differ only
In the method of bringing the He\textsuperscript{3} from the mixing chamber (hereafter the mixer) to the distillation chamber (hereafter the still). The first method uses two superleaks and supercritical LHe\textsuperscript{4} flow between them to force the He\textsuperscript{3} from the mixer to the still. The disadvantage with this method is in the use of superleaks which do not remain reliable after repeated thermal recycling. In the second method, the He\textsuperscript{3} moves from the mixer to the still primarily by osmotic pressure. Hall\textsuperscript{11} suggested a third method in which the separate He\textsuperscript{3} refrigerator is eliminated and the He\textsuperscript{3} from the still is recompressed at room temperature and condensed at 1 K in a He\textsuperscript{4} bath. The LHe\textsuperscript{3} is then passed through heat exchangers containing the dilute He\textsuperscript{3} phase (hereafter concentrated or dilute refers to the relative amount of He\textsuperscript{3} in He\textsuperscript{4}) between the still and mixer. The advantage of this method is that the LHe\textsuperscript{3} is at much lower temperatures when it enters the mixer. The absorption of heat by the dilute phase from the concentrated phase is limited only by the thermal conductivity of the liquids and the heat exchangers and at low temperatures by the Kapitza boundary resistance. In fact, the dilute phase in the still must be heated to keep it at the right temperature since the dilute phase has such a large cooling capacity.

At the LT9 Conference, Das and co-workers\textsuperscript{12} reported the first dilution refrigerator built at Leiden obtained a temperature of 0.22 K. In 1966, Neganov and co-workers at Dubna\textsuperscript{13} and Hall and co-workers at Manchester\textsuperscript{14} independently reported the first really successful He\textsuperscript{3}-He\textsuperscript{4} refrigerators. The Dubna group reported an ultimate temperature of 0.056 K on their first model and 0.025 K after modifications. The Manchester group obtained a temperature of 0.07 K with their initial design
and with improvements extended its lower limit to 0.05 K.\textsuperscript{15} The design changes of both refrigerators were primarily in increasing the surface area in the heat exchangers and mixing chamber since the heat transfer varies with the area of contact.

Vilches and Wheatley initially built a He\textsuperscript{3}-He\textsuperscript{4} refrigerator which has a continuous operating temperature of 0.020 K.\textsuperscript{16} Their refrigerator differed principally in the design of the heat exchangers and is discussed in Section I.3. Vilches and Wheatley used this refrigerator as a starting point for a single cycle He\textsuperscript{3}-He\textsuperscript{4} refrigerator. By removing He\textsuperscript{3} from a mixture without recycling the He\textsuperscript{3} they obtained a temperature of 4.5 mK for at least 30 minutes.\textsuperscript{17} Wheatley and co-workers have improved their refrigerator so that they are now able to maintain 10 mK continuously.\textsuperscript{18,19} There are many other He\textsuperscript{3}-He\textsuperscript{4} refrigerators either under construction or operating but they are based on the general designs of the earlier refrigerators.\textsuperscript{20-36}
I.2 The Dilution Refrigerator

The essential features of the refrigerator which has been built are shown schematically in Figure 1. Beginning in the still, the dilute solution is typically about 1% He\(^3\). The vapor is highly concentrated He\(^3\). The He\(^3\) is pumped from the still by a pumping system at room temperature where it is compressed to pressures greater than 20 torr. The He\(^3\) is then condensed in a He\(^4\) bath at a temperature of about 1.2 K. Heat exchange is made to the dilute solution before it enters the concentrated phase in the mixer. In order to maintain the liquid phase in the return line, several flow impedances are introduced. There is a steady state He\(^3\) concentration gradient from the mixer to the still which is a result of the temperature gradient. As He\(^3\) is removed from the still by pumping on the still, the osmotic pressure of the solution drives the He\(^3\) from the dilute phase of the mixer to the still so that this concentration gradient is unchanged. He\(^3\) then diffuses across the phase boundary in the mixer into the dilute phase so that phase equilibrium is maintained. The amount of He\(^3\) in the mixer is kept constant by the returning He\(^3\) pumped from the still. The fountain pressure which drives normal He\(^4\) is small because the temperature is low enough that the normal fluid concentration in the He\(^4\) is very low. Various parts of the refrigerator and its operation will be discussed later in greater detail.

Physically, the operation of the dilution refrigerator is somewhat analogous to the cooling process that occurs when the vapor is pumped from above a liquid. The upper concentrated phase corresponds to the liquid phase and the lower dilute phase corresponds to the less dense
**Figure 1.** He₁₃−He₄ Dilution Refrigerator

- He₁₃ Film Flow Limiter
- Condenser $T_c = 1.2$ K
- Flow Impedance
- Still Heat Exchanger
- Heat Exchange
- Dilute Phase (He₁₃ Diffuses Through He₄)
- Concentrated Phase (He₃)
- Phase Boundary $T_i = 0.02$ K
- 80 - 99% He₄
- $T_S = 0.7$ K
- 99% He₃ Gas
- 1% He₃ in He₄

Room Temperature Pumps
gas phase. In an ordinary He$^3$ refrigerator, for example, liquid He$^3$
evaporates (moves across the phase boundary) into the less dense He$^3$
gas where it is pumped away. In the dilution refrigerator, the He$^3$
diffuses (moves) across the phase boundary into the non-interacting He$^4$
background containing the dilute He$^3$ where it is removed by diffusion to
the still and eventually pumped away. However, the analogy is not
complete and the two methods of cooling differ in several important
respects. In a pumped liquid He$^3$ refrigerator, the latent heat of
evaporation per atom or the energy required to remove one atom from
the liquid to the gas is only weakly temperature dependent while the
vapor pressure decreases exponentially as the temperature is reduced so
that it becomes physically difficult to remove sufficient numbers of
atoms to achieve cooling. In a dilution refrigerator at low temperatures,
the concentration of He$^3$ atoms in the dilute phase is only weakly tempera-
ture dependent while the heat of mixing per atom or the energy required
to remove a He$^3$ atom from the He$^3$ phase to the dilute He$^3$ phase is pro-
portional to the square of the temperature. Since most mechanical
pumps have pumping speeds, $V$, which are relatively independent of pressure
unless it is near the ultimate pressure, the amount of gas pumped or
the throughput will be proportional to the pressure. The cooling
available will be the heat of mixing or evaporation times the throughput.
Thus for a He$^3$ refrigerator, the cooling capacity decreases exponentially
with decreasing temperature while for a dilution refrigerator the cooling
capacity decreases as approximately the square of the temperature. Making
some simple assumptions about the throughput of a single Welch 1402 pump
on either a single charge He$^3$ refrigerator of a dilution refrigerator
indicates that below about 0.42 K the dilution refrigerator has the larger cooling capacity of the two refrigerators.

A more accurate thermodynamic analog can be made between a dilution refrigerator and certain thermoelectric effects. The cooling process in the dilution refrigerator is analogous to the Peltier effect and the heat transfer in the heat exchangers is analogous to the Thomson effect. Both thermoelectric effects result from the coupled flow of heat and electricity in conductors where small temperature and potential differences exist (see for example Ref. 37). The electrons in two conductors which make a junction are analogous to the He\textsuperscript{3} atoms in the two phases. A current of one ampere corresponds to approximately 10^{-5} mole/sec of electrons which is a typical molar flow rate for He\textsuperscript{3} atoms in dilution refrigerators. The Peltier cooling rate at a junction at a temperature T is proportional to the current and is related to the absolute thermoelectric power $\varepsilon$ of both material

$$\dot{Q}_c = [\varepsilon_1(T), \ldots, \varepsilon_2(T)] T I$$

The absolute thermoelectric power is proportional to temperature for a free electron system. The cooling capacity depends on temperature at least qualitatively in the same manner as cooling in the mixer. The heat transfer in heat exchangers is the analog of the Thomson effect in which a current in either conductor which is part of a junction causes a small heat flow out of the conductor along its length. Joule heating is analogous to the viscous heating of the flowing He\textsuperscript{3} but here the analog is not exact. The He\textsuperscript{3} flows laminarily so that the viscous heating in long circular tubes of diameter $d$ is

$$\dot{Q}_v = \frac{1}{d^4}$$
Joule heating is

$$\dot{Q}_J \propto \frac{1}{d^2}$$

The heat flow in either liquid He\(^3\) (neglecting convection and the conduction of the tube itself) or a conductor is

$$\dot{Q}_T \propto d^2$$

It can be seen from the different dependences on diameter that viscous heating can be reduced faster than thermally conducted heat is increased, but that this is not true for joule heating. There is a final important difference between the two systems which explains why dilution refrigeration is important and thermoelectric refrigeration is not as important. The entropy per particle for weakly interacting degenerate Fermi systems is directly proportional to the effective mass which is much larger in the dilute phase than in conductors or semiconductors.
I.3. Properties of Helium Solutions

There are several properties of liquid helium which are important to the successful design and operation of the dilution refrigerator.

In 1956, Walters and Fairbank\textsuperscript{38} first detected the phase separation of He\textsuperscript{3}–He\textsuperscript{4} solutions below 0.8 K. The lighter He\textsuperscript{3} phase floats above the He\textsuperscript{4} phase. Peshkov and Zinov'eva\textsuperscript{39} visually observed the phase separation as a well-defined meniscus. The existence of this phase separation enables a refrigerator to be built in which mechanical isolation of the mixing components (e.g. a superleak) is unnecessary. The detailed phase diagram of He\textsuperscript{3}–He\textsuperscript{4} solutions is given in Figure 2. The maximum temperature for the coexistence of the two phases is at 0.872KK and at a He\textsuperscript{3} concentration of 66.9\%.\textsuperscript{40}

As indicated on the phase diagram, the phases do not completely separate as the temperature approaches 0 Kelvin. The upper phase is essentially pure He\textsuperscript{3} at temperatures less than 0.15 K\textsuperscript{41} while the lower phase has a limiting solubility of He\textsuperscript{3} in He\textsuperscript{4} at T = 0 Kelvin of 6.4 to 6.8\%.\textsuperscript{42–46} The finite solubility of He\textsuperscript{3} at 0 Kelvin enables the dilution process to be carried out at any low temperature. In this respect the analogy to liquid helium evaporating breaks down, since as the temperature is decreased the vapor pressure above the liquid decreases monotonically to zero.

The heat capacity of liquid He\textsuperscript{3}, He\textsuperscript{4} and their solutions is important in the design of heat exchangers between the concentrated and dilute phases. The heat capacities in the region of interest are given in Figure 3.\textsuperscript{47} The heat capacity per mole of He\textsuperscript{3} is smaller in the concentrated phase than in the dilute phase. The small heat capacity of liquid
FIGURE 2. PHASE DIAGRAM OF He$^3$-He$^4$ SOLUTIONS

$\lambda$ LINE

NORMAL REGION

SUPERFLUID REGION

PHASE SEPARATION REGION
FIGURE 3. THE SPECIFIC HEATS OF $^4\text{He}$, $^3\text{He}$ AND $\text{He}^3$ AT CONSTANT CHEMICAL POTENTIAL FOR MIXER TEMPERATURES OF 0.0 K AND 0.10 K
He⁴ is a result of the relative absence of excitations in liquid He⁴ at low temperatures in particular the lack of rotons below ~0.6 K.

The liquid He⁴ can be described in terms of the two-fluid model in which the excitations make up the normal interacting fluid with a relative density ρ_n/ρ which is small below 1 K. Thus the liquid He⁴ is primarily a noninteracting background through which the He³ moves. This noninteracting property of the He⁴ means that the circulating He³ does not drag the He⁴ with it. The heat capacity of dilute solutions of He³ in He⁴ at constant He⁴ chemical potential is determined by the quantum properties of dilute He³ solutions. In the low temperature region, the dilute He³ can be described as an ideal Fermi gas with an effective mass resulting from weak He³-He³ interactions so that the specific heat is approximately linear in temperature. In the region around 0.5 K where the heat capacity is relatively temperature independent and equal to the classical constant pressure specific heat of 5/2 R, the dilute He³ behaves as a classical ideal gas. The increase in heat capacity at higher temperatures results from the increased number of He⁴ quasiparticles which interact with the He³.

The enthalpy of He³ and dilute solutions of He³ in He⁴ is given in Figures 4 and 5.⁴⁷ The enthalpy of He⁴ is not shown on the graphs since at T = 0.0 K the enthalpy of He⁴ is -39.06 J/mole relative to the enthalpy of He³ and appears constant with temperature by comparison with the enthalpy of He³. The general property of interest is that the enthalpy of pure He³ is lower than that of mixtures of He³ in He⁴ so that an adiabatic dilution of He³ results in a cooling process.

The relative vapor pressures of He³ and He⁴ are given in Figure 6.⁴⁸,⁴⁹
FIGURE 4. ENTHALPY DIAGRAM FOR PURE He³ AND He³–He⁴ SOLUTIONS
FIGURE 5. THE ENTHALPY OF PURE He$^3$ AND He$^3$ IN LIQUID He$^4$ AS A FUNCTION OF TEMPERATURE FOR VARIOUS He$^3$ CONCENTRATIONS.
VAPOR PRESSURE (torr)

FIGURE 6. THE VAPOR PRESSURES OF He3 AND He4
For typical still temperatures of 0.7 K or less, the vapor pressure of He\textsuperscript{3} is about 10\textsuperscript{3} times that of He\textsuperscript{4}. For an ideal solution, the enrichment factor \( X_V/X_L \) depends directly on the ratio of the vapor pressure of the pure constituents.

\[
X_V/X_L = P_{30}/P_{40}
\]

Dilute solutions of He\textsuperscript{3} in He\textsuperscript{4} at low temperatures are not ideal but nevertheless the vapor pressure ratio indicates large enrichment factors. Experimentally, one finds for solutions with He\textsuperscript{3} concentrations \( X_L \approx .01 \) at \( T = 0.7 \) K, the vapor concentrations \( X_V \approx .97 \). Thus fractional distillation is a powerful means of separating He\textsuperscript{3} and is a convenient method of separating He\textsuperscript{3} in dilution refrigerators for closed cycle circulation.

In contrast to the several properties of liquid helium which enhance the operation of the dilution refrigerator, there are several properties which essentially limit the capabilities of the technique unless special precautions are taken to minimize their effects.

Convective instability of the dilute phase was a problem in earlier refrigerators.\textsuperscript{14} The convective instability results from the higher density of the dilute phase in the still since the He\textsuperscript{3} concentration is smaller. Convective instabilities are damped by the viscosity of the dilute phase and by the diffusion of He\textsuperscript{3}...H. London estimated that the critical inner diameter of the dilute phase tube is of the order of 1 mm for the typical conditions.\textsuperscript{14} Later refrigerators have not experienced this difficulty since the dilute phase tube inner diameters are kept less than 1 mm. Peshkov used a 1 mm inner diameter dilute phase tube and found it necessary to have a loop in it to prevent instabilities.\textsuperscript{24}
The loop provided a region where the density gradient was in the proper direction (i.e. the heavier liquid was at the bottom of part of the loop).

