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## A METHOD OF PRODUCING VERY LOW TEMPERATURES BY DISSOLVING He<sup>3</sup> IN He<sup>4</sup>

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A method for the steady production of very low temperatures is described, based on the continuous dilution of liquid He<sup>3</sup> with liquid He<sup>4</sup> and the continuous separation of the isotopes at a higher temperature. At a He<sup>3</sup> circulation rate of about  $1.84 \times 10^{-4}$  mole/sec the temperature 0.1° K can be maintained when heat is added at the rate 1800 erg/sec. In the absence of an external heat input the temperature of the solution is maintained at  $\sim 0.056^\circ$  K. Under constant external conditions the temperature fluctuations do not exceed  $\sim 0.001^\circ$  K.

### 1. INTRODUCTION

THE existing method of producing very low temperatures by the adiabatic demagnetization of paramagnetic salts can be used, in principle, for the continuous production of very low temperatures by repeating periodically the magnetization and demagnetization of the salts. However, the practical realization of cyclical magnetic apparatus (which is, to begin with, very complicated) encounters fundamental difficulties and this technique can hardly be expected to yield refrigerators having a cooling capacity much above 1 erg/sec at  $\sim 0.01^\circ$  K. An example is the refrigerator of Daunt et al.,<sup>[1]</sup> in which adiabatic demagnetization was repeated every two minutes. The lowest temperature attained with this apparatus was  $0.2^\circ$  K, and its refrigerating capacity was 50 erg/sec at  $0.26^\circ$  K.

The basic difficulty lies in the poor heat transfer at the interface between two media, where the thermal resistance increases proportionally to  $1/T^3$ . One can therefore hardly expect to achieve

great success using this technique, and another direction must be investigated.

A new method of producing very low temperatures, which was proposed by London, Clarke, and Mendoza in 1960,<sup>[2]</sup> utilizes the latent heat of dilution of liquid He<sup>3</sup> by He<sup>4</sup>. It has become clear that this technique enables the construction of apparatus possessing several advantages over the existing magnetic refrigerators. These advantages include simplicity, reliability, greater cooling capacity, temperature stability and controllability, and the possibility of cooling any material in any form without requiring the metallic cooling ducts that have interfered seriously with many measurements. This new method should therefore be utilized more extensively than the magnetic method, and will permit many investigations under pure conditions, at least at temperatures of the order of hundredths of a degree, which were previously unattainable for many solid state investigations.

Development of the solution (or dilution) method began in the Laboratory for Nuclear Problems at the end of 1961, when the need arose for a simple refrigerator to be used in nuclear polarization at

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low temperatures etc. Unfortunately the first attempts did not yield positive results for a long time, and the work was suspended for a time. The cause of the failure lay in the fact that since we wished to obviate any external circulation of  $\text{He}^3$  in  $\text{He}^4$  (in order to avoid having extremely large gas flows and the accompanying heat input), we used the incorrect procedure of attempting to separate the isotopes by thermoosmosis at  $T < 1^\circ\text{K}$ . The only positive result of these experiments was the construction of a system capable of absorbing external heat input without a temperature change in the dissolving bath because of continuous circulation and the dissolution of  $\text{He}^3$  at  $\sim 0.3^\circ\text{K}$ . A single mixture of  $\text{He}^3$  and  $\text{He}^4$  at  $\sim 0.3^\circ\text{K}$  demonstrated the cooling effect at this temperature and therefore the possibility of achieving a cooling cycle.

We later utilized the observed temperature stabilization in some experimental work aimed at producing a good thermal contact with the  $\text{He}^3$  bath at a considerable distance from the latter utilizing three capillaries as shown in Fig. 1. The circulation of  $\text{He}^3$ ,  $\text{He}^4$ , and their mixture arises automatically when heat flows to bath A in the directions indicated by arrows, and this heat is transferred efficiently to bath B, which has good thermal contact with the  $\text{He}^3$  bath that maintains bath B at  $0.3^\circ\text{K}$ .

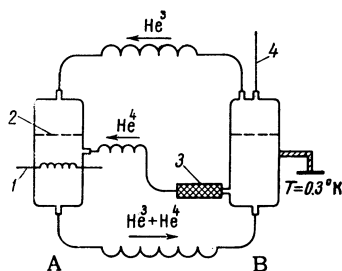


FIG. 1. Sketch of experiment on heat transfer between baths A and B by diluting  $\text{He}^3$  continuously with  $\text{He}^4$ . 1 – heater, 2 – phase separation, 3 – superleak, 4 – capillary for filling the system with the working mixture.

