PHYSICAL REVIEW VOLUME 91, NUMBER 5 SEPTEMBER 1, 1953

The Rapid Separation of He^3 from He^4 by the "Heat Flush" Method[*]

T. Soller, W. M. Fairbank,[†] and A. D. Crowell
Amherst College, Amherst, Massachusetts

(Received May 25, 1953)

A practical method is decribed for separating the He^3 which occurs in well helium from the liquefied mixture using the "heat flush" technique. Approximately 2 liters/hr of liquid can be processed, a rate considerably in excess of any previous separations. When the separated isotope is finally withdrawn at room temperature, the concentration of He^3 is approximately 0.5 percent. This concentration corresponds to an enrichment factor of 3×10^4, which is an order of magnitude greater than enrichment factors reported previously in the literature.

I. INTRODUCTION

THE peculiar superfluid properties of liquid helium below the λ point have been the object of intense experimental and theoretical investigation since their discovery in 1937. In 1947 it was discovered that the rare isotope He^3 apparently does not exhibit these superfluid properties.[1] This seemed to lend evidence to the idea proposed by London in 1938[2] that the transformation of He I into the superfluid He II is analogous to the degeneration which occurs at low temperatures in a perfect Bose-Einstein gas. He^3, obeying the Fermi-Dirac statistics, would not undergo such a transition.

Experiments on the properties of He^3 and solutions of He^3 in He^4 have been made more difficult because of the small quantities of He^3 which have been generally available. Since He^3 occurs in ordinary well helium gas only to the extent of 1.5 parts in 10^7,[3,4] separation of He^3 by conventional methods is prohibitively slow.[5–7] However, the fact that He^3 does not participate in the superfluid flow of He^4 makes possible a very rapid separation of the two isotopes in the liquid phase.[8,9] Lane, Fairbank, *et al.*[8,9] have demonstrated that if heat is applied at one end of a vessel containing liquid helium below the λ point and refrigeration at the other end, the He^3 flows with the normal liquid away from the heater and toward the cold end of the tube. They called this method of separation a "heat flush method." Other investigators[10–12] have thus enriched the concentration of He^3 in relatively small samples of liquid by factors up to 5000.

This paper reports essentially complete removal of the He^3 at a higher enrichment factor than has been previously obtained in a single step, as well as the development of a method for performing this separation inside the helium cryostat at about the rate of liquefaction.

II. APPARATUS AND EXPERIMENTAL PROCEDURE

The apparatus shown in Fig. 1 was designed to be suspended below the main liquid reservoir in a Collins type liquefier built at Amherst College. A and B are copper spheres, of volume 700 cc and 1000 cc, respectively. Both bulbs can be filled with liquid helium through valves V_1 and V_2. The temperature of the liquid in bulb B can be reduced by pumping through tube T_4, and the liquid in A is simultaneously cooled because of the good thermal contact with B afforded by the thin copper cup C separating the two liquids. A small tube T_1 leads from the bottom of C to an external Toeppler pump.

The procedure in performing a separation is as follows:

(1) The bulbs are filled with liquid, using the four carbon resistors R_1 through R_4 to indicate when the bulbs are empty and when they are full.[13]

(2) Valves V_1 and V_2 are closed.

(3) The temperature of the liquid in both spheres is reduced to below the λ point by pumping through T_4.

(4) Heat is applied to the liquid in A by means of heater H_1 at the top of A, while the liquid in B is maintained below the λ point by continued pumping on B. This produces a "heat flush," causing the He^3 to collect at the bottom of C in the small tube D which is the coldest part of the vessel A. Upon application of a quantity of heat equal to or greater than that required to raise the temperature of the liquid in A from absolute zero to the temperature actually existing in A, all of

---

[*] This work has been assisted by the Research Corporation and the U. S. Office of Naval Research.

[†] Now at Duke University, Durham, North Carolina.

[1] Daunt, Probst, Johnston, Aldrich, and Nier, Phys. Rev. **72**, 502 (1947).

