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EXPERIMENTS ON THE DETECTION OF THE TRANSITION OF He³ INTO THE SUPER-FLUID PHASE

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An instrument with three-stage demagnetization of paramagnetic salts is constructed, utilizing tin superconducting thermal switches. With the aid of this instrument, the temperature dependence of the specific heat of liquid He³ was determined in the temperature range from 0.0035 to 0.015° K. A significant increase in the specific heat of He³, roughly twice that expected on the basis of an assumed linear temperature dependence, was observed at a temperature between 0.005 and 0.006° . The results obtained may be explained by the transition of He³ into the superfluid phase at 0.0055° K.

As already mentioned earlier, [1] the question of whether there is a superfluid state of He³, analogous to that of He⁴, immediately arises as soon as one begins to study the properties of liquid He³. Both He⁴ and He³ are quantum liquids (according to the relation $\Delta x \Delta p > h$, at $T = 2^{\circ} K$ the uncertainty in the position of an individual atom is $\Delta x \sim h/\sqrt{3mkT} = 10$ Å, but the distance between atoms is 4 Å); however, He^4 has spin equal to zero and obeys Bose statistics, but He³ has a nonzero nuclear spin and obeys Fermi statistics. In this connection, one can regard the thermal motion in liquid He⁴ as a collection of thermal excitations – rotons and phonons, whose number decreases to zero. In liquid He³ it appeared that, because of the presence of nuclear spin, the number of thermal excitations must remain constant and equal to the number of atoms, all the way down to absolute zero. The latter circumstance has led to the conclusion that observation of superfluidity in He³ is very improbable. Buckingham,^[2] developing an idea of Rice, ^[3] arrived at the conclusion that

superfluidity does not appear either in an ideal Fermi gas or in Rice's model. However, after the development of the theory of superconductivity by Bardeen, Cooper, and Schrieffer, ^[4] and by Bogol-yubov, ^[5] the appearance in liquid He³ of a mechanism analogous to the "pairing" of electrons appeared very probable to us, and consequently its transition into a superfluid phase ^[6] also seemed quite likely. That is when the experiments described below were begun. Exactly the same point of view was justified theoretically by Cooper, Mills, and Sessler, ^[7] and by Pitaevskii, ^[8] and later on by a number of other authors.

A rapid increase of the thermal conductivity and viscosity with decreasing temperature is a property of liquid He³ to be expected according to the theory of a Fermi liquid proposed by Landau.^[9] According to the measurements of Anderson, Salinger, and Wheatley^[10] the thermal conductivity of He³ for T < 0.04° K is equal to $\kappa = 4.8 \times 10^{-6} \text{ T}^{-1} \text{W/cm-deg}$, and according to the data of Abel, Anderson, and Wheatley^[11] the viscosity amounts to $\eta = 2.8 \times 10^{-6} \text{ T}^{-2}$ poise for T < 0.07°K. Therefore, at very low temperatures it would be impossible to expect a sharply delineated anomaly in the heat transfer, as occurs at the lambda-point of He⁴, and it was decided to look for an anomaly in the temperature dependence of the specific heat of He³ in order to detect its transition into the superfluid state.

A sphere of lightly compressed cerium magnesium nitrate (CMN) powder containing liquid He^3 in its pores was selected as the refrigerating medium and thermometer. The sphere was the third stage of adiabatic demagnetization and served as a calorimeter. Since, according to estimates, the specific heat of He^3 must be very small, the experimental arrangement must guarantee negligible (of the order of a few ergs) "parasitic" heat influxes, and because of the presence of a large thermal resistance at the boundary between a solid and liquid He^3 , the contact surfaces must be highly developed. The instrument described below meets these conditions.

DESCRIPTION OF INSTRUMENT AND ARRANGEMENT

A metallic dewar with a capacity of two liters of liquid helium and two liters of liquid nitrogen was used for the experiments. The lower part of the dewar is shown in Fig. 1. Bath 1 containing liquid He³ was placed inside the dewar 2 which was filled with liquid He⁴, whose temperature was maintained at about 1.3° K. The dewar containing He⁴ was surrounded by a polished copper casing 3 which is connected at the top to the bath containing liquid nitrogen. Both the dewar containing He^4 as well as the nitrogen bath were located in a single vacuum space, bounded at the bottom by the copper casing 4. The lower part of the dewar containing He⁴ was fabricated out of thin-walled stainless tubing. He³ was pumped out of the bath by a diffusion pump with the aid of the arrangement described earlier.^[12] The temperature in the bath was maintained at about 0.3° K.

One end of the tin thermal switch 5 was soldered to the bottom of bath 1, a cluster of 640 copper wires of 50 μ diameter was soldered to the other end. The tin thermal switch was assembled out of strips of 0.04 mm thick foil, total cross section 3.8 mm², and length 25 mm. The purity of the material of the foil for the switches was estimated to be 99.9998%.