Further work on the dilution method continued after London, Clarke, and Mendoza published a paper at the end of 1962<sup>[3]</sup> in which they supplied answers to some of our important questions. The most promising of the three variants proposed by them was, in our opinion, that suggested by Hall;<sup>[3]</sup> therefore all our subsequent work was confined to this procedure. Here there is no need at all of  $\text{He}^4$  circulation. A “natural” means of isotope separation was proposed (as in other variants), utilizing high enrichment of the gaseous phase with the lighter isotope above a weak solution of  $\text{He}^3$  in  $\text{He}^4$ ; this led to a relatively simple solution of the entire problem.

We shall now discuss briefly the fundamental

physics of the method, describe the most important parts of the experimental apparatus, and present results obtained in trials of this apparatus.

## 2. FUNDAMENTAL PHYSICS OF THE METHOD

The cooling effect produced by dissolving liquid  $\text{He}^3$  in  $\text{He}^4$  results from the large difference between the molar heat capacities of  $\text{He}^3$  and its weak solutions in  $\text{He}^4$ .<sup>[4]</sup> Since no chemical reaction actually occurs during the dilution process enthalpy should be conserved. Therefore, if the process is adiabatic the solution will be cooled if its heat capacity exceeds the mean heat capacity of the original constituents.

The specific heat of  $\text{He}^3$  differs from that of its weak solutions at very low temperatures because of the Pauli principle, which leads to spin ordering in concentrated  $\text{He}^3$  and to degeneracy of the system at  $T \leq 0.45^\circ\text{K}$ . This actually means that a quantum collective interaction occurs between fermions under certain conditions; one consequence of this interaction is the applicability of the Pauli principle itself. This interaction is not present in a weak solution at the same given temperature and the spin system is not degenerate. Therefore, as was first suggested by Pomeranchuk,<sup>[5]</sup> a system of  $\text{He}^3$  atoms in the solution should behave like an ideal gas in a  $\text{He}^4$  “vacuum,” with its “pressure” represented by the osmotic pressure of  $\text{He}^3$  in the solution, where the van’t Hoff equation is obeyed; the molar heat capacity is restored to the classical value ( $3/2$ )R. Indeed, since  $\text{He}^4$  is a superfluid the  $\text{He}^3$  atoms experience almost no collisions with the  $\text{He}^4$  atoms, so that their mean free path depends mainly on the concentrations alone. On the other hand, because of zero spin the  $\text{He}^4$  contribution to the heat capacity of the solution at  $T < 0.5^\circ\text{K}$  becomes negligibly small. Therefore with respect to  $\text{He}^3$  we have  $\text{He}^4$  playing the part of a “vacuum” in which  $\text{He}^3$  atoms behave like free particles that do not interact with each other.

The described solution is the only model, as well as a very good one, of an ideal gas at any low temperature above the phase separation point. With decreasing temperature the equilibrium concentration of the solution should evidently obey the  $T^{3/2}$  law because the spin system gradually becomes degenerate. This is associated with a reduction of the phase volume and the corresponding increases in the occupation numbers of quantum states. To attain constancy of the degeneracy conditions the phase volume per particle,  $p^3V/N$ , must remain constant. Therefore

$$n_0 \sim p^3 \sim T^{3/2}, \quad (1)$$

where  $n_0$  is the particle density per unit volume and  $p$  is the maximum momentum of the particles. At  $T = 0$  obviously any system will be degenerate; therefore no system with the foregoing properties will exist at  $T = 0$ .

The cooling mechanism associated with the dilution of  $\text{He}^3$  by  $\text{He}^4$  is therefore necessarily of quantum spin character and is formally analogous to adiabatic demagnetization, where cooling also results from reordering of the spin system through interaction with an external field. In the dilution process the role of the external field is played by internal interaction within the system; this disappears as the separation of the particles increases through dilution. Therefore, in contrast with adiabatic demagnetization, where enthalpy is not conserved because the potential energy of the particles varies with the field, in the case of dilution the enthalpy conservation is based on energy conservation so long as the interatomic potential energy of the particles remains constant. The difference between the heats of vaporization of  $\text{He}^3$  from concentrated and weak solutions in  $\text{He}^4$  is a measure of the correctness of this hypothesis, which, as will be shown, seems thus far to be consistent with experiment.

In contrast with adiabatic demagnetization, where the minimum attainable temperature is bounded by the Curie law of temperature dependence for the interaction energy between paramagnetic ions, there is no analogous limitation in the dilution method because the energy of the corresponding dipole-dipole interaction between  $\text{He}^3$  atoms in the solution is negligibly small compared with the energy of thermal excitations for all equilibrium concentrations and temperatures.

If superfluidity of  $\text{He}^3$  exists it should be manifested during the dilution process. Cooling below the transition temperature, for example, could be absent if for some reason the solution should cease to be of ideal character but should consist, instead, of coupled  $\text{He}^3$  atoms, although this is extremely unlikely.