The method of separating $\text{He}^3$ from $\text{He}^4$ by fractional distillation has already been discussed. However the vapor in equilibrium with the dilute phase is not the only gas that can be pumped from the still. Mixing $\text{He}^3$ into $\text{He}^4$ does not eliminate the superfluid film property of liquid $\text{He}^4$ at all concentrations and temperatures (see Figure 2). The gas pumped from the still can include the evaporated film, thus decreasing the concentration of $\text{He}^3$ in the circulated gas. Since the pumps used for circulation are normally run at near capacity, the amount of $\text{He}^3$ circulated is decreased. The cooling capacity of the dilution process, which depends on the number of atoms diffusing across the boundary, is therefore reduced. For a fixed heat leak, the minimum temperature obtainable in the mixing chamber is increased. A circulation of 20% $\text{He}^4$ results in at least a 10% increase in the minimum temperature available. Earlier $\text{He}^3$-$\text{He}^4$ refrigerators all used constrictions of the order of 1 mm in the $\text{He}^3$ pumping tube to minimize the $\text{He}^4$ film flow and subsequent evaporation. Taking the film flow rate as $8 \times 10^{-5}$ cm$^3$/sec$^{50}$ of L$\text{He}^4$ per cm perimeter, a 1 mm orifice allows about $9 \times 10^{-7}$ moles/sec of film flow. Experiments in which the circulated gas was replaced by a pure $\text{He}^3$ source resulted in the mixer temperature decreasing from 13 mK to 10 mK.$^{35}$ Several designs which eliminate the $\text{He}^4$ circulation resulting from the $\text{He}^4$ film will be discussed later.

Heat flow between two materials at low temperatures is impeded by a thermal boundary resistance which is known as the Kapitza boundary resistance when one of the materials is liquid helium.$^{51,52}$ The thermal
boundary resistance establishes a temperature discontinuity at the boundary whenever there is a heat flow. The Kapitza conductance, $h$, for heat transfer from liquid helium is defined

$$h = \lim_{\Delta T \to 0} \frac{\dot{Q}}{A \Delta T}$$

where $\dot{Q}/A$ is the heat flow per unit area across the interface and is found experimentally to vary as

$$h = a T^n \quad [\text{erg/cm}^2 \text{ sec K}]$$

where $a$ depends on the material and $2.5 \leq n \leq 4.5$. The Kapitza conductance is at present an unexplained physical phenomenon.$^{52}$ The acoustic-like mismatch of the thermal phonons in the different materials suggests that there should be a thermal boundary resistance but the predictions of this theory differ by several orders of magnitude from experimental results. The heat flow across a boundary is

$$\dot{Q} = h A \Delta T = a T^n A \Delta T$$

As the temperature decreases, the only way to compensate for the decreased conductance is to increase the interface area across which the heat is transferred.

Early refrigerators used heat exchangers made of capillary tubes. The Leiden group used a capillary for the concentrated phase which was coiled inside another coiled tube containing the dilute phase.$^{53}$ Hall$^{14}$ pointed out that the failure by the Leiden group to reach temperatures less than 0.1 K was probably due to convective instabilities in their 2.5 mm diameter dilute phase tube. The Manchester group$^{14}$ used a heat exchanger similar to the Leiden group but instead of one tube containing the dilute phase, they coiled the concentrated He$^3$ capillary
between two concentric tubes after bringing it down the inside of the inner tube. The He\(^3\) moved through the dilute phase down the annular region and back up the center tube. This design eliminated the need to coil the capillary inside a tube of diameter less than 1 mm but it required that the heat exchanger extend below the mixing chamber. The Dubha group\(^{13}\) used a much simpler heat exchanger of three concentric tubes. The outer annular region contained the dilute phase. The He\(^3\) flowed straight down the outer annular region and diffused straight back up the inner annular region. A different type of heat exchanger was developed by Wheatley at Illinois.\(^{16,18,54}\) In order to increase the area of contact between the phases and the heat exchanger, the cores of the heat exchangers were made of sintered copper. The porous sintered copper blocks were made by pressing copper powder and coësaking it below the melting point. Wheatley's heat exchanger\(^{18}\) consisted of five separate heat exchangers between the still and the mixer. Each phase communicated with a separate sintered piece set into a copper block so that the whole block was in thermal equilibrium at a single temperature. Recently built refrigerators have combined both general types of heat exchange systems.\(^{22,25,36}\)

Viscous heating of the He\(^3\) diffusing through the dilute phase is a limiting factor in determining the minimum temperature of the mixing chamber.\(^{19}\) However estimates of the effect of viscous heating indicate that it is negligible above 5 mK (see Ref. 17 for detailed discussion).
II. THEORY OF OPERATION

II.1. Temperature Measurement

The primary temperature measurement in the experimental cell is the magnetic temperature, $T^*$, of powdered cerium magnesium nitrate, $\text{Ce}_2\text{Mg}_3(\text{NO}_3)_{12}\cdot 24\text{ H}_2\text{O}$. CMN is useful as a thermometer since its magnetic susceptibility, $\chi$, follows Curie's law quite well to very low temperatures. Good thermal contact between the CMN thermometer and both the sample and mixing chamber is provided by thermal contact with liquid He$^4$ in the experimental cell which is enhanced by the increased surface area of the powdered CMN and the decreased liquid He$^4$ flow impedance of the relatively low packing fraction (~1/2). The powdered CMN salt pill used in the experimental cell is in the shape of a right circular cylinder with diameter equal to height because of the convenience with which the lightly packed salt pill can be constructed.

An ideal paramagnetic system obeys Curie's Law where the susceptibility is inversely proportional to the thermodynamic temperature, $\chi T = \text{constant}$ (hereafter the temperature refers to the thermodynamic temperature unless otherwise indicated). Weak interactions between the paramagnetic entities in a salt make Curie's Law only approximately valid, $\chi T = c$. A magnetic temperature, $T^*$, can be defined by $\chi T^* = c$. If the relation between $T$ and $T^*$ is determined, then a measurement of the susceptibility provides a determination of the temperature. The relation between $T$ and $T^*$ for CMN has been reviewed most recently by Hudson$^{55}$ and Wheatley.$^{35}$ Experimental evidence indicates that the magnetic temperature for a single crystal sphere, $T^2$, is related to the temperature above 6 mK
by the expression $T^\circ = T - 9 = T + 0.27 \text{ mK}$. Deviations from this Curie-
Weiss behavior occur below 6 mK\textsuperscript{56-58} and are detectable as high as 8 mK. There is conflicting experimental evidence on the relation between the magnetic temperature, $T^\circ$, for a right circular cylinder of powdered CMN with diameter equal height and the temperature $T$. There are many experiments indicating that $T^\circ = T + \Delta$ down to 2 or 3 mK, where $\Delta$ is the order of a few tenths of a millikelvin.\textsuperscript{59-66} A right circular cylinder of powdered CMN has several properties which might increase departures from Curie's Law. The demagnetizing field of a right circular cylinder is not uniform. Even if some average demagnetizing field is used, it depends on the susceptibility so that the effective demagnetization coefficient is temperature dependent for a Curie salt. The filling factor $f$ of a powdered salt and the averaging factor $n$ over a highly anisotropic powdered material both account for corrections to Curie's Law. It is somewhat surprising that all these factors combine to enhance the thermometric property of CMN in that the magnetic temperature of a right circular cylinder of powdered CMN follows the thermodynamic temperature to a lower temperature than the magnetic temperature of a single crystal sphere of CMN. Heat capacity measurements on powdered CMN yield values of $\Delta$ larger than a millikelvin\textsuperscript{66,67} and cast doubt on previous determinations of the thermometric properties of CMN. Nevertheless the preponderance of evidence supports a small value of $\Delta$. Included in the support of a small $\Delta$ is the fact that the value of $\Delta$ obtained from heat capacity measurements is not as accurate as that obtained from other measurements.\textsuperscript{61,66,68} Because of the uncertainty in the $T - T^\circ$ relation, all data in this work have been left in terms of $T^\circ$ noting here that $T^\circ \approx T$ over the range of the experiment.
II.2. \( ^{4}\text{He} \) Superfluid Film Limiting Design

The consequence of the \( ^{4}\text{He} \) film in both \( ^{4}\text{He} \) refrigerators and \( ^{3}\text{He}^{4}\text{He} \) dilution refrigerators has already been pointed out. Constrictions in the pumping line are not desirable since the cooling rate and ultimate temperature of the mixer depend directly on the circulation rate which is limited by constrictions. It should be possible in the \( ^{3}\text{He}^{4}\text{He} \) refrigerator to use another method to limit the \( ^{4}\text{He} \) film which is not possible in ordinary \( ^{4}\text{He} \) refrigerators. Since the still must be heated to keep it at the proper temperature for optimum preferential evaporation of \( ^{3}\text{He} \) from the dilute phase, there is a heat input available to control the \( ^{4}\text{He} \) film which is not unfavorable to the low temperature operation of the \( ^{3}\text{He}^{4}\text{He} \) refrigerator as it would be in \( ^{4}\text{He} \) refrigerators. If the \( ^{4}\text{He} \) film can be evaporated in a geometry where it is very probable that it will recondense, then the \( ^{4}\text{He} \) content of the gas in the still can be minimized. The gas pumped from the still will then have a higher \( ^{3}\text{He} \) concentration.

The \( ^{4}\text{He} \) superfluid film flows along the walls of its container with a thickness on the order of 200 Å. It flows until it reaches some part of the apparatus at a higher temperature and evaporates. The temperature of the \( ^{4}\text{He} \) superfluid is nearly uniform because of the large thermal conductivity of superfluid \( ^{4}\text{He} \).

Studies on heat flow from fine wires in \( ^{1}\text{He}^{2} \) \(^{69}\) have shown that a cylindrical vapor bubble of diameter of the order of 1 mm forms along the entire heated wire in bulk \( ^{1}\text{He}^{2} \). A wire heated in \( ^{1}\text{He}^{2} \) between 1.6 K and 2.17 K is first surrounded by \( ^{1}\text{He} \) and then at wire temperatures 1-2 K above \( T_{\lambda} \) a vapor bubble forms. Various experimental studies have
indicated that characteristic maximum heat fluxes which determine the onset of vaporization in bulk He II are temperature and geometry dependent.\textsuperscript{69,70} For single-wire heating in the He II film, there is no possibility for zero mass flow heat convection. The normal component is locked into position. Therefore heating of the film by a wire results first in He I and then in vaporization. The temperature of the vapor will be approximately $T_\lambda(P)$. The idea is to evaporate the film so that there is a high probability that the He\textsuperscript{4} gas will recondense either on the liquid surface or on a film surface between the evaporator and the liquid. The discussion of the concept will be in terms of the present design but the ideas are applicable to other designs.

If the separation between the evaporator and a surface on which film is flowing is on the order of the gaseous He\textsuperscript{4} mean free path, then it is very probable that any evaporated He\textsuperscript{4} atom will be able to reach the He\textsuperscript{4} film. The He\textsuperscript{4} gas will recondense on the film similar to He\textsuperscript{4} recondensation on the bulk LHe\textsuperscript{4} in ordinary He\textsuperscript{4} refrigerators.

Figure 7 is a diagram of the design. The LHe\textsuperscript{4} film is evaporated by heating a wire jet into the top surface of the inside stainless steel plate. The evaporated He\textsuperscript{4} will recondense on the He\textsuperscript{4} film above the evaporator. If there is good heat transfer between the recondensed He\textsuperscript{4} and the upper brass wall, the recondensed He\textsuperscript{4} will be cooled to the temperature of the still and the original heat input from the wire will be conducted by the still walls to the dilute phase. The still in this design could have been made of copper for better thermal conductivity. When this recondensed He\textsuperscript{4} is cooled to the still temperature, it should behave exactly like the superfluid flowing from the still and join the
FIGURE 7. EVAPORATION AND RECONDENSATION OF THE $^4$He SUPERFLUID FILM IN THE STILL
flow to the heat source. By causing recondensation of the evaporated He\(^4\) onto the film prior to its passage over the smallest perimeter at the maximum height above the liquid level, the recondensed He\(^4\) must be included in the amount of flow determined by the smallest cross-section. When the He\(^4\) film flows from a LHe\(^4\) reservoir, the flow rate at the reservoir is determined by the smallest cross-section of the film as it flows up the container. In other words, the film flow rate "knows" its maximum before it reaches the limiting cross-section. It is possible in the present design that the He\(^4\) film flowing from the dilute phase will "know" that there is another film source which supplies the limiting amount of He\(^4\) film which will flow over the smallest perimeter. If this is possible then the dynamic process of evaporation, recondensation, cooling and flow to the heated wire may stop the film flow from the dilute phase. However, the question about the film knowing it cannot flow is not answered even by the wealth of data about He\(^4\) films. If the heat transfer is not sufficient, then there will be an increase in the normal LHe\(^4\) density above the evaporator. Either of these two unknown factors will cause an increase in the volume of He\(^4\) in the region of the evaporator. If most of the He\(^4\) that flows from the still reaches the dilute phase again instead of being pumped out with the He\(^3\) gas, the design will still work. There are two possibilities which would prevent the idea from working. The evaporated He\(^4\) could be mixed with the equilibrium vapor above the dilute phase and pumped from the still. The film flow could pass unbroken over the heater. The latter might result from insufficient heater power or from the film bypassing the heater due to the geometry.

A calculation of the He\(^4\) mean free path yields information on how
likely the He\(^4\) is to be pumped out with the dilute phase vapor. The He\(^4\) mean free path \(\lambda_4\) is calculated from experimental gaseous He\(^3\)-He\(^4\) diffusion coefficient data by making certain assumptions concerning the application of the calculations to the conditions in the still. Then the He\(^4\) mean free path is compared to the gap width to indicate the feasibility of the design. The operation limits of the refrigerator which indicate the design may work is then shown to be consistent with the general design. The heat input to the still is calculated for the film limiting design and is shown to be acceptable for controlling the temperature of the still.