[2] F. London, Phys. Rev. **54**, 947 (1938); J. Phys. Chem. **43**, 49 (1939).

[3] L. T. Aldrich and A. C. Nier, Phys. Rev. **74**, 1590 (1948).

[4] J. H. Coon, Phys. Rev. **75**, 1355 (1949).

[5] McInteer, Aldrich, and Nier, Phys. Rev. **74**, 946 (1948).

[6] A. Andrew and W. R. Smythe, Phys. Rev. **74**, 496 (1948).

[7] Schuette, Zucker, and Watson, Rev. Sci. Instr. **21**, 1016 (1950).

[8] Lane, Fairbank, Aldrich, and Nier, Phys. Rev. **73**, 256 (1948).

[9] B. V. Rollin and J. Hatton, Phys. Rev. **74**, 508 (1948).

[10] K. W. Taconis, Ned. Tydschr. Natuurk. **16**, 101 (1950).

[11] B. N. Eselson and B. G. Lazerev, Doklady Adad. Sci. (U.S.S.R.) **20**, 748 (1950).

[12] B. V. Rollin and J. Hatton, Phys. Rev. **74**, 508 (1948).

[13] We have found that a very simple and effective way of measuring the height of liquid is to observe the change in current flowing in a composition resistor of nominal value of 300 ohms when it becomes wet with liquid, with a constant voltage of 10 volts applied to the circuit. The response is both rapid and large, the current changing by at least a factor of two.

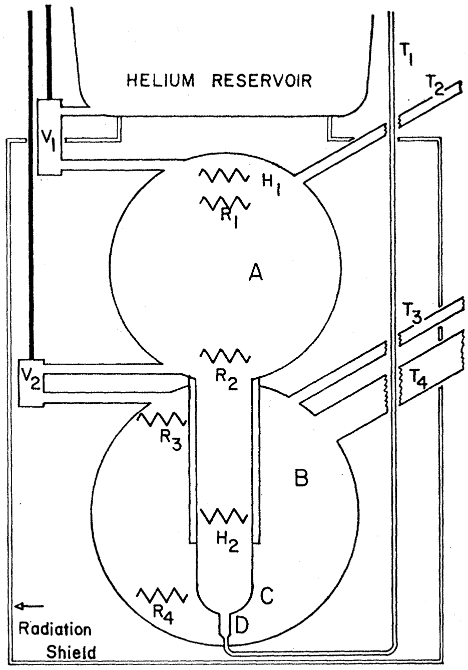

FIG. 1. Schematic diagram of He^3 separator. V_1V_2 values; H_1H_2 heaters; $R_1R_2R_3R_4$ carbon resistors; T_1 tube to Toeppler pump; T_2T_3 tubes to manometers; T_4 tube to pump.

the He^3 originally present will be collected in the small tube D.[14]

(5) The He^3 is now removed from D by opening a valve between tube T_1 and an external Toeppler pump of one-liter capacity. As evaporation takes place from the surface of the liquid in T_1, a second "heat flush" stage of separation takes place, since the surface is cooled by the evaporation, and consequently there is a flow of heat (and with it, of He^3) from D to the evaporating surface. The valve is left open until a total of 1000 cc of gas at approximately 20-mm Hg pressure have been collected in the Toeppler pump at room temperature.

(6) After drawing off the sample into the Toeppler pump, the "waste" He^4 in A is drained into B, which by this time has been sufficiently emptied by continual pumping to accept the liquid from A. Bulb A may now be refilled from the reservoir by opening V_1, and the previous process repeated from step 3.

An alternative method of producing the heat flush after the first filling is to fill bulb A very slowly by just cracking V_1. In this case the liquid evaporates on leaving V_1, and recondenses on the surface of the liquid remaining in A. If this liquid is maintained below the λ point, this recondensation is a heating process, and thus performs the same function as heater H_1. Both methods of flushing work satisfactorily.

