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The Nonparticipation of He^6 in the Superfluidity of He^4

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In a steady-state experiment, a stream of He^4 containing an extremely small concentration of He^6 has been cooled below the lambda point and passed through a narrow slit as superfluid. If the He^6 atoms had passed through the slit, most of them would have been detected by their radioactive decay in the gas stream leaving the cryostat. The fraction actually found was 0.04 ± 0.07 (standard deviation) of those expected if there had been no separation of the isotopes. This result is consistent with the Bose-Einstein condensation model for the superfluidity of He^4. A new type of generator for He^6 is described.

INTRODUCTION

THE striking differences in properties[1] between liquid He^3 and liquid He^4 have been widely interpreted as evidence that the superfluidity of the latter is essentially the result of Bose-Einstein condensation. However, the relatively large mass difference and the importance of zero-point energy in these systems make it desirable to attempt some independent confirmation of this hypothesis. The isotope of mass 6 has a half-life[2] of 0.82 sec, which, though short, allows the possibility of determining its behavior in the presence of superfluid He^4. We chose to measure the fraction of He^6 carried with He^4 through narrow channels below the λ point, because only a flow-type experiment seemed to have much chance of success, and because a critical property was thus being investigated.

One would predict that He^6 in dilute solution, like He^3, would not be transported with superfluid He^4, if superfluidity is basically a property of a system of identical atoms consisting of an even number of fundamental particles. On the other hand, if the only relevant properties are the interatomic forces and the atomic mass, then He^6, unlike He^3, should participate in the superflow of He^4 and be found unchanged in concentration (except for radioactive decay) in the material emerging from the channel. However, we should remark that Landau and Pomeranchuk[3] have predicted that He^3, He^6, and any other dilute impurities would not participate in superflow of He^4, from a model which does not invoke Bose-Einstein statistics.

METHOD AND APPARATUS

The experiment consists of establishing a steady state in which He^6, generated by the reaction $Be^9(n,\alpha)$-He^6, is carried by a stream of ordinary helium to a narrow slit kept at about 2°K. The He^4 liquefies, passes through the slit as superfluid, evaporates, and is warmed again to room temperature. The rate of He^6 generation is monitored continuously by passing the gas stream through an annular Geiger counter between the generator and the cryostat. Another annular counter of large volume after the cryostat records the decay (to Li^6, with the emission of betas of maximum

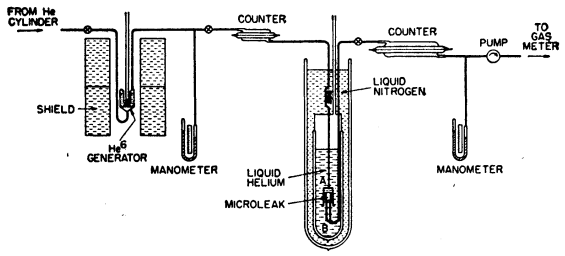

FIG. 1. Schematic diagram of apparatus.

[1] Abraham, Osborne, and Weinstock, Science **117**, 121 (1953); J. G. Daunt, Advances in Phys. (Phil. Mag. Supplement) **1**, 209 (1952).

[2] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[3] L. Landau and I. Pomeranchuk, Doklady Akad. Nauk S.S.S.R. **59**, 669–70 (1948).

energy 3.5 Mev) of a large fraction of those He^6 atoms which have survived passage through the slit.

The apparatus design was dictated by several conflicting requirements. A large rate of gas flow is obviously desirable to shorten the transit time between counters. However, the rate is limited by the slit cross-sectional area. Slits greater in width than about 1 micron would permit flow not typical of true superfluidity,[4] while increasing the slit circumference requires a large volume at low temperature and a correspondingly long hold-up time. There is also a limitation on the rate imposed by the need to transfer to or from the bath the heat content of the gas in heating or cooling, the heat of vaporization, and the thermomechanical heat. The pressure in the parts of the system in the helium bath cannot greatly exceed the bath pressure without causing accumulation of liquid, yet the pressure at the exit counter must be large enough to allow a major fraction of the He^6 to decay there. The volumes of all parts of the system must be kept small to reduce the transit time. The dimensions of the microleak, of the connecting tubing, and of the Geiger counters are all determined within narrow limits by the foregoing requirements, together with such other "boundary conditions" as pump capacity, counter shielding, available neutron intensity, etc.