### 3. METHOD OF CONTINUOUS DILUTION

In principle, continuous dilution can be achieved in several ways; we shall consider the simplest technique, which is represented by Fig. 2. The baths A and B, connected by the tube 1 of a given cross section, are filled with a mixture of  $\text{He}^3$  and  $\text{He}^4$  up to level 5 at the initial temperature  $1^\circ\text{K}$ . When vapor is pumped from bath A and returned as a liquid to bath B through the coil 3 (at the bot-

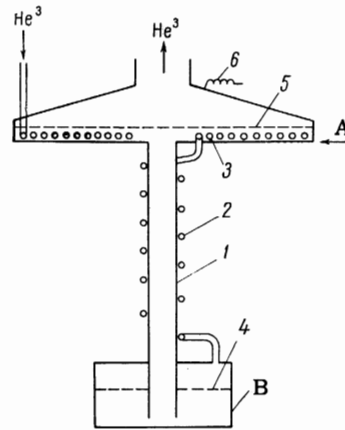


FIG. 2. Schematic diagram of cooling cycle.

tom of bath A) and the heat exchanger 2, circulation of  $\text{He}^3$  results mainly from its high enrichment with the gas phase above the solution in bath A. The temperature of baths A and B begins to drop as in an ordinary refrigerator operating on the basis of  $\text{He}^3$  evaporation. When the phase separation temperature is reached the  $\text{He}^3$ , which was originally dissolved uniformly in the volumes of A and B, begins to accumulate at the top of B, filling the coil 3 and the heat exchanger 2. When steady-state equilibrium is reached the circulation of  $\text{He}^3$  slows down to a level depending on the external heat influx. The  $\text{He}^3$  concentration in A is maintained constant by  $\text{He}^3$  diffusion from B to A through the connecting tube 1, and the loss of  $\text{He}^3$  from the lower heavy phase in B is continuously restored by the dissolution of the upper phase at the phase separation 4. Through the heat absorption that accompanies dilution the temperature of B drops a little lower than that of the evaporating bath A. The osmotic pressures in baths A and B strive toward equality, and by the van't Hoff law the concentrations of the solutions become inversely proportional to their temperatures.

When the heater 6 of the evaporating bath A is switched on the solution here gradually becomes impoverished. This leads to increased decline of osmotic pressures and to intense  $\text{He}^3$  diffusion through the connecting tube from the lower phase in bath B. The results are a lower  $\text{He}^3$  concentration in the solution and intense dissolving of the upper phase. The temperature of B then begins to drop rapidly and the entire system enters a new steady state, where the temperature drop depends only on the level of  $\text{He}^3$  circulation, the power supplied to bath A, and the quantity of heat entering bath B. The last of these depends mainly on the quality of the reflux exchanger 2 and the size of tube 1 where heat also flows to bath B through a helium column. It must be noted, however, that

the temperature of A inevitably rises both because of diminishing  $\text{He}^3$  concentration in the solution and increasing pressure in the pumping system because of its limited capacity. Therefore, as the concentration decreases the isotope separation coefficient is reduced somewhat and the heat influx through the tube 1 to B grows. At a very low concentration the system begins to operate with lower efficiency, thus limiting the lowest permissible concentrations of the solutions and, accordingly, the temperature of the dissolving bath that is attainable by this technique for a given pumping rate. Thus, despite the presence of the heat exchanger 2, which could in principle reduce extremely the heat influx accompanying  $\text{He}^3$  to the dissolving bath, the system as a whole cannot reduce temperatures without limit.

In addition to the foregoing limitation that is associated with the temperature rise of the evaporating bath, there is another more fundamental limitation. The latter results from fountain pressure in the superfluid helium column when a temperature differential exists therein. This pressure impedes  $\text{He}^3$  diffusion and opposes the drop of osmotic pressures in the connecting tube. The concentration, and therefore the temperature of the bath, can clearly not drop below the values at which the osmotic pressure becomes comparable with the fountain pressure and diffusion ceases.

We shall now calculate this limit, taking as the temperature of the upper bath  $0.6^\circ\text{K}$ , which could hardly be lower for concentrations under  $0.1\%$  because of the decreased vapor pressure according to Rayleigh's law. The fountain pressure can be obtained by integrating the entropy of liquid helium at the pressure of saturated vapor:

$$\Delta P_{\text{fount}} = \int_{T_B}^{T_A} \rho S dT, \quad (2)$$

where  $\rho$  is the density of  $\text{He}^4$  and  $S$  is the entropy per gram. To calculate the entropy we can use the value of the specific heat of  $\text{He}^4$  given in [6], represented below  $0.7^\circ\text{K}$  by the formula  $C = 0.0235 T^3 \text{ J/g-deg}$ . Substituting

$$S = \int \frac{C}{T} dT = 0.0075 T^3 \text{ J/g},$$

into (2), we obtain at  $0.6^\circ\text{K}$

$$P_{\text{fount}} = 1/4 \rho \cdot 0.075 T^4 = 354 \text{ dyne/cm}^2. \quad (3)$$