III. RESULTS

(1) A single flush yields 20–25 cc of gas at NTP, of which approximately $\frac{1}{2}$ percent is He^3 corresponding to an enrichment factor of 3×10^4. The He^3 concentration is determined by differential pressure measurements between the sample and pure He^4, above the λ point, assuming the validity of Raoult's Law.[15–19]

(2) Within experimental error of measurement, all of the He^3 originally present in the liquid was collected in the Toeppler pump, assuming an original concentration of He^3 of 1.5×10^{-7}.

(3) With the apparatus described, it is possible to perform approximately 3 complete flushes per hour, thus keeping up with a liquefaction rate of somewhat over 2 liters/hour.

IV. DISCUSSION

Several factors must be carefully considered in the design of the draw-off tube T_1 in order to assure that He^3 will travel from D to the surface of the liquid in T_1. In the first place, the size of the tube in this region must be kept to a minimum so that the amount of heat removed in the evaporation will be sufficient to flush all of the He^3 from D. Also, heat conduction down the tube T_1 from the warm upper end to the liquid surface in T_1 must be minimized. This is accomplished by anchoring T_1 at a point a short distance above the liquid surface to a copper strip making contact with the main reservoir at 4°K. Furthermore, the rate at which the Toeppler pump withdraws the gas must be considerably greater than the rate at which the creeping film evaporates in T_1, in order to prevent undue dilution of the He^3–He^4 mixture. To accomplish this, the diameter of the tube T_1 above the anchor point is made much larger than at the bottom.

The enrichment factor of 3×10^4 is not necessarily the maximum attainable since it has not as yet been determined that it is necessary to draw off as large a volume of gas as here reported in order to collect all of the He^3. Also, it should be possible to collect in C the He^3 resulting from several fillings of A before withdrawing the sample into the Toeppler pump thus speeding up the entire process considerably when large amounts are to be processed. In order to hold the He^3 in C during step 6, heater H_2 is turned on during this draining. Experiments to date using this multiple flush method have not been as effective in recovering all of the He^3 originally present as when it is removed

---

[14] Reynolds, Fairbank, Lane, McInteer, and Nier, Phys. Rev. **76**, 64 (1949).

[15] Fairbank, Reynolds, Lane, McInteer, Aldrich, and Nier, Phys. Rev. **74**, 345 (1948).

[16] Eselson, Lazarev, and Alekseevski, J. Exptl. Theoret. Phys. (U.S.S.R.) **20**, 1055 (1950).

[17] J. G. Daunt and C. V. Heer, Phys. Rev. **86**, 205 (1952).

[18] Abraham, Osborne, and Weinstock, Phys. Rev. **80**, 366 (1950).

[19] H. S. Sommers, Jr., Phys. Rev. **88**, 113 (1952).

after each cycle, probably because of a certain amount of backdiffusion of He^3 into the liquid being drained from A to B.

For larger-scale operation, there is no reason why the present design of separator should not be scaled up to take care of any given larger available rate of liquefaction. The number of flushes which can be performed per hour is limited by the rate at which heat can be transferred from A to B, consequently the critical dimension is the area of the cup C.

It should also be observed that with careful design, at least 50 percent of the liquid helium processed remains as a "by-product," and can be used for other cryogenic experiments.

PHYSICAL REVIEW VOLUME 91, NUMBER 5 SEPTEMBER 1, 1953

The Magnetoresistance of Bismuth Crystals at Low Temperatures

P. B. Alers and R. T. Webber
U. S. Naval Research Laboratory, Washington, D. C.

(Received January 22, 1953)