The apparatus is shown schematically in Fig. 1. The neutron source, a hollow sphere of beryllium coated internally with polonium and covered with nickel, was loaned to us by the Los Alamos Scientific Laboratory. During the measurements reported, its strength was about 3 curies, and it emitted about 3.5×10^6 neutrons/sec. Its great advantage over a radium-beryllium source is, of course, the freedom from gamma radiation, which enables an adequately shielded counter to be placed within four feet of the source with little rise in background. The decision against using a cyclotron source was based on the great inconvenience of transporting the cryogenic apparatus from the low-temperature facilities, as well as the greater steadiness of the radioactive source. The source was shielded for health purposes with an array of square 5-gallon cans filled with saturated sodium metaborate solution; this simple and inexpensive shield proved quite adequate.

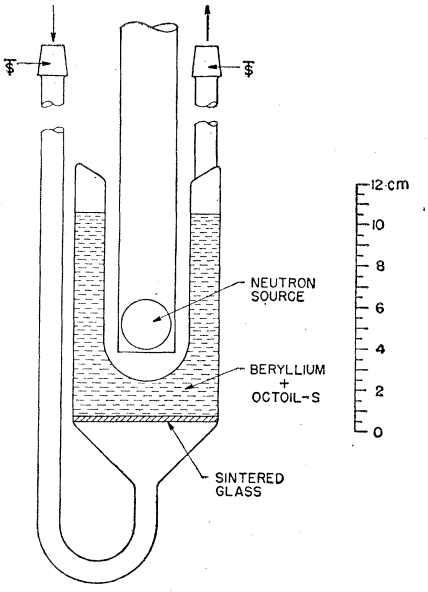

FIG. 2. He^6 generator.

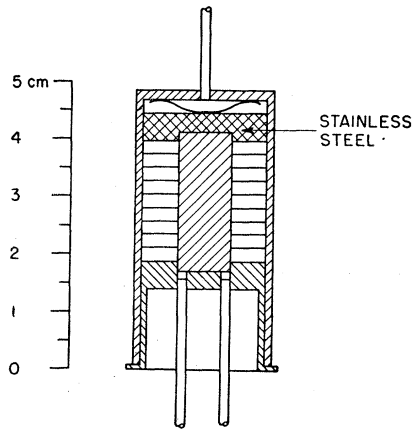

FIG. 3. Microleak assembly.

The He^6 generator is based on the fast-neutron reaction $Be^9(n,\alpha)He^6$. The cross section of this reaction as a function of energy is given by Allen, Burcham, and Wilkinson;[5] its value is about 4×10^{-26} cm^2 from 2 to 4 Mev. A generator arrangement based on that of Allen, Paneth, and Morrish[6] was first tried. In this arrangement 100 g of beryllium powder (-200 mesh) was placed in an annular space surrounding the source and resting on a fritted disk. The He^6 nuclei formed in the reaction recoil and are carried into the gas space. If the average neutron energy is 6 Mev, the recoiling He^6 nucleus carries 2.2 Mev and has a range of approximately 1.8 mg/cm^2 or 10 microns. Thus most of the He^6 atoms are carried out of the particles in which they originate, but in the great majority of cases they re-enter and are stopped in other particles. A stream of helium carries those remaining in the gas phase into the system. The yield of this type of generator was disappointingly small.

A new technique was then tried, in which the beryllium powder was immersed in a nonvolatile liquid (Octoil-S or dibutyl-phthalate), and the helium gas was bubbled through the slurry. In this case most of the He^6 atoms are stopped in the liquid, while the gas volume in the generator is greatly decreased. The rate-determining step is now the transfer of He^6 from liquid to gas. Some data of Sugarman[7] on short-lived fission

[4] R. Bowers and K. Mendelssohn, Proc. Roy. Soc. (London) A213, 158 (1952), and references cited there.

[5] Allen, Burcham, and Wilkinson, Nature **159**, 473 (1947).

[6] Allen, Paneth, and Morrish, Phys. Rev. **75**, 570 (1949).

[7] N. Sugarman (private communication).

products indicated that this might be rapid enough to be useful. After suitable adjustment of conditions, it was found that the addition of liquid improved the yield of He^6 fivefold. The final form of the apparatus is shown in Fig. 2.

The calculation of the fraction of He^6 atoms produced which are recovered in the gas phase is a difficult matter, the chief uncertainties being the energy distribution of the neutrons and the source geometry. A rough value of 0.7 percent was obtained, uncertain by perhaps a factor two.

The Geiger counters used are thin-walled jacketed tubes of "Eck and Krebs" design, the monitor counter having an annular volume of 10.0 cc. The exit counter, designed to provide sufficient time for most of the remaining He^6 nuclei to decay in it, has a counter volume of 500 cc and an annular volume of 800 cc. Its wall thickness was not measured directly, but comparison of counting rates with the small counter in series led to a correction factor of 0.52 ± 0.02 for the difference in wall thickness (and geometry, although this effect was small), suggesting a wall thickness of about 275 mg/cm^2 for the large counter.