The mean free path which characterizes the He\(^4\) is for the conditions of He\(^4\) at a temperature of about 2.17 K moving through nearly pure He\(^3\). The temperature of the He\(^3\) gas should be approximately that of the still so that the He\(^4\) is not in thermal equilibrium with the He\(^3\) gas. In addition the vaporization of He\(^4\) may change the pressure and concentration of the nearly pure He\(^3\) gas above the evaporator from the equilibrium conditions in the still. Thus the problem is not an equilibrium problem but calculations from an equilibrium situation may be helpful. The mean free path of gaseous He\(^4\) can be estimated from the gaseous diffusion coefficient \(D_{G34}\). The diffusion coefficient for He\(^3\)-He\(^4\) gas has been measured down to 1.74 K. Both theoretical and experimental work\(^71\) are described by a temperature dependence of the form

\[
\text{n} \mu_{34} D_{G34} = 2.77 \times 10^{-6} \ T^{7/4} \ [\text{gm/cm sec}]
\]

where \(n\) is the number of atoms per unit volume and \(\mu_{34}\) is the He\(^3\)-He\(^4\) reduced mass. The gaseous diffusion coefficient is related to the average hard sphere diameter \(\sigma_{34} = (1/2)(\sigma_3 + \sigma_4)\) by \(^72\)
\[ D_{34} = \frac{3}{8n\sigma_{34}} \left\{ \frac{k_B T (m_3 + m_4)}{2\pi m_3 m_4} \right\} \]

where \( k_B \) is Boltzmann's constant. Neglecting He\(^4\)-He\(^4\) collisions, the average cross-section is related to the mean free path of the He\(^4\) atoms by

\[ \lambda_4 = \left[ 1 + \frac{m_4}{m_3} \frac{\hbar}{\pi n_3 \sigma_{34}} \right]^{-1} \]

so that for a concentration \( X \)

\[ D_{34} = \frac{3\pi}{8} X \lambda_4 \sqrt{\frac{M_4}{\rho}} \left( \frac{RT}{2\pi} \right)^{\frac{1}{2}} \frac{M_3 + M_4}{M_3 M_4} \]

and substituting the experimental temperature dependence for the diffusion coefficient one finds

\[ \lambda_4 = 2.002 \times 10^{-2} T^{1.24} \frac{\text{cm}}{\text{P}} \]

where \( P \) is the pressure in units of \( 10^{-3} \) torr. If the He\(^4\) mean free path is sufficiently long so that most of the He\(^4\) recondenses, then the concentration and pressure in the gap will not be significantly affected by evaporation of He\(^4\). As a first approximation, the temperature is taken as \( T_\Lambda \) and the pressure is taken as the still pressure.

\[ T = T_\Lambda = 2.17 \text{ K} \]

\[ T_S \approx 0.75 \text{ K} \]

\[ P_S \approx 0.129 \text{ torr} \]

\[ \lambda_4 = 4.06 \times 10^{-4} \text{ cm} \]

This first approximation gives a mean free path approximately \( 1/6 \) a gap distance of \( 2.54 \times 10^{-3} \) cm. Figure 8 is a graph of pressure versus temperature for the mean free path calculation. The solid line is for the mean
FIGURE 8. THE DEPENDENCE ON TEMPERATURE AND PRESSURE OF THE MEAN FREE PATH OF He⁴ EVAPORATED IN THE STILL FILM LIMITER

\[ \lambda = 0.1d \]

\[ \lambda = d \]

\[ \lambda < d \]

\[ \lambda = 10d \]

\[ \lambda > d \]

\[ P_s \text{ vs. } T_s \]

FOR \( X_s = 0.01 \)

\[ d = 0.00254 \text{ cm} \]

CALCULATED FROM REF. 71
free path equal to the gap distance. The broken lines indicate a mean
free path which is an order of magnitude higher or lower. Also indicated
on Figure 8 is the temperature $T_\lambda = 2.17$ K which should be the minimum
temperature of the evaporated He$^4$ and a curve representing the still
pressure versus the still temperature for a still concentration of 1%. The
Since the mean free path given by the first approximation is on the order
of the gap distance, the He$^3$ concentration and pressure between the
plates should not be greatly changed from the still conditions. In
addition, since He$^4$-He$^4$ collisions do not contribute to diffusion, it is
not unreasonable to let the pressure be determined by the still
temperature.

The GHe$^3$ between the plates will be in thermal equilibrium with the
GHe$^3$ in the still. Since the He$^4$ mean free path is on the order of the
gap distance, there are few He$^3$-He$^4$ collisions before the He$^4$ reaches the
film. Therefore the temperature of the He$^4$ is more important for the
diffusion process than the He$^3$ gas temperature. The temperature used
can be taken to be $T_\lambda = 2.17$ K. Figure 8 indicates that for $T_S \lesssim 0.55$ K
the mean free path for evaporated He$^4$ is greater than the gap between the
heater and the He$^4$ film. It has not been possible in previous He$^3$-He$^4$
refrigerators to operate the still much below 0.65 K because the con-
struction in the He$^3$ pumping line reduces the flow rates. The throughput
depends linearly on the pressure but it depends on the fourth power of
the diameters through which the gas flows. The He$^3$ pumping line from
the still has a diameter at least 10 times the constriction diameters used
in the best previous refrigerators so that even at pressures 10 times less
than the previous normal operating pressures, the throughput can be $10^3$
times as big.

It is important to remember that while these calculations indicate the design may work, three approximations have been made. The gas diffusion coefficient was extrapolated to temperatures below 1.0 K and may be inaccurate. \( \text{He}^4 \) does not behave like a hard sphere at these low temperatures. It is in fact the strong Van der Waals forces which give rise to the \( \text{He}^4 \) film in the first place. The evaporated \( \text{He}^4 \) is not in thermal equilibrium with the \( \text{He}^3 \) and so estimates of the temperature and pressure which enter the expression for the \( \text{He}^4 \) mean free path have been used. \( \text{He}^4 \) film flow has been widely studied and the results are as widely varied. The condition of the surface over which the film flows causes wide variations in flow rates. However, the \( \text{He}^4 \) film flow is linearly dependent on the smallest cross-section of the film above the liquid and is usually determined by the smallest perimeter along the flow path. The minimum perimeter the film crosses before the gap heater is 3.99 cm. For a clean surface, the average film flow rate per cm of perimeter from 0.4 to 0.8 K is about \( 8 \times 10^{-5} \text{ cm}^2/\text{sec} \), giving an approximate film flow rate for this perimeter of \( 31.9 \times 10^{-5} \text{ cm}^2/\text{sec} \). Taking the density of \( \text{He}^4 \) below 1.5 K to be \( 0.1450 \text{ gm/cm}^3 \), this flow rate is \( 1.16 \times 10^{-5} \) moles/sec. The amount of heat required to vaporize the \( \text{He}^4 \) film is the difference in the enthalpy of \( \text{He}^4 \) between \( T_\lambda \) and \( T_\lambda \) plus the heat of vaporization at \( T_\lambda \). The heat of vaporization of \( \text{He}^4 \) at \( T_\lambda \) is approximately 91 J/mole. The enthalpy difference from 0.75 K to 1.5 K is about 140 J/mole. The enthalpy difference from 1.5 K to 2.17 K is calculated from the saturated vapor pressure specific heat since the difference between the saturated vapor pressure specific heat and constant
pressure specific heat is less than 1% below 2.5 K.76 This enthalpy difference is about 10.3 J/mole. The total enthalpy difference from 0.75 K to 2.17 K and the latent heat of vaporization at $T_\lambda$ is 102.3 J/mole so the heat needed to evaporate the He$^4$ film is about 1.19 mW. The heat leak into the still is calculated to be less than 10 erg/sec. The heat used to evaporate the film is absorbed by the still when the He$^4$ recondenses. This heat is absorbed in the still by the heat of vaporization of the dilute phase and by the cold He$^3$ diffusing into the still from the mixer. The heat of vaporization alone requires approximately 25 J/mole.47

Other still designs have been tried in an effort to limit the He$^4$ film flow.26,36,77 A method in which a disc (placed in a similar position to the stainless steel plate in the present design) was heated did not appear to work.26 It has been suggested that due to convective mixing of the vapor in the still, it is difficult to heat a surfact hot enough to evaporate the film.77 The separation of the disc from the top surface of the still was several millimeters.78 The previous calculation indicates that this is on the order of $10^3$ times the mean free path of the evaporated He$^4$. Either the inability to heat the film due to convective cooling or the mixing of the evaporated He$^4$ with the dilute phase vapor due to the short mean free path could have resulted in the large He$^4$ circulation observed (-20%). Another method in which a 1.19 mm o.d. capillary tube was heated eliminated the convective cooling problem by localizing the heat on a small surface.77 The problem of mixing the evaporated He$^4$ into the pumped dilute phase vapor was eliminated by isolating that part of the vapor containing the evaporated He$^4$ from the
pumped dilute phase vapor with a metallic baffle. This device produced a ratio $\text{He}^3/\text{He}^4$ of about 30. The flow impedance of the pumping line to the still is higher than a still containing a 1 mm orifice since the heated tubing through which the still was pumped had a 1.04 mm i.d. and was 30 mm long. The present design is different from the heated disc method since the heater is insulated from the disc and comes in direct contact with the film. The distance from the heater to a cold surface on which the $\text{He}^4$ can recondense is within an order of magnitude of the mean free path of the evaporated $\text{He}^4$. Finally, the pumping tube to the still is not restricted as in the heated capillary design. The heated tube method has been studied recently for several tube sizes and found to work for larger tubes.36
II.3. Cooling Capacity

The cooling process in the dilution refrigerator can be understood in terms of the enthalpy using approximations valid as the temperature approaches zero Kelvin (T→0). The enthalpy for a pure component $H^0$ at constant pressure is

$$H^0(T) = \int_0^T C^0_P(T) dT$$

where $C^0_P$ is the constant pressure specific heat for a pure component. As T→0, the constant pressure specific heat is approximately equal to the specific heat measured at saturated vapor pressure $C^0_S$ so that the specific heat for the pure component can be written simply as $C^0$. Thus

$$H^0(T) = \int_0^T C^0(T) dT$$

Comparison of the specific heats of the pure components $\text{He}^3$ and $\text{He}^4$ given above in Figure 3 indicates that as T→0

$$C^0_4 \ll C^0_3$$

The dilute phase which is primarily $\text{He}^4$ does have a significant specific heat due to the $\text{He}^3$. Landau and Pomeranchuk\textsuperscript{79} first proposed that for dilute $\text{He}^3$ concentrations, the $\text{He}^3$ would be noninteracting with both the superfluid $\text{He}^4$ and other $\text{He}^3$. The $\text{He}^3$ in dilute solutions was indeed found experimentally to behave as an ideal gas as predicted down to temperatures of 0.2 to 0.5 K depending on the concentration. This can be observed in Figure 3 by noting the region in which the approximately constant pressure specific heat $C_{\mu 4}$ has the classical value of $5/2 \, R$.

Early analysis of the dilution refrigerator was made considering the $\text{He}^3$ to behave entirely as an ideal gas.\textsuperscript{12-14,80} This is not accurate since at some low temperature, the quantum properties of the $\text{He}^3$ Fermi particles
must be considered. The model which allows adequate description of $^3\text{He}$ in dilute solutions is to treat the problem as an ideal Fermi gas in which the effective mass is a function of concentration and reflects the weak interaction which also depends on concentration. The $^3\text{He}-^4\text{He}$ refrigerator has been analyzed quite successfully using this weakly interacting Fermi gas model.\textsuperscript{16-18, 31, 47, 81-83} The specific heat of an ideal Fermi gas is (see for example Ref. 84 and 85)

$$C_V = \frac{\hbar^2}{2} R \frac{T}{T_F}$$

where $R$ is the gas constant and $T_F$ is the Fermi temperature which is related to the Fermi energy $\varepsilon_F$ by Boltzmann's constant $k_B$

$$k_B T_F = \varepsilon_F = \frac{\hbar^2 k_F^2}{2 m^*}$$

where $2\pi\hbar$, $k_F$ and $m^*$ are Planck's constant, the Fermi momentum and the effective mass of the weakly interacting $^3\text{He}$ particles. At $T = 0$, the Fermi sphere contains the translational states of the $N$ particles so that the total number of states $N$ is

$$N = 2 \frac{V}{(2\pi)^3} \frac{4}{3} \pi k_F^3$$

Therefore

$$T_F = \frac{\hbar^2}{2 k_B m^*} \left(3\pi^2 N/V\right)^{2/3}$$

where the number density $N/V$ is

$$N/V = x \frac{N_A}{V_m}$$

where $x$, $N_A$ and $V_m$ are the $^3\text{He}$ concentration, Avogadro's number and the molar volume of the solution. Thus the specific heat of the dilute
solution is

\[ C_3(X,T) = C_V(X,T) = \frac{\pi^2}{2} R T \frac{T}{T_F(X)} \]

During steady state operation of the refrigerator in the ideal case where the noninteracting He\(^4\) forms a stationary background through which the He\(^3\) moves, only the thermodynamic properties of the He\(^3\) in the He\(^4\) need to be considered in the cooling process. The entropy and the enthalpy of the He\(^3\) in He\(^4\) which are just the thermodynamic properties of the ideal Fermi gas are

\[ S_3(X,T) = S_F(X,T) = \int_0^T \frac{C_V(X,T)}{T} \, dT \]

\[ H_3(X,T) = TS_3(X,T) + \mu_3(X,T) \]

where \(\mu_3(X,T)\) is the molar chemical potential of the He\(^3\) in He\(^4\). The enthalpy of the dilute phase will not be calculated using the second equation due to inaccuracies involved in the near cancellation of some rather large terms which are not well known experimentally.\(^47\)

During the dilution process, the He\(^3\) atoms move across the phase boundary at constant pressure. The enthalpy difference gives the amount of cooling available. If the He\(^3\) is cooled in the upper concentrated phase to the temperature of the mixer, then the He\(^3\) diffuses across the phase boundary at constant temperature and the process is reversible. The cooling available can then be calculated from the change in entropy of the He\(^3\) from the concentrated to the dilute phase.

\[ \dot{Q}/\dot{n} = H_D(X,T) - H_C(T) \]

\[ \dot{Q}/\dot{n} = T(S_D(X,T) - S_C(T)) \]
Taking the concentrated phase to be pure $\text{He}^3$ and the dilute phase to be an ideal Fermi gas with effective mass $m^*$, the enthalpy of the dilute phase can be written

$$H_D(X,T) = H^0_3(T) + T[S_F(X,T) - S^0_3(T)]$$

The previous expression is just a result of the equilibrium between the concentrated and dilute phases, i.e. the partial chemical potentials for the $\text{He}^3\quad \mu_3(X,T) = \mu_3(x=1,T) = \mu^0_3$

$$H_D(X,T) = \int_0^T C^0_3(T) + T \int_0^T \frac{C_v(X,T)}{T} dT - T \int_0^T \frac{\mu^0_3(T)}{T} dT$$

For $6 \leq T \leq 50$ mK, the specific heat of pure $\text{He}^3$ is approximately

$$C^0_3 = 3RT = 25T \quad \text{J/mole K}^2$$

As $T \to 0$, $X \to 0.064$ and $m^*/m \to 2.4831$ from which one finds using the equations above

$$T_F(X = 0.064) = 0.382864 \text{ K}$$

$$C_V(X = 0.064)T = 107.2T \quad \text{J/mole(He}^3\text{)} \quad \text{K}^2$$

The enthalpy of the dilute phase for $T \to 0$ is the

$$H_D(X=0.064,T) = H_D(T) = \int_0^T 25TdT + T \int_0^T 107.2 \frac{T^2}{T} dT - T \int_0^T 25 \frac{T}{T} dT$$

$$= 12.5 T^2 + 107.2 T^2 - 25 T^2$$

$$= 94.7 T^2 \quad \text{J/mole K}^2$$

The cooling capacity is then from either the change in entropy or the change in the enthalpy

$$\dot{Q}/\dot{n} = 107.2 T^2 - 25 T^2 = 82.2 T^2 \quad \text{J/mole K}^2$$

For a typical flow rate of $2 \times 10^{-5}$ moles/sec
\[ \dot{Q} = 1.64 \ T^2 \text{ mW/K}^2 \]