The osmotic pressure in a weak solution can be derived by extrapolating the measurements at  $0.8^\circ\text{K}$ . [2] However, since we must here use the laws of ideal gases, which apply better to small

concentrations, the osmotic pressure can be derived more accurately simply from the van't Hoff equation:

$$P_{\text{osm}} = \frac{x}{v} RT, \quad (4)$$

where  $x$  is the number of moles of  $\text{He}^3$  in the volume  $v$  (the molar volume of  $\text{He}^4$ ). Taking  $x = \alpha T^{3/2}$ , where  $\alpha = 0.615$ , we obtain

$$P_{\text{osm}} = \alpha T^{5/2} R / v. \quad (5)$$

By equating (3) and (5) we obtain

$$T_{B(\text{min})} = (P_{\text{fount}} v / R \alpha)^{2/5} = 0.033^\circ\text{K}. \quad (6)$$

This limit will be somewhat higher in actuality, since we have neglected the osmotic pressure in the evaporator, assuming in infinitesimal concentration.

#### 4. REFRIGERATING CAPACITY AND STABILITY OF THE COOLING CYCLE

The enthalpy diagram in Fig. 3 is a sufficient basis for determining the refrigerating capacity of the cooling cycle. Here the higher curve represents the enthalpy  $H$  of the solution, derived from the ideal-gas model. The system under consideration operates at a constant pressure equal to the osmotic pressure, while the liquid  $\text{He}^3$  is vaporized in a volume that is practically unlimited because this "gas" is pumped continuously from the  $\text{He}^4$  reservoir; therefore when calculating the enthalpy of the solution we must take into account the work of expansion and therefore start from the specific heat at constant pressure. We then have

$$H = \int C_P dT = 5/2 RT. \quad (7)$$

The enthalpy of pure  $\text{He}^3$  calculated from specific heat measurements of  $\text{He}^3$  down to  $0.1^\circ\text{K}$ , [7] is represented by the lower curve. The dissolving process is represented by the horizontal straight line AB if the temperature to which  $\text{He}^3$  can be

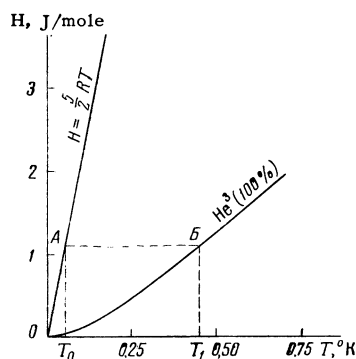


FIG. 3. Enthalpy diagram for calculating the refrigerating capacity of the cycle.

cooled before reaching the dissolving bath is  $T_1$ ; the temperature of the resultant solution is then  $T_0$ . The heat that can be absorbed by the solution, thus heating it to some temperature  $T$ , is obviously

$$Q(T) = (H - H_0)z = \frac{5}{2}R(T - T_0)z, \quad (8)$$

where  $z$  is the rate of circulation of  $\text{He}^3$  in the system.

The refrigerating capacity of the cycle is thus found to be a linear function of temperature, depending only on the  $\text{He}^3$  circulation rate, so long as the concentration of the solution permits us to use the ideal-gas model.

Equation (8) is valid while the solution temperature  $T$  is much lower than  $T_1$ . The change of  $T$  resulting from external heat input then has very little influence on  $T_1$ , or correspondingly on  $T_0$ , and can be neglected. This case obviously pertains to a version of the cooling cycle having a low-efficiency heat exchanger. Since the enthalpy of the solution at a given temperature is many times greater than the enthalpy of concentrated  $\text{He}^3$ , it is possible in principle to cool the  $\text{He}^3$  flow to the minimum temperature of the solution. With improvement of the heat exchanger  $T_0$  will approach  $T_{\min}$ , and in the case of an "ideal" heat exchanger when  $T_{\min}$  is reached the dissolving process under adiabatic conditions comes to an end. However, dissolution can still continue under isothermal conditions at  $T_{\min}$ , with external heat input

$$\dot{Q}_{T_{\min}} = H_{T_{\min}} - \int_0^{T_{\min}} G_{\text{He}^3} dT = 0.65 \text{ J/mole}. \quad (9)$$

In this limiting case the refrigerating capacity at any  $T$  above  $T_{\min}$  is

$$\dot{Q}(T) \cong \frac{5}{2}RTz - z \int_0^T C_{\text{He}^3} dT. \quad (10)$$

Improvement of the heat exchanger can thus greatly affect the operation of the cycle even though it cannot greatly lower the minimum attainable temperature of the solution. As a result, the lowest temperature of the solution is maintained even with some heat input, which in any experiment can be the effective amount.