Using bismuth monocrystals at 4.2°K, the magnetoresistive behavior of the metal has been investigated in fields ranging to 100 000 gauss. The crystals were mounted transverse to the field with the principal crystalline axis parallel to the specimen axis. It is observed that the magnetoresistance oscillates at twice the frequency of the susceptibility oscillations of the de Haas-van Alphen effect. The resistance oscillation is separated into two frequency components, one twice the other, and each component is related to a group of electrons having a certain effective mass. The group which is thought to be primarily responsible for the de Haas-van Alphen effect produces magnetoresistance oscillations of small amplitude and of the same frequency. The other group yields larger amplitude resistance oscillations of twice this frequency. Resistance minima have been found at fields of 23.2, 12.5, 8.8 and 6.5 kilogauss when the field is perpendicular to a binary axis of the crystal. The results are discussed in terms of the theory of Davydov and Pomeranchuk, and yield a degeneracy temperature of about 210°K.

INTRODUCTION

In general, when a metal is placed in a magnetic field, its resistance is observed to increase. This increase is usually monotonic, having at first a roughly quadratic dependence on field which gradually becomes linear as the field is increased.[1] As a function of temperature,[2–5] the effect is proportional to $1/\rho$, where ρ is the resistivity. Thus, at helium temperatures one should expect the magnetoresistance to be quite large. It is at these temperatures that anomalies in the magnetoresistive behavior of bismuth[6] have been observed, and similar, though much weaker, effects in zinc[7] and tin.[8] The bismuth anomalies are such as to suggest that the magnetoresistance contains an oscillatory component, and the proposal has been made[9] that this is linked to the susceptibility oscillations of the de Haas-van Alphen effect.[10] Indeed, the discovery of such behavior in bismuth prompted de Haas and van Alphen to make their original investigation of the susceptibility.

The experiments reported here were undertaken to extend existing data to higher fields and to attempt to establish an experimental correlation between the anomalies in the magnetoresistance and those of the susceptibility.

PREPARATION OF SPECIMENS

The magnetoresistive effect in bismuth is quite sensitive to impurities, especially at low temperatures, and care was taken to obtain pure material and prevent contamination during the process of growing single crystals. The bismuth used in crystals 4 through 7 was obtained from the Cerro de Pasco Copper Company and was stated by them to be 99.999 percent pure. The work of Shoenberg and Uddin[11] on the de Haas-van Alphen effect indicated that small amounts (~0.01 percent) of certain impurities affected the susceptibility oscillations considerably, and it appears that the same is true for the magnetoresistance. For crystal No. 3, reported earlier,[12] Johnson & Matthey bismuth (Lot No. 2824) was used and the crystals grown less carefully. We have concluded, in the light of subsequent

---

[1] P. Kapitza, Proc. Roy. Soc. (London) **A119**, 358 (1928); **A123**, 292 (1929).

[2] D. K. C. MacDonald and K. Sarginson, Repts. Progr. Phys. **15**, 249 (1952) give a resume of the work done on magnetoresistance and the Hall effect, with an extensive bibliography.

[3] E. Justi and H. Scheffers, Physik. Z. **39**, 105 (1938).

[4] M. Kohler, Physik. Z. **39**, 9 (1938).

[5] J. W. Blom, Physica **16**, 144–182 (1950). The second article (pp. 152–170) gives a discussion of the Kohler diagram and its application to gallium.

[6] de Haas, Blom, and Schubnikow, Physica **2**, 907 (1935); see also L. W. Schubnikow and W. J. de Haas, Proc. Acad. Sci. Amsterdam **33**, 130, 363, and 418 (1930).

[7] N. M. Nachimovich, J. Phys. (U.S.S.R.) **6**, 111 (1942).

[8] E. S. Borovik, Doklady Akad. Nauk. (S.S.S.R.) **69**, 767 (1949).

[9] W. J. de Haas, Nature **127**, 335 (1931).

[10] W. J. de Haas and P. M. van Alphen, Leiden Comm. **212a** (1930) and **220b** (1932).

[11] D. Shoenberg and M. Z. Uddin, Proc. Roy. Soc. (London) **A156**, 687 (1936).

[12] P. B. Alers and R. T. Webber, Phys. Rev. **84**, 863 (1951).