If He\(^3\) enters the mixer at a higher temperature, \(T_N\), produced in the last heat exchanger and is subsequently cooled to the mixer temperature, \(T_m\), before diffusion across the phase boundary, then the ratio \(T_N/T_m\) can be calculated by equating the change of enthalpy of the incoming He\(^3\) to the cooling capacity of the mixer. Assuming the T+0 approximations for the thermodynamic functions, and that all other heat leaks to the mixer are negligible, one finds

\[
\frac{\dot{Q}(T_m)}{n} = H_C(T_N) - H_C(T_m)
\]

\[ 82.2 \ T_m^2 = 12.5 \ (T_N^2 - T_m^2) \]

\[ T_N/T_m = 2.75 \]
II.4. Heat Exchanger

It is apparent from a comparison of the characteristics of various He\textsuperscript{3}-He\textsuperscript{4} dilution refrigerators that several heat exchanger designs work reasonably well. The problem of designing the heat exchanger for the dilution refrigerator is not a simple one and has not yet been satisfactorily solved. The physical properties of the liquid heliurums are fairly well known. The most general heat flow equation for the concentrated phase in either a continuous or discrete heat exchanger is found by equating the enthalpy change per unit length to the sources and sinks of heat flow per unit length\textsuperscript{86}

\[
\hat{n}_3 C_3 \frac{dT}{dx} = A \left[ K \frac{d^2 T}{dx^2} + \frac{dK}{dT} \left( \frac{dT}{dx} \right)^2 \right] - \frac{d\sigma}{dx} \int_{T_b}^{T_R} \frac{dT}{R} \pm \eta V_m^2 \hat{n}_3^2 \frac{dz}{dx}
\]

where the three terms on RHS are due to thermal conduction, Kapitza conduction and viscous heating respectively and where \( \hat{n}_3 \) is the molar flow rate, \( C_3 \) is the specific heat, \( A \) is the cross-sectional area of the liquid, \( K \) is the liquid thermal conductivity, \( d\sigma/dx \) is the surface area per unit length, \( R \) is the Kapitza resistivity, \( \eta \) is the viscosity, \( V_m \) is the molar volume and \( dz/dx \) is the flow impedance per unit length. The heat exchanger material has a thermal conduction much greater than the Kapitza conduction so that thermal gradients in the heat exchanger body are neglected. There is a similar equation for the dilute phase. The heat flow out of the concentrated phase is equal to the heat flow into the dilute phase

\[
\hat{n}_3 C_3 \frac{dT_C}{dx} = \hat{n}_3 C_3(X) \frac{dT_D}{dx}
\]

where \( X \) is the He\textsuperscript{3} concentration in the dilute phase and \( dT_D/dx \) and
\( \frac{dT_D}{dx} \) are the temperature gradients in the concentrated and dilute phases respectively. These equations together with the physical properties of helium solutions and the fixed experimental parameters are sufficient to allow the temperature distribution in the heat exchanger system to be determined. The only temperature which can be varied experimentally in achieving low mixer temperatures is the still temperature \( T_S \). To find a complete solution including all parameters affecting the heat exchange is a difficult problem which has not yet been solved for either continuous or discrete heat exchangers.

The present discussion is directed at the problem of discrete heat exchangers rather than continuous heat exchangers since the discrete or step heat exchanger provides better heat transfer at lower temperatures.\(^{19,35}\) Less effort has been made in solving the complete problem of the continuous heat exchanger even though this type of heat exchanger is perhaps more important at higher temperatures in the dilution refrigerator.\(^{22,36}\)

In the general solution of the equations written above, the temperature distribution is a function of \( x \) (the distance along the heat exchanger module) and cooling and heating effects may extend beyond the modular heat exchanger due to thermal conduction and viscous heating in the liquid helium phases. At temperatures above about 10 or 20 mK, viscous heating effects are small.\(^{36,87}\) This term can be neglected in describing the heat exchangers. The problem can be further simplified by eliminating the detailed temperature distribution by disregarding the effects of the liquid thermal conductivity. While neglecting the liquid thermal conductivity is certainly incorrect, it allows an approximate solution to be obtained from which certain physical characteristics of
the heat exchanger may be deduced.

The problem is to determine the temperature distribution of a heat exchanger system consisting of N modules between the still and the mixer when only the still temperature and properties of liquid helium are known. For the notation on the temperature distribution, see Figure 9. T₁ and T₃ are the inlet temperatures of the concentrated and dilute phases respectively. T₂ and T₄ are the outlet temperatures of the concentrated and dilute phases respectively. The heat flow between the two phases at low temperatures is limited by the Kapitza thermal boundary resistance and not the thermal conductivity of the heat exchanger body material. At low temperatures the heat exchanger body will be in thermal equilibrium at a temperature T₅. Since the concentrated phase is the hotter phase

\[ T₁ > T₂ > T₅ > T₄ > T₃ \]

The three heat flow equations given above yield 3N equations for N heat exchanger modules. The temperatures of various modules are related

\[ T_{n1} = T_{n-1,2}, \quad n = 2, N \]
\[ T_{n4} = T_{n-1,3}, \quad n = 2, N \]

This provides an additional 2N-2 equations. The input to the first heat exchanger is taken to be Tₛ which is specified

\[ T₁₁ = Tₛ \]

The output of the concentrated side in the bottom heat exchanger module is related to the mixer temperature Tₘ as shown in Section II.3. The relation is actually temperature dependent and can be obtained more accurately from Table 16 in Ref. 47. The mixer temperature is taken as
FIGURE 9. HEAT EXCHANGER SYSTEM
the input temperature on the dilute side of the lowest heat exchanger

\[ T_{N2} = 2.75 \quad T_m = 2.75 \quad T_{N3} \]

Thus there are a total of 5N equations.

The equations necessary for the solution of the N module heat exchanger taking into account heat transfer are not difficult to derive but the solution of the 5N transcendental equations is quite difficult. One can simplify the problem by solving for the temperature distribution of N ideal heat exchange modules. An ideal heat exchange module is one that provides the necessary heat transfer so that the two outlet temperatures are equal and therefore equal to the heat exchanger body temperature, i.e. \( T_2 = T_4 = T_5 \). For the ideal heat exchanger, the detailed temperature distribution is unimportant as in the case of a real heat exchanger if the liquid thermal conductivity is neglected. By balancing the change in enthalpy in each heat exchanger module with the boundary conditions simplified by \( T_2 = T_4 = T_5 \), the 3N equations are sufficient to solve the ideal heat exchanger. Both the ideal and the real heat exchanger problem must be solved by either a graphical or numerical iterative process.

It is important to note how the physical data has been represented in various solutions to the problem. It was mentioned above that solutions have been obtained by both graphical and numerical iterative processes. The difficulty in representing the physical properties is that there is no simple temperature dependence over the entire temperature range of interest. Some authors have used a graphical iterative process in which the physical properties of interest are obtained from a graph. This works well for obtaining enthalpy differences in ideal heat exchangers but cannot be extended to real heat exchangers where the Kapitza thermal
boundary resistance enters. Another approach has been to do the numerical integrations necessary by fitting the necessary data over a small temperature region with a low order polynomial. This approach would allow the solution over the entire temperature range for real heat exchangers to be obtained. An alternative approach which has been adopted in the present work is to fit the necessary physical data over a wide temperature range with a higher order polynomial. There is no disadvantage in having more terms to evaluate since it is done on a computer. Some accuracy is traded however in keeping the number of terms to a minimum in the expression for the specific heat of the dilute solution. The power series fit to the experimental data for the specific heat of pure He\textsuperscript{3} is taken from Radebaugh\textsuperscript{47}

\begin{align*}
0 \leq T \leq 0.2 \text{ K} & \quad c^0_3 = 25.333T - 87.475T^2 + 284.618T^3 - 1097.33T^4 + 1903.57T^5 \text{ J/mole K} \\
0.2 < T \leq 0.5\text{ K} & \quad c^0_3 = -1.2846 + 48.3061T - 228.9698T^2 + 556.0303T^3 - 666.821T^4 + 315.643T^5 \text{ J/mole K} \\
0.5 < T \leq 2 \text{ K} & \quad c^0_3 = 2.0914 + 3.948T - 4.5068T^2 + 3.37857T^3 - 0.68783T^4 \text{ J/mole K}
\end{align*}

The specific heat of the dilute phase has been evaluated by Radebaugh\textsuperscript{47} and is a function of the mixer temperature due to its dependence on concentration. Thus to be strictly correct in evaluating the mixer temperature one would have to adjust the specific heat at constant chemical potential by an iterative method until the solution for T\textsubscript{m} was obtained by the specific heat determined by T\textsubscript{m}. This process complicates the solutions so that the initial approach has been to take the values of the specific heat at constant chemical potential determined by T\textsubscript{m} = 0.010 K
and fit these values with a power series expression. The error for $T_m \leq 50$ mK is less than 5%.

\[
\begin{align*}
0 \leq T \leq 0.2 \text{ K} & \quad C_4 = 107.22046T - 30.939288T^2 + 10597.364T^3 \\
& \quad - 122474.15T^4 + 298785.25T^5 + 913259.34T^6 \\
& \quad -3496609.9T^7 \quad \text{J/mole K}
\end{align*}
\]

\[
\begin{align*}
0.2 \leq T \leq 0.9 \text{ K} & \quad C_4 = 6.6451662 + 104.59628T - 294.67706T^2 \\
& \quad +315.67187T^3 + 94.288951T^4 - 434.60441T^5 \\
& \quad + 243.70703T^6 \quad \text{J/mole K}
\end{align*}
\]

Simple expressions for the Kapitza thermal boundary resistance are particularly important in the present method of solution. In order to allow integrations to be made in the heat balance equation, the expressions for the thermal boundary resistance are required to be a single term which is a function of an integral power of the temperature and to have constant coefficients over the same temperature range for which the specific heat coefficients are constant. The experimental data\textsuperscript{19,88} can be fit rather nicely with single terms if the power of the temperature is non-integral. However, the requirements above restrict the agreement of the expressions to the experimental data. The expressions overestimate the thermal boundary resistance except at higher temperatures for the He\textsuperscript{3} data. Since the thermal boundary resistance is smaller at higher temperatures and the actual thermal boundary resistance in a heat exchanger may be far from ideal, the expressions given below are considered to be a reasonable approximation. For the concentrated phase, the thermal boundary resistance $R = \Delta T/\dot{Q}$ is

\[
R_{3Cu} = 1.988 \times 10^{-2}T^{-3} \frac{m^2 s K^4}{J} \quad 0 \leq T \leq 0.2 \text{ K}
\]
\[ R_{34Cu} = 7.952 \times 10^{-4} t^{-5} \frac{m^2 s K^6}{J} \quad 0.2 \leq T \leq 0.9 \text{ K} \]

For the dilute phase, the thermal boundary resistance is

\[ R_{34Cu} = 6.356 \times 10^{-3} t^{-3} \frac{m^2 s K^4}{J} \quad 0 \leq T \leq 0.2 \text{ K} \]
\[ R_{34Cu} = 1.27 \times 10^{-3} t^{-4} \frac{m^2 s K^5}{J} \quad 0.2 \leq T \leq 0.9 \text{ K} \]

All the data just discussed can be represented at very low temperatures quite accurately by simple expressions.

\[ R_{3Cu} = 1.988 \times 10^{-2} t^{-3} \frac{m^2 s K^4}{J} \quad 0 \leq T \leq 0.136 \text{ K} \]
\[ R_{34Cu} = 6.356 \times 10^{-3} t^{-3} \frac{m^2 s K^4}{J} \quad 0 \leq T \leq 0.3 \text{ K} \]

\[ C_m = 107.2 \text{ T} \quad \text{J/mole K}^2 \quad 0 \leq T \leq 0.05 \text{ K} \]
\[ C_m^0 = 25 \text{ T} \quad \text{J/mole K}^2 \quad 0 \leq T \leq 0.05 \text{ K} \]

The initial work on the heat exchanger problem including heat transfer properties was done using the very low temperature approximations. Neglecting liquid thermal conductivity and viscous heating, the heat flow equation given above becomes

\[ \dot{m}_3 C_3 \frac{dT}{dx} = \frac{d}{dx} \int_0^T T \frac{dT}{R_3} \]

Using the temperature dependent helium properties

\[ 25A_3 T dT = -d \sigma \int_T^\infty \frac{T^3 dT}{1.988 \times 10^{-2}} \]
For the concentrated phase, $T_5 < T$

$$\frac{T \Delta T}{T^4 - T_5^4} = -\frac{d\sigma}{1.988 \hat{n}_3}$$

If the integral is carried out over an entire heat exchanger, then from

$$\int_{T_1}^{T_2} \frac{T \Delta T}{T^4 - T_5^4} = -\int_0^{\sigma_C} \frac{d\sigma}{1.988 \hat{n}_3}$$

one obtains

$$\frac{4T_5^2 \sigma_C}{1.988 \hat{n}_3} = \ln\left(\frac{T_1^2 - T_5^2}{T_1^2 + T_5^2} \cdot \frac{T_2^2 + T_5^2}{T_2^2 - T_5^2}\right)$$

A similar equation is obtained for the heat flow balance on the dilute phase side.

$$\hat{n}_3 C_{m} \frac{dT}{dx} = -\frac{d\sigma}{dx} \int_{T_3 R_{34}}^{T} \int_{T_5}^{T_3} \frac{dT}{dx}$$

$$107.2 \hat{n}_3 T \Delta T = -\frac{d\sigma}{dx} \int_{T_5}^{T} \frac{T^3 dT}{6.356 \times 10^{-3}}$$

For the dilute phase, $T_5 > T$

$$\frac{T \Delta T}{T_5^4 - T^4} = -\frac{d\sigma}{2.725 \hat{n}_3}$$

If the integral is done over a heat exchanger module, then

$$\int_{T_3}^{T_4} \frac{T \Delta T}{T^4 - T_5^4} = \int_0^{\sigma_d} \frac{d\sigma}{2.725 \hat{n}_3}$$
\[
\frac{kT_5^2 \sigma d}{2.725 \dot{n}_3} = \ln \left( \frac{T_5^2 + T_4^2}{T_5^2 - T_4^2} \right) \frac{T_5^2 - T_3^2}{T_5^2 + T_3^2}
\]

The total heat flow from the concentrated side equals the heat flow into the dilute side.

\[
\int_{T_2}^{T_1} \dot{n}_3 C_3 dT = \int_{T_3}^{T_4} \dot{n}_3 C_{\eta 4} dT
\]

\[12.5 (T_1^2 - T_2^2) = 53.6 (T_4^2 - T_3^2)\]

Each of the three equations just derived can be written for each heat exchanger module. The outlet temperature of one module is taken to be the inlet temperature of the next module for a given phase so that

\[T_{n1} = T_{n-1,2}\]

\[T_{n4} = T_{n-1,3}\]

The relation between the mixer temperature \(T_m = T_{N3}\) and the temperature to which the concentrated phase is cooled in the last heat exchanger \(T_{N2}\) was given in Section II.3. and was approximated here as

\[T_{N2} = 2.8 T_m = 2.8 T_{N3}\]

If some initial temperature of the concentrated phase is specified, then the equations can be solved for the various temperatures. Results were obtained but it was soon apparent that the very low temperature approximations could not provide all the physical insight needed to describe the complete heat exchanger system. The decision to change approaches was
also influenced by the relative inefficiency of the computer program
used at this stage. These results, however, do provide some interesting
observations about the modular heat exchanger at very low temperatures.