Finally, we shall discuss the stability of the cycle and the possible causes of thermal fluctuations. The given cycle is a system with positive feedback. The lower the temperature of the dissolving bath, the smaller the amount of heat entering it because of additional cooling of  $\text{He}^3$  in the reflux heat exchanger. In the presence of positive

feedback, fluctuations of a system are theoretically possible depending on the degree of the feedback, which in the present case is determined by the quality of the heat exchanger. If the exchanger functions poorly the feedback will be small and the system will be stable. With an "ideal" exchanger, in which practically all heat is transferred, the bath temperature will drop continuously until the solution is "supercooled." Further diffusion and dissolution will then cease if the osmotic pressure becomes equal to the sum  $P_{\text{fount}} + P_{\text{osm}}$  in the evaporating bath. This leads immediately to severe deterioration of the thermal regime and to enhanced heat influx into the dissolving bath, both because of the increasing temperature of the  $\text{He}^3$  flowing continuously into the bath and because of the enhanced heat flow through the liquid helium column in the connecting tube when diffusion is interrupted.

All processes occurring in the system will engage in mutual regulation, generally speaking; this leads to a "self-consistent" state, although thermal fluctuations of a regular nature are possible. However, when an external heat load exists the conditions for the excitation of fluctuations will be absent and the cycle should be stable. It is also possible to find temperature oscillations of the dissolving bath having a different character, a possible cause of which would be an incorrect amount of  $\text{He}^3$  in the system. With insufficient  $\text{He}^3$  content dissolution can begin partially in the exchanger and an optimal operating regime will not be achieved. With excess  $\text{He}^3$  content dissolution can occur periodically in the connecting tube and also in the evaporator; the operation of the system will then exhibit sharp breaks, accompanied by strong heating of the dissolving bath followed by cooling because of a temporarily reduced amount of  $\text{He}^3$  in the dissolving bath. The operation of the cycle will then have the character of relaxational oscillations.

The amount of  $\text{He}^4$  in the system has only a very slight influence on the position of the phase separation, because the concentrations are small and should therefore have practically no effect on the cycle.

We note that no mechanical devices are needed to obtain optimal and stable operating cycles if the coil in which  $\text{He}^3$  condenses is almost completely filled with  $\text{He}^3$ . Since in this case the amount of  $\text{He}^3$  in the condenser is constant, a change in the amount of  $\text{He}^3$  contained within the external system will be reflected immediately by the amount of  $\text{He}^3$  in the dissolving bath, thus enabling simple regulation of the operation of the cycle.

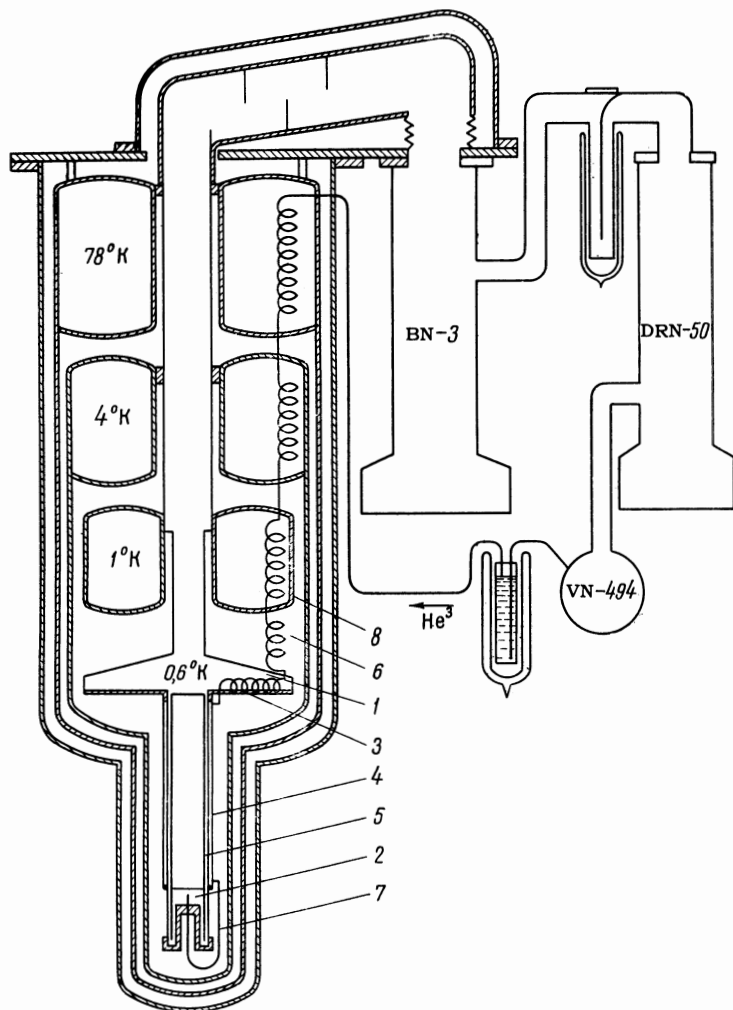


FIG. 4. Diagram of apparatus.