In preliminary studies, the half-life was approximately verified, and it was shown that the fraction of longer-lived activities (such as O^{15}) at the monitor counter was negligible.

The cryostat, of the design standard in this laboratory, consists of a helium Dewar, 7 cm i.d. by 65 cm long, surrounded by a liquid nitrogen bath. The helium bath pressure is regulated by manual adjustment of a needle valve and is measured by a mercury manometer and a manometer filled with Octoil-S. The helium stream leaving the monitor counter passes through 75 cm of copper tubing, 0.48 cm o.d., 0.30 cm i.d., immersed in the liquid nitrogen, through the cap of the inner Dewar, and then through stainless steel tubing, 0.16 cm o.d., 0.13 cm i.d., to the microleak, which is near the bottom of the helium bath. The exit tube in the helium space is also 0.16 cm o.d., but is larger in the parts expected to be at higher temperatures.

The helium, specially purified by passing it at 2000 lb/in.^{2} over charcoal at liquid nitrogen temperature, is supplied through a pressure regulator and needle valve, the settings of which determine the rate of gas flow. Mercury manometers measure the absolute pressure near each of the counters. Octoil-S manometers connected to the system close to the microleak (points A and B, Fig. 1) can be arranged to show either the absolute pressure or the difference from the helium bath pressure. A mechanical pump (capacity 33 l/min) is connected to the exit counter through a rubber tube. By constricting the tube it is possible to adjust the pressure in the counter to a suitable value. The pump discharge leads to a wet-test gas meter.

The microleak design as finally evolved is shown in Fig. 3. A pile of 9 copper washers and a stainless steel cap are centered by a copper cylinder. The assembly is contained in a closely fitting copper case soldered together with Rose's metal at a joint far from the washers. Moderate pressure is applied to the pile by a phosphor-bronze leaf spring. The plane surfaces of the washers and cap and the face of the case on which the end washer rests were polished flat to about $\frac{1}{4}$ wavelength of visible light (0.1 to 0.2 micron), and the superflow takes place in the narrow gap between these surfaces. By measuring the rate of gas flow through the leak at known pressures and temperature, an average slit gap can be calculated from the formula for flow between parallel plates, which applies approximately:

$$\dot{v}_2 = wh^3(p_1^2 - p_2^2)/(24\eta L p_2).$$

Here w, L, and h are the total width, length, and gap of the slit, η is the gas viscosity, p_1 and p_2 are the pressures at the ends of the slit, and $\dot{v}_2$ is the volume rate of flow, measured at p_2. For L, we took the difference between the inner and outer radii of a washer and for w the inner circumference of a washer multiplied by the number of gaps, namely 10. The value of $[\langle h^3\rangle_{Av}]^{\frac{1}{3}}$ obtained this way was always in the range 1 to 2μ, based on measurements at room temperature and at the boiling points of nitrogen and helium.

RESULTS

It was found that the apparatus could be operated successfully only in the temperature range from about 1.88 to 2.00°K. At higher temperatures the transit time was too great due to the higher vapor pressure

TABLE I. Summary of results.

Average temperature, °K	Total duration of flow, min	Average flow rate, cc/sec (STP)	Average transit time, sec	Exit counting rate Expected min^{-1}	Exit counting rate Observed min^{-1}	Ratio observed/expected
1.98	100	2.75	3.2	15.0±0.6	1.4±1.6	0.09±0.11
1.93	20	2.9	2.9	21.4±1.2	4.0±3.9	0.19±0.18
1.91	60	2.1	3.4	11.5±0.5	8.6±2.2	0.75±0.19*
1.90	41	2.2	3.2	13.1±0.6	0.3±2.4	0.02±0.18
1.89	100	2.2	3.1	15.0±0.7	−0.8±1.7	−0.05±0.11
				Weighted mean {all results		0.12±0.06
				Weighted mean {excluding the run marked*		0.04±0.07