The mixer temperature obtained for various values of the total
surface area on each side of the modular heat exchanger is shown in
Figure 10. The surface area on the dilute side is equal to the surface
area on the concentrated side. The mixer temperature is plotted as a
function of this surface area divided by the molar flow rate (hereafter
referred to as a reduced surface area). Thus if one increases the molar
flow rate, the surface area must be increased by the same fraction. As
the reduced surface area is increased, the mixer temperature approaches
a limiting value which cannot be decreased by increasing the reduced
surface area. Radebaugh obtained for the case of zero liquid thermal
conductivity the same value of the mixer temperature with the same surface
area on the dilute side (~4.2x10^7 cm^2 s/mole) but with only half the
surface area on the concentrated side (~2x10^7 cm^2 s/mole). Radebaugh
studied the ratio of the surface area on the dilute side and the
concentrated side and found the relation

\[
\frac{a_d}{a_C} = 2.1
\]

yielded the highest heat transfer in the lowest temperature module. It
is interesting to note that one can get maximum heat transfer without
knowing the optimum surface area ratio but the flow impedance will not be
a minimum. It is also important that one should build the modular heat
exchangers with the optimum surface ratios only if both sides have
sufficient surface area for the maximum heat transfer and minimum mixer
FIGURE 10. THE MIXER TEMPERATURE DEPENDENCE ON THE SURFACE AREA OF THE LAST HEAT EXCHANGER FOR AN INPUT TEMPERATURE OF 60.0 mK WITH $\sigma_c = \sigma_d$
temperature. This is illustrated in Figure 11 in which the mixer temperature is plotted as a function of the reduced surface area ($\xi_d/\xi$) on the dilute side for a surface area ratio

$$\frac{\sigma_d}{\sigma_C} = 2.0$$

The lowest temperature point on the curve was calculated to be 8.4 mK. The mixer temperature can be reduced to 8.3 mK by increasing just the surface area on the concentrated side so that $\sigma_d = \sigma_C$. Although it was not calculated, the optimum surface area ratio should provide for the limiting temperature once both surface areas are sufficiently large. Therefore, the utility in using the optimum surface area ratio is in minimizing material and flow impedance but is valid for optimizing heat transfer only when the surface area on each side is sufficiently large for the optimum heat transfer to occur.

The limiting surface areas for various input temperatures are given in Figure 12. As might be expected because of the dependence of the heat transfer on surface area, the minimum surface area for optimum heat transfer increases for operation at lower temperatures. Figure 13 indicates the minimum surface area on the dilute side which allows the limiting mixer temperature to be reached.

Various calculations indicate that for a given total surface area, one obtains lower temperatures if two smaller modules are used rather than one large one. For example, with a reduced area of 8.19x10^7 cm^2 sec/mole on each side of a module, a mixer temperature of 8.3 mK is obtained whereas two modules each having reduced surface areas of 4.1x10^7 cm^2 sec/mole allow a mixer temperature of 5.5 mK to be obtained.
Figure 11. The mixer temperature dependence on the surface area of the last heat exchanger near the optimum ratio $\sigma_d/\sigma_c = 2.1$ and for $T_1 = 50$ mK.
THE MIXED TEMPERATURE DEPENDENCE ON THE SURFACE AREA OF THE LAST HEAT EXCHANGER FOR VARIOUS INPUT TEMPERATURES TO THE LAST HEAT EXCHANGER

Figure 12.
FIGURE 13. THE SURFACE AREA OF THE LAST HEAT EXCHANGER ($\sigma_c = \sigma_d$) FOR WHICH THE MIXER TEMPERATURE REACHES ITS LIMITING VALUE.
This is true even though the smaller heat exchangers in the example are not of sufficient size to obtain maximum heat transfer.

The main purpose of the present study is to determine how the heat exchange system depends on the surface area and how this surface area should be arranged. The low temperature calculations indicated that it was important to the design of efficient heat exchanger systems in dilution refrigerators to know how to break the total surface area up into modules. Figure 14 indicates the resulting mixer temperatures as one continues to add heat exchanger modules of the same surface area. One obtains lower temperatures with the addition of each module. Nevertheless, the discussion above indicates that limiting temperatures are not being reached since the lower temperature modules need to have larger surface area. Additional calculations in which the mixer temperature of the eight module system was reduced from 12.8 mK to 8.3 mK by increasing the surface area of each module by a factor of four indicate that the surface area used in the calculations for Figure 14 was indeed not sufficient for optimum heat transfer. The very low temperature calculations confirm the physical argument that the increasing Kapitza resistance to heat flow at lower temperatures requires increasing surface area for optimum heat flow. The poor results of adding additional heat exchanger modules with small effective heat exchange surfaces can be understood by the preceding discussion which requires larger surface areas at lower temperatures.

In order to test the power series fits to the heat capacity of both helium phases in the heat exchange system, an analysis of a set of perfect heat exchanger modules was carried out. The results are shown
FIGURE 14. THE MIXER TEMPERATURE DEPENDENCE ON THE NUMBER OF EQUAL AREA MODULES WHEN THE SURFACE AREA IS NOT SUFFICIENT FOR MAXIMUM HEAT TRANSFER. $T_1 = 50$ mK

$\sigma_c/\dot{h} = \sigma_d/\dot{h} = 6.4 \times 10^5$ cm$^2$ sec/mole
in Figure 15 and compared to the results obtained by Radebaugh\(^{86}\) by a different numerical technique. The agreement of the mixer temperatures is reasonably good. The value of the mixer temperature obtained for six perfect heat exchanger modules is 0.825 the value obtained by Radebaugh. The numerical estimates of Pipes\(^{31}\) are not correct since his estimate of the enthalpy of the dilute solution was based on the enthalpy along the solubility curve. The single numerical result of Wheatley\(^{19}\) is in reasonable agreement with the present work and that of Radebaugh.

The results for the perfect heat exchange system indicate that the power series expressions are accurate enough to find the dependence of the mixer temperature on the arrangement of the surface area in a real heat exchange system.

Radebaugh\(^{86}\) has included the effects of the liquid He\(^3\) and dilute solution thermal conductivity in the analysis of the real heat exchanger. The results indicate that this thermal conductivity is not only important to the detailed temperature profile but also to the efficiency of the heat exchanger module. For real heat exchangers of the same size, the temperature change of a phase is much larger for zero liquid thermal conductivity than for a finite thermal conductivity. At first sight, one might expect the finite thermal conductivity to improve the heat transfer process but instead it reduces the heat transfer since in effect, it provides a thermal short between the higher and lower temperatures in the phase. It is the same problem that occurs in the body of the heat exchanger module since it also is an effective thermal short. Radebaugh estimates that \(\ell/A > 100\) cm\(^{-1}\) (recall that \(\ell\) is the length of the heat exchange surface and \(A\) is the liquid cross-sectional area) in order for the liquid thermal conductivity not to provide an effective
FIGURE 15. THE DEPENDENCE OF THE MIXER TEMPERATURE ON THE NUMBER OF PERFECT HEAT EXCHANGER MODULES

- Present work \(T_s = 0.70 \text{ K}\)
- Ref. 86 \(T_s = 0.70 \text{ K}\)
- Ref. 18 \(T_s = 0.73 \text{ K}\)
thermal short. The solution for $\frac{\lambda}{A} \approx 1 \text{ cm}^{-1}$ already behaves like the case $\frac{\lambda}{A} \ll 1 \text{ cm}^{-1}$ where no temperature gradients exist due to thermal conductivity. Thus, one should make the heat exchanger modules either extremely long, $\frac{\lambda}{A} \gg 1 \text{ cm}^{-1}$, for high efficiency or extremely short, $\frac{\lambda}{A} \ll 1 \text{ cm}^{-1}$, if the flow impedance problems in the long modules cannot be overcome.

Further analysis of the dependence of the heat exchanger on surface area distribution among modules should include the effects of the liquid thermal conductivity. These calculations have not yet been performed but would provide valuable information on the properties of low temperature heat exchangers.
III. APPARATUS

III.1. Liquid Helium-4 Bath

Figure 16 is a drawing of the nested dewar set. The dewar was designed to obtain long running times by having a volume of LHe\textsuperscript{4} above the refrigerator of about 4 liters while minimizing the heat leak to the He\textsuperscript{4} bath by using a reasonably narrow neck. The heat leak into the bath by radiation from surfaces at room temperature was reduced by five polished brass discs which were tightly clamped to the still pumping line. These discs not only acted as radiation shields but provided good heat transfer between the cold He\textsuperscript{4} gas being pumped out of the dewar and the still pumping tube. The tail section of the dewar set was designed to fit a 2" gap in an electromagnet. With the refrigerator cooling from 77 K to 4.2 K without using an exchange gas, the first LHe\textsuperscript{4} transfer of about 12 \& lasted 38.5 hours before falling below the top of the vacuum can. A second transfer of about 6 \& while still cooling to 4.2 lasted about 42.5 hours. The LHe\textsuperscript{4} bath was pumped by a Kinney DVD 8810 (110 cfm) vacuum pump to a temperature of 1.2 K. It is important to pump the bath down at the right speed in order to obtain the maximum amount of liquid helium at 1.2 K. The procedure used was to open the needle valve to the bath as fast as was possible without causing pressure oscillations above the bath. The gate valve was opened its minimum amount only when the rate of pressure change was nearly zero which occurs at pressures of about 10 torr. The pressure was reduced above the bath to its lowest value before the gate valve was fully opened. Using this procedure, about 2.6 \& were available at temperatures below the \(\lambda\) transition giving running times of about 20 hours.
Figure 3.1-1. Nested Dewar Set:
Outside - N₂
Inside - He⁴
Dewars are silvered with 1/2" slits on N₂ Dewar and 1/4" slits on He⁴ Dewar

Total length of He⁴ Dewar is 49"
III.2. Circulation and Gas Handling System

Figure 17 shows the He$^3$ circulation and gas handling system. The He$^3$ was pumped from the still by a 4" diffusion pump containing Dow Corning 704 diffusion pump fluid. For a higher circulation rate, a diffusion pump oil with a higher viscosity can be used. The backing forepump was a specially sealed Welch 1402 KGB vacuum pump.\textsuperscript{89} The circulation rate can also be increased by putting another 1402 KGB in parallel with the first one. The circulation rate can be measure by observing the time rate of change of the pressure in the known volume of tank 1.\textsuperscript{90} The pressure was measured by a TI Precision Pressure Bourdon Gauge with an accuracy of better than ± 0.02 torr. The TI gauge was also used to accurately measure the pressure in the still and gas return. The pressure measurement in the still was made via a 1/8" OD x 0.010" wall stainless steel tube inside the main pumping line so that there was no pressure gradient due to gas flow. The pressure was monitored during the initial high vacuum cleaning and during the refrigerator startup and operation by various Bourdon, thermocouple and Phillips ionization gauges (indicated by the initials in a small circle on Figure 17). Most of the valves were Nupro B-4H bellows-sealed valves. The bellow-sealed valves between the diffusion pump and the mechanical pumps were 1.5" Veeco FL 150S and the bellow-sealed valve between the gas return line and the Welch 1402 was a .5" Veeco FL38S. The check valve was opened by a pressure differential and allowed the tank 2 to be evacuated and then used as a safety tank in case there was a pressure build-up in the gas return while the refrigerator was unattended. This check valve was a
FIGURE 17. $^3\text{He}$ CIRCULATION AND GAS HANDLING SYSTEM
Circle Seal 559B-2MP-5. It was found to be necessary since in an early attempt at running the refrigerator the normal evaporation of He\textsuperscript{3} in the condenser line as the bath level dropped a small amount overexpanded a bellows in the gas return line necessitating its replacement. The needle valve connecting the vacuum can with the He\textsuperscript{4} bath was a Whitey 0IRS2 and was used both for leak detection and temperature calibration. The gate valve above the diffusion pump was a CVC VCS41-B. The purpose of the various valves will become apparent when the various procedures necessary to the operation of the refrigerator are described. Tank 3 was initially charged with 4\% of pure He\textsuperscript{3} and was filled first upon shutting down the refrigerator so that it contained primarily He\textsuperscript{3}. Tanks 2 and 4 were initially charged with approximately 31.7\% with He\textsuperscript{4} from a high pressure He\textsuperscript{4} cylinder of nominal purity. During charging and during the operation of the refrigerator, the gas was passed through a liquid nitrogen cooled charcoal trap. This trap was pumped out at room temperatures between runs. The cold trap above the diffusion pump was a NRC 0315-1-004 cooled by a 1/3 H.P. freon refrigeration unit set to maintain a temperature of about -35°C in the baffle.

The sample gas handling system is given in Figure 18. It consisted primarily of a Toeppler pump with suitable valves for its operation. The volume of the left side of the Toeppler pump was accurately known so that an accurate amount of He\textsuperscript{4} gas could be condensed in the sample cell via a LN\textsubscript{2} trap made of approximately 3 m. of 1/8" copper refrigeration tubing coiled in a dewar.