## 5. DESCRIPTION OF APPARATUS AND DISCUSSION OF EXPERIMENTAL RESULTS

The experimental apparatus is represented schematically in Fig. 4. The helium cryostat and the  $\text{He}^3$  pumping system are practically identical with those customarily employed in cooling devices operating by means of the evaporation of  $\text{He}^3$ . The  $\text{He}^3$  vapor was pumped by means of BN-3 and DRN-50 booster pumps and a VN-494 mechanical pump with the respective speeds 450, 30, and 0.2 liters/sec. (Sealing of the mechanical pump required replacement of the factory housing and supplementary packing of the pump shaft.) The  $\text{He}^3$  pumping line was made of stainless steel 0.5 mm thick. The diameter of the tube was 20 mm in its lower part, 34 mm as far as the nitrogen bath, and 100 mm thereafter to the pump. The length of the line from the evaporating bath to contact with the nitrogen bath was  $\sim 60$  cm.

Figure 4 indicates clearly how the  $\text{He}^3$  vapor was fed to the BN-3 pump at nitrogen temperature. The reflux  $\text{He}^3$  stream passing through the carbon

trap at nitrogen temperature was first cooled in the nitrogen and helium baths of the cryostat and was condensed in the coil 8, consisting of a 136-cm long copper tube of 0.65-mm inside diameter lying on the bottom of the lower helium bath. The temperature of the latter was maintained at about  $1^\circ\text{K}$  by pumping out the helium vapor with the 40-liter/40-liter/sec VN-4 pump.

The design of the dilution stage is very simple. The evaporating bath 1 (Fig. 4) has a thick flat bottom, of 15.5-cm diameter, with helical grooves containing the coil 3 that consists of four copper capillaries each 3 m long and  $\sim 0.3$  mm in diameter. The capillaries are connected in parallel to increase the throughput of the coil. Liquid  $\text{He}^3$  enters the coil through stainless-steel capillary 6, which is 18 cm long and 0.1 mm in diameter and acts as the throttle valve required to make  $\text{He}^3$  condensation occur in the helium bath at the working rate of circulation without boiling of the liquid at the coil exit. This obviously requires that the pressure at the exit of coil 8 should not drop below

8 mm Hg, because the capillary filled with the liquid is in contact with the helium bath at 1° K. (This circumstance is sometimes ignored in designing cryostats operating by means of He<sup>3</sup> evaporation; the result is then an appreciable heat input during continuous operation.)

The heat exchanger and connecting tube were made of three stainless steel tubes, ~22 cm long with ~0.3 mm walls and about 20 mm diameter, inserted into one another as shown in Fig. 4. Through the annular 0.15-mm wide space 5 between the sealed inside tube and the middle tube He<sup>3</sup> diffuses into the evaporating bath; the outer space 4, which is about 0.1 mm wide, acts as a heat exchanger. Cold He<sup>3</sup> from the heat exchanger flows through capillary 7 into the dissolving chamber 2, of 19-mm diameter, which is fastened to the lower end of the inside tube.

About 2 to 3 liters of He<sup>3</sup> and 8–12 liters of He<sup>4</sup> were usually needed under normal conditions to fill the entire system to its operating level. The operation of the cycle depends to a large extent on the amount of He<sup>3</sup> in the system. Therefore the optimal concentration of the mixture was determined experimentally for each experiment. During most of the experiments that were performed to determine the optimal parameters the operation of the circuit was monitored with carbon resistance thermometers of different sensitivities which were fastened to the outside walls of the apparatus.

The temperature of the evaporating bath varied within wide limits depending on the input power and the concentration of the contained solution. In most experiments its working temperature was maintained at ~0.6° K with He<sup>3</sup> circulation at about  $1.84 \times 10^{-4}$  mole/sec when the power supplied to the separating bath was  $4.7 \times 10^{-3}$  W. As we shall show, the minimum temperature then attainable in the dissolving bath was ~0.056° K, which is very close to the theoretical limit for the given technique. If our assumed formula  $x = 0.615 T^{3/2}$  is correct, the corresponding concentration of the solution is ~0.8%; therefore the concentration of the solution in the evaporating bath, with the fountain pressure taken into account, could not have been above 0.056%.