and hence greater gas density. At lower temperatures, the pressure drop necessary to force the gas through the connecting tubing was excessive compared to the bath pressure. The results of five series of measurements at various temperatures in this range are given in Table I. Usually, once steady conditions were established, counting periods with flow were taken alternately with background measurements. The decay of He^6 in the monitor counter gave 50 counts/min above a background of 25–30 counts/min; the helium pressure at this counter was 2.5 to 3.0 cm Hg. The background of the exit counter was about 225 counts/min, and the decay of He^6 there could have given an additional 12–21 counts/min, as shown in Table I; the operating pressure was 6 to 10 mm Hg. The method of calculation is described in the next paragraph. The pressure at the slit entrance was between 0.3 mm of oil greater and 0.5 mm less than the bath pressure. The pressure drop across the slit was 1 to 8 cm of oil, and hence the *maximum* flow of gas, as calculated from the performance at 4.2°K, was about 1 percent of that due to the flow of superfluid. Operation with the pressure *less* than the bath pressure was probably possible due to capillary condensation in the narrower parts of the slit; this is confirmed by the fact that the observed flow was only about $\frac{1}{5}$ of the superflow which should occur in a slit of these dimensions. Under these conditions, one is assured that no liquid is present except in the slit and that the amount of gas adsorbed on the walls of the system is negligible.

Let t_1, t_2, and t be the times required for the gas to traverse the first counter, the second counter, and the system between the counters. Then, if $\dot{C}_1$ is the observed counting rate at the first counter, it is easy to show that the counting rate at the second counter, assuming no separation of He^6 and He^4, will be

$$\dot{C}_2=\dot{C}_1 e^{-\lambda t}\frac{1-e^{-\lambda t_2}}{e^{\lambda t_1}-1},$$

where λ is the decay constant of He^6. The times t_1 and t_2 can be obtained simply from the known volumes of the counters, the observed flow rate and the pressures at the counters. To calculate the values of t given in Table I, the connecting tubing was divided into 6 regions of known volumes: two at room temperature (one on each side of the leak), two assumed to be at 78°K, and two at the helium bath temperature. The pressure in each region was assumed to be uniform and equal to that shown by the nearest manometer, or, for each region at 78°K, to be the mean of the values on the two manometers on the same side of the leak. The microleak itself had a small volume, calculable with sufficient accuracy from its dimensions, and assumed to contain gas at the bath temperature. The time spent by the superfluid in traversing the slit is certainly less (since the liquid must evaporate before going the entire length) than the value obtained by dividing the slit length (0.6 cm) by the critical flow velocity[8] at these temperatures (~6 cm/sec). The total time t is then just the quantity of He^4 in the system between the counters divided by the rate of flow as measured by the gas meter and a timer.

A check on the amount of gas in the system under the operating conditions was obtained as follows. At the end of a flow period, the oil manometers were cut off from the system by closing stopcocks, and two stopcocks, one just before each counter, were turned off as nearly simultaneously as possible (probably within 0.1 to 0.2 sec). The remainder of the system was pumped to a good vacuum, and the gas trapped between the stopcocks was then pumped out and collected over water in an inverted graduated cylinder. Except for small known quantities of gas at room temperature, the volume measured in this way was that between the counters plus that of the first counter plus that in the leads to the manometers. The correction for the last contribution was unfortunately not at all negligible and was also subject to some uncertainty, since we do not know the temperature distribution above the bath. Since the heat leak down each tube can be calculated[9] to be only about 1 percent of the observed total heat leak into the bath, it seems likely that essentially the entire lengths of the manometer leads in the double-walled portion of the inner Dewar were cooled nearly to the bath temperature by the evaporated gas even when the Dewar was only half-filled with liquid. When the measured volume exceeded that calculated on the preceding assumptions by more than 1 cc (STP) (out of about 15 to 20), the corresponding run was rejected.

Earlier experiments with a different design of microleak, a disk fabricated by pressing, sintering, and rolling copper powder (−325 mesh), gave anomalous results. Below the λ point no He^6 was observed to pass the microleak, in agreement with the result using the slit. Above the λ point, however, a leak of the same design but pressed and rolled less, so that a large gas flow was possible, allowed no He^6 to reach the exit counter at 2.5°K. At higher temperatures there was an increased passage of He^6, and at 4.2°K the expected amount was observed. Attempts to explain this effect as due to adsorption, with concentration of He^6 in the adsorbed phase resulting in a long transit time, were not very successful, and the behavior with this type of leak remains obscure.

Experiments in which the microleak assembly was replaced with an open tube gave the expected exit counting rate both above and below the λ point.

The errors assigned in Table I are the standard deviations due to counting statistics. It is difficult to estimate the error in the transit time. If we have overlooked as much as about 2.5 cc (STP) of gas (which

[8] L. Meyer and W. Band, Naturwiss. **36**, 5–16 (1949); L. Meyer and J. H. Mellink, Physica **13**, 197 (1947).
[9] R. Berman, Phil. Mag. [7] **42**, 642 (1951).

seems unlikely), the ratios in Table I would all be approximately doubled, and so would their standard deviations.