The He\textsuperscript{3} pumping line from the flange which mates to the He\textsuperscript{4} bath pumping tee consisted first of a 35 mm i.d. He\textsuperscript{3} pumping tee with an
FIGURE 18. SAMPLE GAS HANDLING SYSTEM
effective pumping length of about 12 cm. The next 36.8 cm length of pumping line had a 27 mm i.d. This was followed by a 44 mm i.d. tee directly above the 4" gate valve. The effective pumping length of the 7.62 cm tee was approximately 15.5 cm. Connecting the diffusion pump to either mechanical pump was a 38 mm i.d. line. The remaining connections in both the circulation and gas handling systems were 4.72 mm i.d. Cu refrigeration tubing.
III.3. Still

The significant difference in this refrigerator from other dilution refrigerators is in the design of the still. The still was designed to maximize the amount of He$^3$ circulated by incorporating a method which should have limited the amount of He$^4$ film entering the still pumping tube. The still was constructed out of brass and put together with indium O-rings as shown in Figure 19. A resistor thermometer and a heater coil were attached to the outside of the still. The heat leak into the still due to radiation from surfaces at room temperature was minimized by the upper optical baffle.\textsuperscript{19} The pumped gas cooled the lower baffle so that the heat leak down the stainless steel pumping tube was reduced. The film evaporator consisted of one turn of AWG 36 nichrome wire which was set in a grooved nylon ring pressed into the inside stainless steel plate. The inside top surface was precisely machined with a 2.54x10$^{-3}$ cm recess parallel to the surface which mates the stainless steel plate. The stainless steel plate was pressed against the mating surface and held by a stainless steel nut on the threaded lower end of the pumping tube. The lower plate was made as thin as possible to limit heat conduction to the film but sufficiently thick to be rigid. Very little heat was conducted along the hylon ring and stainless steel plate so that the evaporator was a very localized source of heat. The evaporator leads were brought through a hole in the stainless steel plate and epoxied. The leads then went up the He$^3$ pumping tube to room temperature electrical feedthroughs.
FIGURE 19. STILL
III.4. He\textsuperscript{3} Condenser and Still Heat Exchanger

The He\textsuperscript{3} gas returned from the external pumping system via a 5.84 mm i.d. stainless steel 304 tube to a 1 m length of 0.65 mm i.d. x 0.076 mm wall Cu-Ni 70-30 capillary coiled in the pumped LHe\textsuperscript{4} bath which acted as a heat exchanger to precool the He\textsuperscript{3} gas. The precooler was connected directly into a sintered silver condenser which was mounted in the vacuum can on the vacuum can flange. The sintered core condenser was similar to half a modular heat exchanger described below and was in mechanical contact with the LHe\textsuperscript{4} bath. The LHe\textsuperscript{3} flowed from the condenser through a flow impedance to the still heat exchanger which consisted of 104.2 cm of 0.30 mm i.d. x 0.05 mm wall Cu-Ni 70-30 capillary. This heat exchanger precoalled the LHe\textsuperscript{3} to the still temperature before it entered the modular heat exchanger system.
III.5. Flow Impedance

In order to maintain the liquid phase everywhere in the concentrated phase side of the refrigerator, it is necessary to adjust the flow impedance. The pressure everywhere in the concentrated phase must be greater than or equal to the vapor pressure of LHe\textsuperscript{3} at the given temperature. Since the LHe\textsuperscript{4} bath temperature is about 1.2 K, the condensing pressure of the LHe\textsuperscript{3} must be greater than approximately 200 torr. The vapor pressure in the still is less than 0.1 torr. In order to have stable liquid levels, there must be a pressure drop resulting from the impedance to flow from the condenser to the dilute phase in the still equal to the backing pressure on the condenser. The still temperature can be from about 0.5 K to about 0.8 K depending on the operating characteristics of the refrigerator. The LHe\textsuperscript{3} in the still heat exchanger must have a pressure greater than 0.16 to 2.9 torr depending on the temperature. Therefore a fraction of the total pressure drop described above must occur following the still heat exchanger. The characteristic temperature of the LHe\textsuperscript{3} after passing through the first heat exchanger is approximately 0.3 K with a corresponding vapor pressure of about 2 millitorr. Thus it is unnecessary to provide additional flow impedance in the heat exchange system. It is advantageous to minimize the flow impedance in the heat exchanger system and mixer so that viscous heating is minimized. Therefore two flow impedances are introduced into the returning LHe\textsuperscript{3} line between the condenser and still heat exchanger and between the still heat exchanger and the first heat exchanger module. The size of the flow impedance needed can be determined from the definition of flow impedance.
\[
Z = \frac{\Delta P}{\eta \dot{V}} = \frac{\Delta P}{\eta V_m n}
\]

provided one knows the pressure drop \( \Delta P \), the viscosity of \( \text{LHe}^3 \) \( \eta \) and the volume flow rate \( \dot{V} \) which can be written as the product of the molar volume \( V_m \) and the molar flow rate \( \dot{n} \). The molar flow rate is constant throughout the circulation system and depends on the condensing rate of the condenser, the capacity of the pumps and the total flow impedance of the circulation system.

Assuming that the condenser has sufficient capacity and that the pumping system is limited by the mechanical pump, the maximum molar flow rate in the present system was approximately \( 2 \times 10^{-5} \) mole/sec. For \( \Delta P \geq 20 \) torr, the total flow impedance should be greater than \( 1.3 \times 10^{12} \) cm\(^{-3}\). With no artificial impedances in the refrigerator, the flow impedance on the order of \( 10^9 \) cm\(^{-3}\) is insufficient. For the same minimum flow impedance found above, the molar flow rate can be increased by increasing the backing pressure on the condenser provided there is nothing limiting the flow rate elsewhere in the circulation system. However, if the flow impedance is designed for \( \Delta P = 20 \) torr, then the flow rate can be increased by a factor of 38 before the backing pressure on the condenser exceeds atmospheric pressure and is harmful to the mechanical pump. Thus, there is a limit of \( 7.6 \times 10^{-4} \) moles/sec unless the total flow impedance is decreased. It is also advantageous to decrease the total impedance slightly so that in obtaining larger flow rates, it is not necessary to use a relatively large condenser backing pressure which requires more \( \text{He}^3 \).
Since the pressure drop following the still heat exchanger is at most 15% of the total pressure drop, the impedance following the still heat exchanger is an order of magnitude smaller and can be dealt with independently. The pressure drop of 0.16 to 2.9 torr which the second impedance must produce depends on the operating temperature of the still. The design must be for the largest value of the minimum pressure drop which may be encountered during operation of the refrigerator. If the refrigerator is operated with the still at a lower temperature then the pressure of the LHe\textsuperscript{3} in the still heat exchanger will be greater than its vapor pressure. For a pressure drop of 2.9 torr and a molar flow rate of 2x10\textsuperscript{-5} torr the flow impedance should be approximately 1.35x10\textsuperscript{11} cm\textsuperscript{-3}.

The refrigerator was designed to allow for higher flow rates to take advantage of the film limiting design. However initial operation of the refrigerator has been with a rather minimal circulation pumping system. Impedance I between the condenser and the still heat exchanger consisted of 1.52 m of nominal 0.1 mm i.d. stainless steel 304 capillary tube. The impedance per unit length was measured to be 3.8x10\textsuperscript{9} cm\textsuperscript{-4} so that $Z_L \approx 5.8x10\textsuperscript{11}$ cm\textsuperscript{-3}. This value is about one-half the value calculated as needed for the assumed pressure drop of 20 torr and molar flow rate of 2x10\textsuperscript{-5} moles/sec. Impedance II following the still heat exchanger consisted of approximately 17.3 cm of nominal 0.05 mm i.d. stainless steel 304 capillary tube. The impedance per unit length was measured to be 7.3x10\textsuperscript{9} cm\textsuperscript{-4} so that $Z_{II} \approx 1.3x10\textsuperscript{11}$ cm\textsuperscript{-3}. This value is approximately equal to the value calculated as needed for the assumed molar flow rate of 2x10\textsuperscript{-5} moles/sec and pressure drop of 2.9 torr.
corresponding to a rather high still temperature.

Once a given impedance is used in the refrigerator, the molar flow rate and pressure drop will adjust until the refrigerator has achieved steady state operation such that

$$\frac{\Delta P}{n} = Z\eta V_m$$

If the impedance is too small and the flow input is limited upstream, then the impedance may pass the LHe$^3$ fast enough to empty the condenser or still heat exchanger of liquid. This is not likely in the case of impedance II because of its high impedance. However, with the present pumping system, it is possible in the case of impedance I. If the condenser is not full of liquid it will place a large heat load on the still as a result of the latent heat of condensation. In the present operation of the refrigerator impedance I appears to be large enough. With the LHe$^3$ return temporarily interrupted, the vapor pressure on the concentrated side indicates that condensation is occurring in the condenser as it should.
III.6. Interconnections

The capillary leading from the still heat exchanger to impedance II and then to the concentrated side of the first modular heat exchanger and subsequently to the second and third heat exchanger on the concentrated side was 0.30 mm i.d. x 0.05 mm wall Cu-Ni 70-30 capillary tube. The remaining connections in the heat exchanger system and to the mixer and still were 0.65 mm i.d. x 0.076 mm wall Cu-Ni 70-30 capillary. Increasing the size of the tubes on the concentrated side of the lower heat exchangers was probably an unfortunate choice since it has been subsequently shown that an increase from 0.27 mm to 0.64 mm of the bore of the tube connecting the concentrated side of the lowest heat exchanger to the mixer results in an increase in temperature.\textsuperscript{35} The connections between heat exchanger modules were approximately 5 cm long. The connection between the still and the dilute phase side of the first heat exchanger was 8 cm long. The capillary tube was looped once between each heat exchanger connection to minimize the length required for the entire heat exchanger system.
III.7. Heat Exchanger

The various types of heat exchangers and the need for large surface area have been discussed above. The heat exchanger consisted of five modular heat exchangers in a vertical arrangement between the still and the mixer. One heat exchanger module is shown in Figure 20. All previous modular heat exchangers have been made of Cu.\textsuperscript{54,91-94} The body of the heat exchanger module and the sintered cores were made of high purity silver in an attempt to improve heat transfer. The thermal conductivity of pure annealed silver is approximately twice that of pure annealed copper at 4.2 K.\textsuperscript{95} The Kapitza conductance of silver is larger than that of Cu although for comparable surfaces, this difference may be less than a factor of two.\textsuperscript{52} The sintered cores were machined several thousandths of an inch oversize and pressed into the heat exchanger body by cooling the sintered piece first in LN\textsubscript{2} to shrink it. The sintered cores expanded upon warming producing a tightly fitting plug.

The silver powder was lightly packed into 5/8 inch i.d. Cu swet couplings to prevent it from falling out and sintered without any applied pressure. The silver powder contracted away from the Cu walls during the sintering process. No attempt was made to find a process which would prevent this since the sintered pieces were to be machined to the proper size. The sintering process used was to insert the Cu fitting containing the powder into a flowing (reducing) gas furnace at a temperature of approximately 495°C for 45 to 50 minutes. The piece was then removed from the hot part of the furnace and allowed to cool for 5 to 10 minutes in the flowing gas. The temperature was constant during the on-off cycles of the furnace to ± 8°C during any sinter. The reducing
FIGURE 20. HEAT EXCHANGER MODULE
atmosphere was a gas of 5% H₂ and 95% N₂. The final sintered pieces used in the heat exchange modules had fractional densities compared to solid silver of 0.321 to 0.352 for the concentrated phase side and 0.355 to 0.370 for the dilute phase side.

In arriving at a procedure which would yield satisfactory sintered pieces, it was necessary to test the flow impedance of a number of preliminary sinters. The flow impedance is a geometrical property defined for incompressible laminar flow as

\[ Z = \frac{\Delta P}{\eta \dot{V}} \]

where \( \Delta P \) is the pressure drop across the piece, \( \eta \) is the viscosity and \( \dot{V} \) is the volume flow rate. The flow impedance can be calculated for several simple geometrical shapes such as cylindrical tubes and wide rectangular channels.\(^96\) For the complicated channels found in a sinter an approximate calculation can be made with estimated particle size, void size, etc.\(^97\) but it is a straightforward procedure to measure the flow impedance. The apparatus used to measure the flow impedance is given in Figure 21. Of particular interest is the flow impedance of the dilute side of the heat exchanger. The total volume flow was timed on the wet test gas meter for several different pressure drops. Since a compressible gas was used, it was necessary to apply a correction to the data.\(^98\) The results plotted in Figure 22 indicate that the total flow impedance of the dilute side of the heat exchanger is 14x10⁸ cm⁻³. The pressure drop of the He³ in the dilute phase was calculated from this to be much less than the change in the osmotic pressure from the mixer to the still so that the diffusion of He³ to the still is not impeded.
FIGURE 21. APPARATUS FOR FLOW IMPEDANCE MEASUREMENT.
FIGURE 22. TOTAL FLOW IMPEDANCE \( z = \Delta P/\eta \dot{V} \) OF THE DILUTE PHASE SIDE OF THE HEAT EXCHANGER

\[ z = 14 \times 10^8 \text{ cm}^{-3} \]
The dilute phase was gravitationally stable (i.e. the density decreased with increasing height).

The heat exchanger system was supported by a nylon tube from which part of the wall was removed leaving three support columns connecting cylindrical bands. Each heat exchanger module was centered inside one of the bands by means of nylon screws passing through the support column and threaded into the heat exchanger module body. The support column was tubular to conserve radial space in the dewar tail. The open structure was designed for access to the heat exchanger modules. This turned out to be unnecessary since any work on the heat exchanger system usually required its removal from the support tube. The support column was bolted to the bottom of the still. The top flange of the mixer was threaded and screwed into the bottom of the support tube. The support column containing the heat exchanger system was flexible and was centered by the standoff on the bottom of the sample cell. The whole assembly appeared to be well centered. Whenever the vacuum can was reassembled and just before contact with the vacuum can flange was made, the refrigerator was tested for an electrical short to the vacuum can. The absence of an electrical short indicated that there were no metal-to-metal contacts which would have provided thermal leaks. The heat exchanger assembly, mixer and sample cell appeared to vibrate freely after the vacuum can was in place so that the conduction thermal leaks through the nylon parts were minimized. Vibration heat leaks could be transmitted to the mixer down the support column.
III.8. Mixer

The mixer was constructed of silver with a brass top flange. The top flange was sealed to the mixer by an indium O-ring as shown in Figure 23. A sintered silver piece was attached to the inside to improve the heat flow to the mixer body. A similar sintered piece was attached to the bottom of the mixer inside the sample cell but had to be removed in order to fix a vacuum leak. The sample filling capillary passed through the mixer. A resistor thermometer and a heater were attached to the outside of the mixer. The attachment of the sample cell is described in the section on the sample cell.
III.9. Sample Cell

The sample cell containing a Mo sample and the CMN thermometer was cooled by the mixer. The sample cell was constructed out of nylon because of its small magnetic susceptibility\textsuperscript{99} and large coefficient of expansion relative to most metals.\textsuperscript{100} The magnetic susceptibility of the sample holder must be small so that its influence on the magnetic susceptibility measurements of the CMN salt pill and the Mo sample are negligible. A demountable superfluid tight sample cell was needed so that studies could be made on both liquid helium and superconductors. In a study of the superconducting transitions involving supercooling, thermal contact using liquid helium is preferable to metallic contacts so that the superconducting transition is not prematurely induced. The sample cell was tightly screwed onto the mixer at room temperature with a very thin coating of Apiezon N grease on the tapered surface of the nylon which contacts the tapered surface of the mixer. The sample cell was vacuum tight at room temperature and became superfluid tight at LHe\textsuperscript{4} temperatures due to differential thermal contraction between the nylon and the mixer.\textsuperscript{101} Nylon ordinarily contracts 3.6 times as much as silver from room temperature to LHe temperatures.\textsuperscript{95,100} Thermal contact between the mixer and the Mo and CMN was made by filling the sample cell with LHe\textsuperscript{4} via the Cu-Ni capillary passing through the mixer. Liquid He\textsuperscript{4} was used as the thermal contact agent in the sample chamber rather than liquid He\textsuperscript{3} primarily because the use of He\textsuperscript{3} would have required gas handling and storage not necessary with He\textsuperscript{4}. There are however a number of advantages in using LHe\textsuperscript{4}.\textsuperscript{102} The heat capacity of LHe\textsuperscript{4} is smaller than that of LHe\textsuperscript{3}. Above about 50 mK, LHe\textsuperscript{4} has a smaller thermal boundary resistance than
He$^3$ and they have comparable thermal boundary resistances down to 10 to 20 mK. The thermal resistance $\Delta T/\dot{Q}$ of a column of LHe$^4$ with the diameter of the sample chamber (1.27 cm) is smaller down to 19 mK than that of LHe$^3$. This results in a smaller thermal time constant for LHe$^4$ in the sample chamber than the time constant LHe$^3$ would have. The thermal time constant in the sample chamber between the Mo and the CMN is estimated (see Ref. 102) by

$$\tau = \frac{2.6 \times 10^{-7}}{T^5} \text{ sec}$$

so that at $T = 30$ mK the thermal time constant in the sample chamber is approximately 10 sec. The primary difficulty in using LHe$^4$ results from the heat leak due to the He$^4$ film which forms above the liquid level. This can be minimized by proper thermal grounding of the filling capillary and by carefully filling the sample chamber so that it is not completely full. The filling capillary was well grounded thermally to the still.
III.10. Mo Sample

The molybdenum sample was spark cut from a high purity, well annealed, zone refined sample used in previous superconductivity measurements in this laboratory (sample 3 in Ref. 1003). The original molybdenum material purchased from Materials Research Corporation typically contains the following impurities as stated by the supplier in ppm: C = 5.0; O = 1.0; N = 0.5; H = 0.3; Fe = 5.0. Single crystals with the (110) plane perpendicular to the axis of the cylinder were obtained by electron-beam zone refining (3 passes). The sample was annealed for 68 hours at 1100°C under a vacuum of about 10⁻⁵ to 10⁻⁶ torr. The average diameter of the sample was 0.290 cm and the length after spark cutting was 5,842 cm. The sample was suspended in the sample cell by two nylon supports which were loose enough to prevent any pressure from being exerted on the ends of the sample.