It was of interest to evaluate the heat of vaporization of He<sup>3</sup> from this weak solution; it was possible to arrive at a fairly determinate result. The external heat input into the evaporating bath was  $\sim 4.7 \times 10^{-3}$  W. The additional heat influx resulting from He<sup>3</sup> circulation was of the order 0.4 cal/mole. Therefore the heat of vaporization of He<sup>3</sup> from a solution with a concentration of the order 0.06% is not less than 6.5 cal/mole. This agrees approxi-

mately with the value given in [8] for the heat of vaporization of pure He<sup>3</sup>, calculated from the Clapeyron-Clausius equation using experimental densities of the liquid and vapor and the temperature dependence of the saturated vapor pressure. (Our value for the heat of vaporization of He<sup>3</sup> from a weak solution must be amended by a small positive correction for the He<sup>4</sup> admixture; we have not yet determined this correction.)

The pressure in the upper part of the pumping line (the region of contact with the nitrogen bath) was ~0.01 mm Hg under the working conditions. Therefore the vapor pressure above the solution with the given low concentration at ~0.6° was not lower. This was a favorable circumstance enabling operation at very low concentrations in the evaporating bath.

The operation of the circuit is greatly dependent on the width of the space through which He<sup>3</sup> diffuses. The size should be consistent with the pumping speed and the desired refrigerating capacity. If the space is very wide a large amount of heat flows from the upper "heat" bath into the dissolving bath. If the space is very narrow diffusion is impeded; the osmotic pressure drop at the ends of the connecting tube is then enhanced, with the result that the minimum attainable temperature of the solution is higher.

The physical process occurring in the space can be pictured as follows using the two-component He<sup>4</sup> model. The superfluid He<sup>4</sup> component moves to the warm end of the tube without entraining He<sup>3</sup> atoms, while a stream of normal He<sup>4</sup> having an equivalent mass moves in the opposite direction and interacts with the opposing stream of cold He<sup>3</sup> "gas." A considerable fraction of the heat transported by the normal He<sup>4</sup> component is removed by the opposing He<sup>3</sup> stream before this heat can reach the dissolving bath. The helium temperature distribution along the tube is altered thereby, so that the principal temperature drop occurs at the upper end, where there also occurs most of the heating of the diffusing cold He<sup>3</sup> stream to the temperature of the upper bath.

It has thus become clear that a simple calculation of heat flow through the helium column without detailed account of the complex heat-exchange process will not represent reality, since such a calculation could not be formulated in the terminology of thermal conduction for even a column of pure He<sup>4</sup>. Moreover, we have none of the necessary experimental information on the thermal conductivity of weak solutions in the required temperature and concentration ranges. Using data in [9], we obtain a rough upper limit of the order

700 erg/sec for the maximum heat flow.

With a large external heat influx to the dissolving bath it was noted that less heating of the solution occurred in the case of a wider space and faster circulation; this is entirely natural. Therefore, depending on the specific purpose of the apparatus, the space can be varied within certain limits to determine its optimal size experimentally. In our present version of a heat exchanger this space is easily rendered adjustable during the experimental work by using tubes of slightly conical shape.

For the purpose of determining the absolute temperature of the solution in the dissolving bath and to clarify the possibility of utilizing the refrigerating circuit in physical experimentation, we experimented with the cooling of cerium-magnesium nitrate crystals by the circulating flow. The scheme of this experiment is represented in Fig. 5.

Thin crystal plates 1 were placed at the bottom

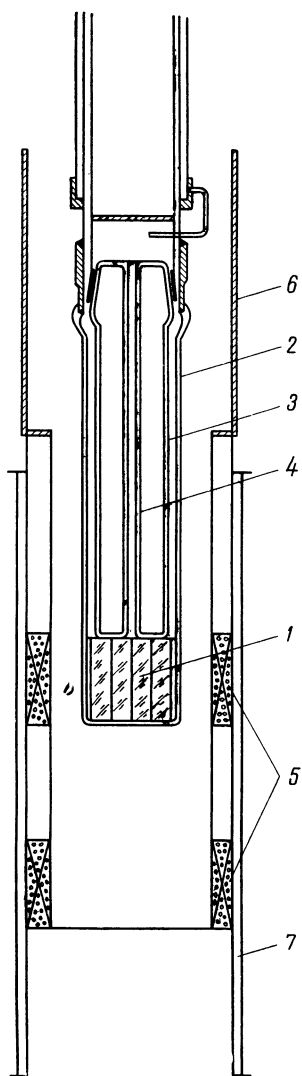


FIG. 5. Diagram of experiment to determine the absolute temperature of the solution.