DISCUSSION

It appears that He^6 behaves like He^3 in dilute solution,[10] in that the concentration of He^6 in material emerging from a fine channel is only a few percent at most of the concentration at the entrance. This is just what would be expected on the Bose-Einstein gas model for liquid He^4. The He^6, although it obeys Bose-Einstein statistics, is present at such a low concentration ($\sim 10^{-19}$ atom fraction) that its degeneracy temperature ($\sim 10^{-12}$ °K) is far below the temperature of the experiment. Since it is distinguishable from He^4, it cannot participate in the condensation of the latter. Hence it behaves like any other foreign solute in being excluded from the superfluid state.

In the present state of theoretical knowledge of liquid helium, it is perhaps still possible to imagine that the conditions for superfluidity are so stringent that there exist both *upper* and *lower* limits for the atomic mass, and that He^4 satisfies these conditions while He^3 and He^6 do not. However, the most natural interpretation of our results, taken together with the extensive work on He^3, is that superfluidity is connected essentially only with the symmetry properties of the wave function of a system with respect to the interchange of identical atoms, and not with the atomic mass.

[10] Abraham, Weinstock, and Osborne, Phys. Rev. **76**, 864 (1949).

ACKNOWLEDGMENTS

We are deeply indebted to the Los Alamos Laboratory and the Argonne National Laboratory for their cooperation in supplying us with the polonium-beryllium neutron source used in this work. The microleak was constructed by Mr. Paul Dolmer, under the supervision of Mr. Joseph Getzholtz. The support of the industrial sponsors of the Research Institutes is gratefully acknowledged.

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Exchange Coupling in $CuK_2Cl_4 \cdot 2H_2O$ by Paramagnetic Resonance Absorption

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$CuK_2Cl_4 \cdot 2H_2O$ contains two inequivalent Cu^{++} ions in each unit cell. At $\lambda = 5.4$ mm two peaks are obviously found corresponding to two different g values, whereas only one peak is found at longer wavelengths. From these facts the exchange frequency between these dissimilar ions is estimated to be about 0.1 cm^{-1}. For the crystal orientation relative to the magnetic field at which the two types of ions are similar, the line width decreases with decreasing wavelength. This may be the result of the "10/3 effect" discussed by Anderson and Weiss.

MEASUREMENTS of microwave paramagnetic resonance absorption in $CuK_2Cl_4 \cdot 2H_2O$ at $\lambda = 10$ cm, $\lambda = 3$ cm, and $\lambda = 1.5$ cm have already been reported.[1,2] As the crystal has two inequivalent ions we can expect to observe two absorption maxima. We found only one maximum at these wavelengths, with, however, a remarkable frequency dependence of the line width. This may be ascribed to exchange coupling. We have recently investigated this salt at $\lambda = 7.7$ mm and $\lambda = 5.4$ mm, and two maxima were found at $\lambda = 5.4$ mm.

The crystal structure of $CuK_2Cl_4 \cdot 2H_2O$ is tetragonal and each unit cell contains two inequivalent Cu^{++} ions situated at the points (0 0 0) $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$.[2,3] Both ions are surrounded by four chlorine atoms and two water molecules. The chlorine atoms are on a rhomb in the *a-a* plane, the two rhombs in the unit cell being rotated relative to each other by 90° about the *c* axis. Thus the electric field acting on the Cu^{++} ion has rhombic symmetry, and two different g values would be expected at those orientations of the cyrstal at which the

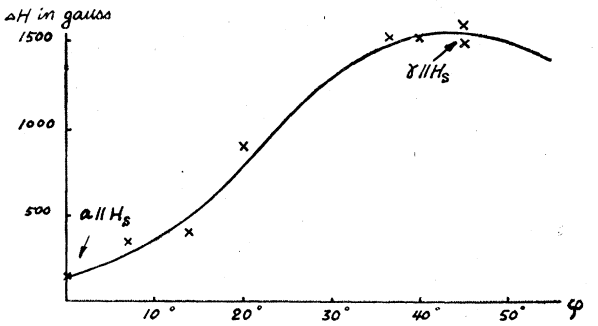

FIG. 1. Variation of line width with orientation for H_s in the *a-a* plane at $\lambda = 7.7$ mm, where φ is the angle between H_s and the *a* axis.

[1] Itoh, Fujimoto, and Ibamoto, Phys. Rev. **83**, 852 (1951).
[2] H. Kumagai *et al.*, J. Phys. Soc. Japan **7**, 535 (1952).
[3] L. Chrobak, Z. Krist. **88**, 35 (1934).