The residual resistivity ratio of a piece of Mo cut from the annealed sample used in the experiment was measured by a four lead d.c. technique using Mo wire spot welded to the Mo sample. The residual resistivity ratio $R_{298.7}/R_{4.2}$ was measured to be 5600 with an estimated error of ±4%. Because of the preparation of the sample for the measurement, this value should be considered as the lower limit of the residual resistance ratio of the Mo sample used in the critical field measurement. This high value of the resistance ratio indicated a high purity crystal relatively free from strain.
III.11. CMN Thermometer

The CMN salt pill was made by grinding to a fine powder 99.9% CMN supplied by Alfa Inorganics, Inc. in the form of a crystalline powder. The powder was sifted through a U. S. Standard Sieve with mesh number 70 so that the powder used was 210µ or smaller. This powder was then packed with slight pressure into a nylon container so that the CMN salt pill was in the shape of a right circular cylinder with the diameter equal to the height of 1.13 cm and a filling factor of 0.545. Cotton cloth was glued with G. E. 7031 over each end of the nylon cylinder to contain the CMN but allow liquid He\textsuperscript{4} to flow through the thermometer.
III.12. Mutual Inductance Coils

The Mo mutual inductance coils consisted of a primary and a secondary wound from AWG 40 heavy Formvar insulated Cu wire on a common nylon coil form with the CMN mutual inductance coils. The spacing of both sets of coils is given in Figure 23. The Mo primary consisted of 2 layers with a total of 1232 turns. The Mo secondary was wound in 19 layers with sloping edges over approximately 1 inch for a total of 4039 turns. It would be possible to combine the primary and secondary for use as a pickup coil.

The CMN mutual inductance coils were wound from AWG 40 heavy Formvar insulated Cu wire and consisted of a primary and two nearly identical secondaries. The two secondaries were connected in opposition so that the mutual inductance of the empty coil set is nearly zero.\(^{104}\) The CMN measuring and compensating secondaries were wound in 8 layers for a total of 1675 turns each. The turns per layer on layers 5, 6, and 7 differ as does the density of turns on the two coils. These two factors mean that the net mutual inductance of the empty coil set was not exactly zero. The temperature-independent part of the measured mutual inductance at LHe\(^4\) temperatures corresponds to the incomplete compensation of the secondaries and is about 2% of the calculated mutual inductance between one secondary and the primary.

The magnetic susceptibility of the CMN salt pill was measured by measuring the mutual inductance of the coil set described above. The mutual inductance is some residual temperature independent mutual inductance resulting from incomplete compensation of the two secondaries and a temperature dependent mutual inductance resulting from the
susceptibility of the salt contained in the measuring secondary. The mutual inductance was measured by a Hartshorn A.C. bridge operating at 33 Hz as shown in Figure 24. The 33 Hz signal is provided by the output of a PAR Lock-In Amplifier which is amplified and isolated from ground via a transformer. Two variable mutual inductors with ranges of approximately ±1 mH and ± 10 µH are used to null the in-phase inductive unbalanced signal. A decade resistor with 0.01 Ω steps is used to null the out-of-phase resistive unbalanced signal. The mutual inductance coil set containing the Curie salt was designed so that the µH variable inductor can be used during the calibration at higher temperatures and the mH inductor can be used at low temperatures where the change in susceptibility is larger. The unbalanced signal was amplified and detected at the input to the lock-in amplifier. Using the null indicator of the lock-in amplifier, a sensitivity was obtained such that a 0.1 µH change in mutual inductance was detectable.

For experiments in which the sample does not require homogeneous transverse magnetic fields, the mutual inductance coil set can be inverted putting the CMN salt pill at the center of the pole pieces of the 6" electromagnet instead of the Mo sample. The sample cell could then be cooled by adiabatic demagnetization starting from some low temperature achieved by the dilution process.
FIGURE 24. MODIFIED AC HARTSHORN BRIDGE
III.13. Secondary Thermometers, Heaters and Leads

Speer nominal 100 Ω, 1/4 watt resistors were used as secondary thermometers on the mixer and the still. The actual room temperature resistance of the mixer resistor was 98.0 Ω and of the still resistor was 100.7 Ω. Both resistors were glued into thermometer wells on the outside of the mixer and still using G. E. 7031. This provided sufficient thermal contact at the still temperatures but not for mixer temperatures below about 0.2 K. The mixer resistor was thermally grounded to the mixer by a solder contact on one side and by thermal contact through a 510 pf. silver mica capacitor on the other side. Heater coils of AWG 40 Nickel Alloy 90 (Cu-Ni 88-12) having a room temperature resistance of 119.1 Ω were wound bifilarly on the outside of the mixer and the still. The leads from the room temperature feedthroughs to both the resistors and heaters were AWG 40 Nickel Alloy 90. In order to reduce rf Joule heating due to the ambient signals primarily in the FM broadcasting band, high frequency electrical filters were put on the leads going to the mixer at the room temperature feedthrough.\textsuperscript{33} The rf filter network for the mixer thermometer is given in Figure 25. The resistance of the thermometers was measured via one common lead using a Leeds and Northrup Wheatstone bridge. The current applied to the thermometer was always kept to a minimum so that the momentary Joule heating in the thermometers was not observable. The two heaters were supplied current using a different common lead from a constant current source. The still and mixer heaters had a resistance at liquid helium temperatures of 103.7 Ω. The leads to the resistors, heaters and mutual inductance coils were brought down the vacuum can pump-out tube from a room temperature electrical feedthrough
FIGURE 25. MIXER RESISTOR RF FILTER NETWORK
to the low temperature terminal strip. The leads from the heaters and resistors were connected to the same terminal strip. The Cu leads from the mutual inductance coils were stretched along the wall of the vacuum can, taped down with adhesive tape and then connected to a miniature hexagonal connector so that the vacuum can could be easily removed. The coil leads were kept against the outer wall of the vacuum can in the region of the mixer by a spring loaded coil with strip of brass. The leads from the female hexagonal connector were attached to the terminal strip. When the refrigerator was operating with the pumped helium bath at 1.2 K, the lead resistance including filter resistance to the still thermometer was 68.6 Ω and to the mixer thermometer was 95.4 Ω. The leads to the mixer were wrapped around the concentrated phase output of each heat exchanger and were glued in place with G. E. 7031. The initial temperature calibration was taken from work at Illinois where they showed that similar resistors behaved almost identically. The resistance thermometers were later calibrated against the vapor pressure of the pumped LHe⁴ bath and the magnetic temperature of the CMN.
IV. OPERATION OF THE DILUTION REFRIGERATOR

IV: l. Startup

The operation of the dilution refrigerator has not been standardized since the optimum operation has not yet been achieved. The procedure given here is the procedure which has yielded the best results and will provide a basis for further work. The valves are numbered in Figure 26 and are referred to as V1, V2, etc. Assuming that the procedures in Appendix II have been followed, the procedure for running the dilution refrigerator is described below starting with the condensation of the He$^{3}$-He$^{4}$ gas.

1a. The He$^{3}$-He$^{4}$ gas mixture is condensed on the concentrated phase side of the refrigerator only to avoid building up impurities in the still which would increase the film flow rate or interfere with the film flow limiting design. The He$^{3}$-He$^{4}$ gas mixture is condensed on the concentrated side by closing V1-16, 22, 24, 26, 28, 31, 32, and by opening V17-21, 23, 25, 27, 29, 30. V25, 27 should be opened slowly so that gas impurities are condensed out in the LN$_{2}$ trap.

1b. Alternatively, the He$^{3}$-He$^{4}$ gas can be condensed preferentially. The advantage in condensing the He$^{4}$ first is that it will tend to be on the dilute side of the refrigerator, as it should.$^{108}$

2. After most of the gas is condensed, it is necessary to reconcentrate the stored gas so that as much gas is condensed into the refrigerator as possible. This is done by closing V21, 23, 29 and pumping tanks 2 and 3 into tank 4 by opening V22, 28, and using the 1402 KGB pump. When the pressure in tanks 2 and 3 reaches 0.1 torr, close V17, 18,
20, 25 to the tanks. Then close the trap bypass V28 and finally close V22. The 1402 KGB pump can then be cut off. Open V23 and then V29 and continue to condense the gas into the concentrated side.

3a. To start circulation, start 1402 KGB pump and open V7 (after checking vacuum in diffusion pump to be less than $10^{-3}$ torr). Next open V1 slowly. Gas is being pumped through V1, 7 by pump 1402 KGB and exhausted into the concentrated side through V23, 27, 29. Care must be taken not to allow the backing pressure of 1402 KGB pump to exceed 1 atm.

3b. Alternatively, since the initial pumped gas contains a large amount of $\text{He}_4$, it can be stored by closing V23 and opening V19 before opening V1 in 3a. After the still is pumped to a low pressure, close V19 and open V23 to allow circulation as in 3a.

4. The still cools first and finally the mixer cools. When the mixer temperature reaches $\approx 0.56$ K, the rate at which it is cooling decreases. If 0.6-1.0 mWpower is applied to the heaters in the still, the still warms up to $\approx 0.75$ K and the mixer continues to cool.
IV.2. Shutdown for Retransfer of Liquid Helium-4

If the dilution refrigerator has been run and the mixer has cooled, it is possible to retransfer LHe\(^4\) and pump the bath to 1.2 K before the mixer warms up to 0.3 K. To retransfer, the still power is cut off and circulation is stopped by closing V1 only. During retransfer the pressure in the still may increase to 40-50 torr. The pressure on the concentrated side may increase above 1 atm. unless tank 4 is opened by opening V19. If V19 is opened, the gas in tank 4 is usually recondensed during the bath pumpdown. After the transfer is over, the bath is pumped down and the gas in tank 4 is recondensed, then V19 is closed. Then begin circulation again by opening V1, and the mixer will cool again.
IV.3. Shutdown

The shutdown procedure is simple but requires as much as 16 hours to pump all the gas from the refrigerator into the tanks using applied power to the still only. In order to store the gas, open only V1, 7, 17-23, 28, 29 and use pump 1402 KGB. The pressure in the still remains high at ~1 torr during most of the storage operation but drops to 0.1 torr an hour or so before emptying the refrigerator. The pressure may then increase again to ~1 torr. When the refrigerator is empty, the pressure drops rapidly to less than 0.01 torr and the tank gauges stabilize. As a practical guide, tanks 2, 3 and 4 are full when their gauges read 2.5, 8.9, and 2.5 respectively. To preferentially store the He\textsuperscript{3}, first cut the still power off and wait for the pressure to drop. Then, fill tank 3 from the concentrated side (to tank gauge indication of 8.9) and pump remaining gas in the refrigerator into tanks 2 and 4.
V. RESULTS OF THE DILUTION REFRIGERATOR

This section describes the progress made in obtaining a routinely operating dilution refrigerator which will be used to study the properties of superconducting molybdenum. Experiments on superconducting Mo and routine operation of the dilution refrigerator have not yet been achieved. At the present time, the problems of various vacuum leaks discussed in Appendix I seem to have been solved. The present problems are the slow rate of condensation of the gas in the condenser and instabilities in the operation of the refrigerator. The two problems may be related.

The rate of condensation during the early attempts at running the refrigerator was approximately $5 \times 10^{-5}$ moles/sec. This is greater than the circulation possible in the present pumping system. However, the rate of condensation has been getting progressively worse so that it is now about $1.9 \times 10^{-5}$ moles/sec. This is slightly less than the circulation of about $2 \times 10^{-5}$ moles/sec possible in the system. A slow rise in the condensing pressure is observed if there is a large throughput in the 1402 KGB pump. Extensive pumping to clean the refrigerator has not solved the problem. This suggests that solid particles or non-volatile liquids have accumulated in the concentrated side of the refrigerator and are restricting the flow of liquid helium. Three sources could provide these impurities. The LN$_2$ cooled charcoal trap and the sintered silver pieces could provide solid particles. Mechanical pump oil exhausted with the pumped gas could provide oil impurities. It is difficult to see how the oil impurities could pass the LN$_2$ cooled charcoal trap and traverse the relatively long connecting line with its increase in height. The sintered silver pieces appeared to be made well enough to prevent their disintegration into powder. The likely impurity
is the charcoal dust even though the small flow rates and pressures would not easily transport the dust over the vertical height of 1 m it would have to travel to reach the condenser. Impurities were seen on the condenser sintered piece where the input tube is positioned. This problem might be solved by cleaning or perhaps machining the sintered core of the condenser to remove the impurities.