of the glass ampoule 2, which was connected by a Kovar seal to the inside tube of the heat exchanger. A sealed glass tube 3 containing a sealed-in capillary 4 was fastened inside the ampoule and connected by a ground glass joint to the dissolving bath. In this way the crystals were isolated from the metal parts of the circuit that would interfere with magnetic susceptibility measurements, and a model was set up for the conditions of cooling by a solution at a considerable distance from the dissolving bath. It is easily seen that the  $\text{He}^3$  stream could enter the evaporating bath only by diffusing through capillary 4 to the crystals, and also in the reverse direction through the space between the glass tubes. Since this was not accompanied by heat input, the solution temperature in the dissolving bath was identical with that at the bottom of the glass ampoule and could be determined from the magnetic susceptibility of the crystals. For the purpose of measuring the latter quantity, on a helium shield 6 we positioned a measuring coil 5 consisting of two 2000-turn sections and a coil 7 that set up an alternating magnetic field. Measurements were obtained using an R-56 ac bridge at 50 cycles. The susceptibility at  $1^\circ\text{K}$  was used as a calibrating point. This temperature was established very quickly after condensing the working mixture in the evaporating bath by means of thermal contact with the helium bath through the gas column in the  $\text{He}^3$  pumping line at a pressure of  $\sim 8$  mm Hg. The temperature was checked independently with carbon resistance thermometers calibrated in liquid helium. Although there was a small registered effect due to change in the crystal temperature from approximately nitrogen temperature to  $1^\circ\text{K}$ , the reproducibility of numerous measurements was quite good.

When the heater in the upper bath was switched on and a circulation of  $\sim 1.8 \times 10^{-4}$  mole/sec was established the observed effect at  $1^\circ\text{K}$  with an optimal mixture filling the system and a 0.15-mm space increased by a factor of about 17.5. The evaporating bath temperature was maintained at about  $0.6^\circ\text{K}$ . The temperature of the crystals, after a small correction to the Curie law and a correction for the demagnetizing factor, was reduced to  $0.056^\circ\text{K}$  with an error that could not have exceeded 10%. Temperature fluctuations during steady operation did not exceed  $0.001^\circ\text{K}$ . To determine the cooling rate of the circuit a measured amount of heat was supplied directly to the dissolving bath and the temperature change of the solution was measured by registering the change of crystal magnetic susceptibility.

The results obtained with two different circula-



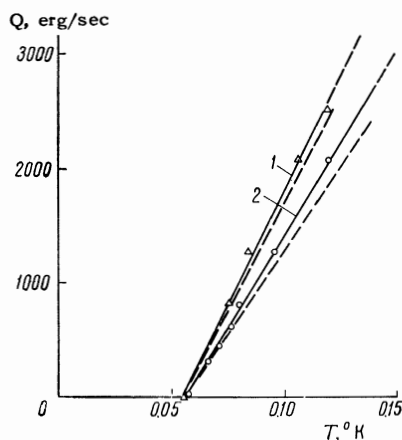


FIG. 6. Cooling rate of the apparatus with 0.15 mm width of space. Curve 1 —  $z = 1.84 \times 10^{-4}$  mole/sec; curve 2 —  $z = 1.43 \times 10^{-4}$  mole/sec.

tion rates are shown in Fig. 6. The cooling rate is actually found to be a linear function of temperature and agrees well with the dashed theoretical lines. The deviations could have resulted both from a  $\text{He}^4$  admixture in the circulating flow and from deviations between the properties of the real solution and the assumed ideal-gas model. The smallness of the deviation attests to the suitability of our concepts in the given temperature region. The heat influx in the dissolving bath is still quite large; this apparently accounts for the considerable difference between the achieved temperature and the possible minimum. This heat, probably amounting to  $\sim 2000$  erg/sec, was carried into the dissolving bath mainly by the  $\text{He}^3$  stream, which should thus have had a temperature  $\sim 0.5^\circ \text{K}$ . We thus have evidence of low heat-exchanger efficiency and, possibly, of a high temperature of the liquid  $\text{He}^3$  flowing from the helium bath, where the coil surface was not large enough for the utilized  $\text{He}^3$  circulation rate.

In conclusion the authors wish to express their deep gratitude to Professor V. P. Dzhelepov for making this work possible, and to L. B. Parfenov for participating in the first attempts to achieve the dilution process. They also thank N. I. Kvitkova and F. A. Tagirova for experimental assistance, as well as A. S. Akimov, A. O. Orlov, and A. N. Nikolaev for excellently prepared units of the

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Note added in proof (May 11, 1966). After Completing the present work we learned of new experimental data on the solubility of  $\text{He}^3$  at  $\sim 0.01 \text{ K}$ .<sup>[10]</sup> It appears that even at  $T = 0$  a finite solubility of  $\sim 5\%$  exists.<sup>[11]</sup> Accordingly it is probable that the described method is not limited theoretically and can be utilized to obtain considerably lower temperatures than those achieved in the present work. We now present results that we obtained with a new version of the apparatus in which the heat-exchanger surface has been increased five times and the  $\text{He}^3$  circulation rate is  $\sim 1.14$  mole/sec:

$T, ^\circ\text{K}$	0.025	0.03	0.04	0.05	0.06	0.07	0.08	0.09	0.1
$\dot{Q}, \text{erg/sec}$	0	50	180	320	480	670	860	1080	1300

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