There is a severe instability in the operation of the refrigerator which manifests itself as a sharp change in the condensing pressure and a rather large change in the mixer temperature over a short period of time. Figures 27 and 28 show measurements as a function of time of the condensing pressure, the mixer temperature and the still power applied to the $\text{He}^4$ film limiting evaporator. It can be seen in Figure 27 that cooling from -0.5 K to -0.22 K with zero still power was rapid. Further cooling from 0.22 K was much slower until a still power of 0.158 mW was initiated. The mixer cooled to 56 mK in about two hours. The still power was increased to 0.18 mW in order to increase the cooling rate. The cooling rate appears larger but the temperature rise on change of still power somewhat offset this so that the net cooling in the 20 minutes following the change is small. In an attempt to find a still power which would lead to further cooling, the operation of the refrigerator seems to have been disrupted. After the still power was stabilized at 0.63 mW, the large oscillations in condensing pressure and mixer temperature began. The temperature stability at the lowest temperature of 36 mK was hardly suitable for an experiment. The next run was begun with 0.63 still power. The mixer reached a temperature of 36 mK in a much shorter time than in the previous run, but the condensing pressure
FIGURE 27. EXPERIMENTAL RUN SHOWING THE RELATION BETWEEN THE MIXER TEMPERATURE \(T_m\), THE CONDENSER PRESSURE \(P_c\) AND THE APPLIED STILL POWER \(\dot{Q}_s\).
FIGURE 28. EXPERIMENTAL RUN WITH APPLIED STILL POWER OF 0.63 mW

- Time: 12/18/70
- 4 PM: 5
- 6 PM: 6
- 7 PM: 7
- 8 PM: 8
- 9 PM: 9
- 10 PM: 10
- 11 PM: 11
- 12 AM: 12
- 1 AM: 1
- 2 AM: 2
- 3 AM: 3
- 4 AM: 4
- 5 AM: 5
- 6 AM: 6
- 7 AM: 7
- 8 AM: 8
- 9 AM: 9
- 10 AM: 10
- 11 AM: 11
- 12 PM: 12

- Temperature (K): 0.029, 0.03, 0.1, 0.2, 0.4 P_c (atm)
and mixer temperature oscillations began again. In order to determine if the pressure and temperature oscillations were actually related, the pressure in the condensing line was suddenly decreased by opening one of the evacuated storage tanks. The mixer temperature immediately began decreasing. The still pressure appeared constant during the condenser pressure fluctuations. However, some pressure fluctuation in the still could have been masked by the large volume of the connecting lines which would act as a reservoir of constant pressure.

It appears that the large fluctuations are associated with a large still power (0.63 mW). The refrigerator needs to be run continuously at a lower still power (0.158 mW) to see if the large fluctuations appear at lower still powers. The refrigerator also needs to be operated at higher still powers of at least 1.2 mW if possible. It is possible that the minimum power required to evaporate all the He\textsuperscript{4} film was not reached (see Section II.2). This would allow a large amount of He\textsuperscript{4} to reach the still pumping tube where it would eventually evaporate and reduce the concentration of He\textsuperscript{3} in the circulating gas. The circulated He\textsuperscript{4} can build up on the concentrated side and form a superfluid column. If this column exists in a large temperature gradient in equilibrium with the concentrated phase on both ends, all the conditions on the chemical potential of the superfluid cannot be satisfied. In order to have a stationary column of superfluid, its chemical potential must be constant. In order for the superfluid dilute phase to be in equilibrium with the concentrated phase at each end, the chemical potential of the superfluid dilute phase must be equal to the chemical potential of the concentrated phase. But since the two ends are at different temperatures,
the chemical potentials of the concentrated phases are unequal. Therefore a chemical potential gradient is required in the superfluid column giving rise to a net force on the column in the direction of higher temperatures. Wheatley\textsuperscript{36} has calculated the magnitude of the driving pressure on the superfluid column to be as high as $1/3$ atm. at higher temperatures in the heat exchanger. The column of superfluid is under tension since the driving pressure is different on each end. The superfluid column under tension may break, and the driving pressure which may be sensed as a change in condenser pressure will suddenly disappear. This sudden disappearance of an appreciable condenser pressure was observed in the present system.

The difficulty in eliminating this problem is that if the still power must be increased to reach the critical value, the temperature of the still may increase too much. This is because one 1402 pump may be insufficient to handle the additional load. It was not determined if larger still powers actually disrupted the refrigerator operation even though the still pressure and temperature did increase. The lower and higher still powers were not run because the condenser pressure changes were not understood at the time. If the refrigerator will run normally at lower still powers, then this will indicate that the critical power occurs for lower applied power to the still. If normal operation is found at higher still powers, then this will indicate that the critical power was not used in the present work. If the refrigerator cannot be made to operate in a stable manner, then the most likely solution is to replace the film evaporator. The basic problem may be that the power per unit minimum perimeter of film flow in the present design appears to
require a high critical power for operation of the film evaporator so that normal operation of the refrigerator is not possible.

Because of the instabilities, it has not been possible to determine conclusively if the He\textsuperscript{4} film flow limiting design in the still works. The indication that it may be limiting the film flow is given by a measurement of the concentration in the condenser. The circulation was stopped while the mixer was at a temperature of 0.106 K and the He\textsuperscript{4} bath pressure and the equilibrium condenser pressure were measured by the TI Quartz Bourdon Gauge. The temperature was obtained from the He\textsuperscript{4} bath pressure using 1958 He\textsuperscript{4} Temperature Scale.\textsuperscript{48} Using the temperature and the measured equilibrium vapor pressure, a concentration of 83\% was found.\textsuperscript{109} While this is not high compared to other refrigerators, it is greater than one would expect for the geometry in the present refrigerator.

One modification of the still which might improve the performance of the He\textsuperscript{4} film limiter would be to place a rigid baffle near the outer wall and concentric to it. It would almost touch the stainless steel plate as shown in Figure 29. The purpose of this baffle would be to aid in condensing any evaporated He\textsuperscript{4} that did not recondense between the gap.

In conclusion, temperatures down to 30 mK have been achieved and temperatures less than 55 mK have been maintained for hours. These are temperatures measured outside of the mixer where a sample could be placed. Instabilities have prevented measurements of cooling power or of the properties of superconducting Mo. There are indications that the design of the still incorporating a He\textsuperscript{4} film flow limiting arrangement may at
FIGURE 29. BAFFLE MODIFICATION IN STILL
least be limiting the film flow if not eliminating it. Once the refrigerator runs routinely, it will be a valuable low temperature refrigerator.
APPENDIX I: VACUUM LEAKS

This appendix is included because of the inordinate amount of time spent on solving problems of vacuum leaks in the hope that it may aid others if problems should reoccur or if any rebuilding should be desired. Vacuum leaks have always been a major difficulty in low temperature research.110,111 Vacuum leaks have been a particular problem in dilution refrigerators because of the large number of joints necessary. The first attempts at building a dilution refrigerator by London at Harwell and then by Mendoza and London at Manchester were unsuccessful because of vacuum leaks.112

Two methods of leak detection were employed, each having certain advantages. A CVC mass spectrograph leak detector sensitive to 2.5x10^{-10} atm cc He^4/sec was the most sensitive method of leak detection. The most sensitive way in which it was used was to place the refrigerator in the dewar and alternately fill with He^4 gas and evacuate various regions of the cryostat to determine if any leaks exist. Of course to locate the leak, the refrigerator must be removed from the dewar and the vacuum can removed from the refrigerator. Preliminary leak checks were always made with the refrigerator outside the dewar. He^4 was either sprayed on the part being tested or contained in a plastic bag about the suspected part.

The second method of leak detection was to pressurize the part with gaseous nitrogen (gaseous helium can be used but increases the background in subsequent He^4 mass spectrographic leak detection), immerse the part in water and look for bubbles. Care must be taken not to exceed the bursting pressure for the part being tested. The first heat exchanger
using nylon seals could not maintain a pressure exceeding approximately 15 psi thus limiting the usefulness of this method. The heat exchange systems and capillary tubes including impedances were routinely pressurized to 50 atm. while the still was never pressurized to more than 2 atm. For many leaks this method, while not a sensitive one, can pinpoint the location of a leak much more easily than the He\(^4\) leak detector, particularly when the leak is in an area of several joints or when a piece of tubing is porous.

In addition to leaks in the construction of the refrigerator which will be discussed, the new pumping equipment purchased for the refrigerator was faulty and was repaired at Rice. In the process of adapting the specially built Welch 1402 KGB mechanical pump, a blow hole in the casting at the intake adapter was discovered. The Al sealing gasket was bypassed by this hole. This certain leak was eliminated by filling the hole tightly with indium and using an indium O-ring to seal the intake adapter. This solution has been satisfactory thus far. After the initial operation of the CVC PMCS-4B diffusion pump, it was discovered that the pump itself leaked. The leak was located by pressure testing under water and was found to be in the body of the stainless steel (not at a joint) just above the bottom quick cool coil. This leak was eliminated by silver soldering the quick cool coil in this area again using extra silver solder to shut the hole. This solution has also been satisfactory thus far.

The first refrigerator leaked in an inaccessible place so that is was deemed easier and more reliable to start over rather than dismantle and fix the first model. Upon inspection the leak was attributed to the
destructive action of corrosive residues on stainless steel tubing. These difficulties were not encountered in the second model. Eutec Rod 157 and Eutector Flux 157 were used for all large stainless steel tube connections, and Eutector Flux 157 and soft solder were used for stainless steel capillary connections. All stainless steel joints were washed carefully in a saturated solution of baking soda and rinsed with distilled water.

Although good stainless steel capillary connections could be made with a little care, the relative ease with which joints of cupro nickel capillary were made supports the original suggestion that cupro nickel capillary is far superior.\(^\text{19}\)

The only reliable solution to a leaky solder joint is to remake the joint. In several parts of the refrigerator, pieces were inadvertently made of brass which was slightly porous. Although the most reliable solution to a part made of porous brass is to remake the piece, in one instance it was advantageous to tin the entire porous surface with soft solder rather than disassemble the structure. This procedure provided a leak tight piece.

The first heat exchanger system used nylon end caps which sealed by differential contractions as discussed in the description of the sample cell. These seals often leaked at room temperature, opened up occasionally at low temperatures and did not thermally cycle well in that they tended to loosen on thermal cycling. It is likely the fault of the design since the nylon sample cell has worked well. These seals were finally replaced by soft-soldered end caps in order to obtain a reliable vacuum tight heat exchange system.
The last vacuum leak problem to be solved in the course of obtaining a vacuum tight refrigerator was leaking electrical feedthroughs in the still. While all the leads were brought into the vacuum can through room temperature vacuum feedthroughs, two Latronics 2-lead feedthroughs were used in the still. These feedthroughs were superfluid tight and thermally cycled several times before one of them cracked. This problem was solved by eliminating the feedthroughs and putting the still heater and resistance thermometer on the outside of the still.

The bottom plate of the vacuum can was silver soldered so that it would not become superconducting in the pumped He$^4$ bath and affect the calibration of the CMN thermometer. This joint leaked only after numerous thermal cycles. The leak was fixed by tinning the joint with Cd-Zn (82.5-17.5) solder which is not superconducting at liquid He$^4$ temperatures.
APPENDIX II: GENERAL PROCEDURES FOR STARTING LOW TEMPERATURE RUN

1. Evacuate the He$^4$ dewar

2. The vacuum jacket of the He$^4$ dewar should be pumped out and flushed with air several times to remove any He$^4$ which has diffused into the vacuum jacket since the previous run. Finally, pump a good vacuum and then fill the vacuum jacket with 1 torr of air. Close the stopcock to the vacuum jacket. It is not necessary to pump the air out of the vacuum jacket after cooling to 77 K.

3. Pump a rough vacuum on the refrigerator and sample cell using an external mechanical pump via external pumping line.

4. Close sample cell valves 2 and 9 (P = 10 torr)

5. Continue pumping refrigerator via circulation diffusion pump and backing external mechanical pump for ~24 hours to clean refrigerator (P$_{\text{final}}$ < 10$^{-7}$ torr).


7. Transfer LN$_2$ from LS160 storage dewar to LN$_2$ dewar via insulated connection. Subsequent LN$_2$ fills to top off LN$_2$ bath are made from 5L dewar via funnel at top.

8. The temperature during cooldown is monitored by measuring the resistance of the Mo sample secondary coil using a Triplet Model 800 V-0-M on X100 above 200 Ω and X10 below 200 Ω. The resistance values of interest are
\[ R_{300 \text{ K}} = 1195 \, \Omega \]
\[ R_{273 \text{ K}} = 1090 \, \Omega \]
\[ R_{77 \text{ K}} = 201 \, \Omega \]
\[ R_{20 \text{ K}} = 74 \, \Omega \]
\[ R_{4.2 \text{ K}} = 72 \, \Omega \]

9. When the temperature of the sample cell is below 273 K, evacuate the sample cell using an external diffusion pump. Continue pumping during cooldown to 77 K since the flow impedance of filling capillary is high. Note that the He\textsuperscript{4} dewar is still evacuated so that cooling to 77 K requires about 6 hours. Whenever it is not necessary to pump the sample cell\textsuperscript{out} during cooldown, the time to cool to 77 K can be reduced by using He\textsuperscript{4} exchange gas in the LHe\textsuperscript{4} dewar.

10. When the refrigerator is cooled to 77 K, the LHe\textsuperscript{4} transfer is begun. The He\textsuperscript{4} dewar is pressurized slowly so that the incoming gas does not warm the refrigerator. The transfer tube is inserted in the storage dewar and the LHe\textsuperscript{4} dewar. The pressure in the storage dewar builds up with no applied gas pressure. Then the pressure in the storage dewar drops off to some intrinsic value. At this time there is no liquid being transferred but the refrigerator is being cooled slowly by cold gas from the storage dewar. The purpose of cooling slowly is to avoid freezing out the gaseous hydrogen before the refrigerator is cooled to about 13 K. Approximately 3-1/2 hours are required to cool to near 13 K. The pressure in the storage dewar
is increased slightly and the LHe\textsuperscript{4} dewar is filled with liquid in about 1/2 hour. The 12% of LHe\textsuperscript{4} used in this method is no more than the amount transferred out of the storage dewar when the gas cooling phase is eliminated and liquid evaporation in the experimental dewar cools the refrigerator from 77 K.

11. The dewar is allowed to remain at 4.2 K for 6 to 8 hours to cool the refrigerator to 4.2 K.

12. Even after longer periods at 4.2 K than 8 hours, there is usually sufficient LHe\textsuperscript{4} in the dewar to pump the bath down to 1.2 K. The pump-down procedure is to open the throttle valve to the bath slowly and evenly. The process of opening the throttle valve requires about 1/2 hour (-200-300 torr). The pump requires another hour after the throttle valve is fully open to reduce the bath pressure to about 10 torr. At this pressure the gate valve is opened slowly to its minimum opening and the throttle valve closed. It is usually unnecessary to open the gate valve fully. The time required for the refrigerator (mixer) to cool to 1.2 K depends on how effective the previous cooling times were.

13. The He\textsuperscript{3}-He\textsuperscript{4} gas can be admitted when the He\textsuperscript{4} bath is at 1.2 K but if there is a possibility of a leak anywhere, it is advisable to wait until the refrigerator is cooled to 1.2 K. With this procedure, a leak from the dewar can be distinguished from a leak from the refrigerator into the vacuum can. Subsequent filling of the sample cell will indicate leaks in that system separately.

14. Most of the He\textsuperscript{3}-He\textsuperscript{4} gas can usually be condensed before the top of the vacuum can and condenser begins to warm. It is usually necessary to retransfer LHe\textsuperscript{4} before the refrigerator circulation can
begin. The bath valves to the pump must be closed and the bath repressurized slowly to avoid warming the refrigerator. LHe\textsuperscript{4} re-transfers are not necessary until the bath level falls below the top of the vacuum can. If there is LHe\textsuperscript{3} in the condenser coil, the pressure will increase before the bath level drops below the vacuum can. In this case, the refrigerator must be shut down and the retransfer can be made at that time. Each retransfer when the bath level is just below the vacuum can top requires about 7.5%. 
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