The first cooling stage is pellet 6 with 60 g of ferric ammonium alum, into which two identical bundles of copper wires were pressed. The pellet



FIG. 1. Three-stage demagnetization instrument (see text for call-outs).

was coated on top with epoxy resin mixed with finely powdered quartz (45% quartz by weight).¹)

The first stage of demagnetization was connected with the second stage of demagnetization 8 in the same way by the tin thermal switch 7 $(l = 20 \text{ mm}, \text{s} = 1.6 \text{ mm}^2)$. The second stage consisted of a set of disks of thickness between 2 and 2.5 mm, cut out of CMN single crystals, and disks of 25 mm diameter cut out of 50 micron copper foil with strips 9 leading up and down. The disks were smeared with silicone valve grease and tightly compressed. The weight of the CMN crystals was equal to 24 g. The capsule for the second stage of demagnetization was also covered with epoxy resin containing quartz.

The thermal switch 10 of length 16 mm and cross section 1.7 mm^2 was assembled, like the first two switches, out of strips of tin foil, and was soldered to the strip of copper foil 9 and to the copper frame 11. (The thermal switches were

¹⁾The prescription for preparation of the coatings was developed in the Technology of Materials Section of the Institute of Physics Problems, under the direction of N. N. Mikhailov.

soldered with tin of the same purity as the switches themselves.) The frame was pressed into sphere 12 of fine CMN crystals, whose average size was about 10 μ , and total weight was 14.1 g. The volume of the sphere amounted to 8 cm^3 , and the volume of the pores filled with liquid He³ was equal to 1.12 cm^3 . In order to equalize the temperature inside the sphere, 640 copper wires 13 of length 20 mm and diameter 50 μ were silver soldered to the frame 11. The total weight of copper in the sphere was 1.55 g. Fine filaments were wound on the outside of the sphere, and it was covered with a mixture of 45% by weight finely powdered quartz and 55% epoxy resin. The total weight of the casing was 1.6 g. In order to ensure that the epoxy resin casing 14 remains vacuum tight and does not crack upon cooling down to very low temperatures, it was subjected to polymerization. For this purpose, the sphere was cooled down to between 0 and -10° C and for several seconds placed in an oven with a temperature of 500° C, then immersed in liquid nitrogen. Then again cooled down to between 0 and -10° C, and again placed in the oven for several seconds. Such an operation was repeated ten times.

To prevent the sphere from touching the walls, a spring-stretched caprone bracing string 15 was fastened at the bottom. In order to decrease "parasitic" heat influx, the sphere was placed inside the shield 16. The shield was cemented together with BF glue out of strips of 50 μ thick copper foil, and was cemented to the second capsule 8. The shield was made laminated in order to not create short-circuited loops which would impede the measurement of the magnetic moment of the sphere. In order to center the sphere and shield, a sleeve containing three sharpened Plexiglas points 17 was installed at the bottom.

The temperature of the sphere was determined from the magnetic moment of the CMN crystals with the aid of a ballistic galvanometer and two bucking identical ballistic coils 18. The coils had up to 7500 turns of enameled copper wire of 60μ diameter. The measuring field was produced by a solenoid located on the lower part of the instrument. During readings, the current in the solenoid was reversed. The magnetic field of the solenoid was chosen such that the deflections of the ballistic galvanometer were of the order of 100 to 250 mm; here the field varied from 0.5 Oe at low temperatures up to 5 to 8 Oe at temperatures of 0.3° K and above.

In order to monitor the cooling of the sphere, a carbon thermometer 19, prepared according to the method described earlier, [13] was set up on

the copper tube connecting the sphere with the frame. Unfortunately, at low temperatures the sensitivity of a carbon thermometer decreases with decreasing temperature, and for $T < 0.02^{\circ}$ its resistance remains practically constant. The resistance of the carbon thermometer varied from a few hundred ohms at 1° K up to 40 k Ω at 0.02° K. The measuring current was 10⁻⁷ A. A heater 20, made out of 50 μ constantan, was coiled around the same copper tube.

The sphere was filled with He³ through a German silver capillary 21 with inside diameter 0.125 mm. The capillary passed through the center of both of the upper capsules and basically served to strengthen the entire construction. He³ specially purified of He⁴ in a distillation column was used to fill the sphere; according to an estimate, the amount of He⁴ present in it was less than 0.001%. ²⁾ The helium, stored in a six liter flask, was supplied for the experiment and re-covered with the aid of a mercury Toepler pump.

A carbon thermometer 22 was mounted on the copper tube cemented in pellet 6 in order to monitor the cooling and heating of the first stage of adiabatic demagnetization. Since the heat capacity of the complete system of salts is appreciable, but heat transfer by the thermal switches is rather moderate, about 0.1 cm³ (at 0° C and 760 mm of Hg) of He³ heat exchange gas was put in the space between the capsules and the helium dewar, in order to accelerate the cooling down to liquid helium temperatures. He³ was chosen in order to guarantee proper purity of the studied He³ inside the sphere which, due to diffusion or leakage, might be contaminated by the heat exchange gas. Such an operation enabled us to reduce the time required to cool the salts from liquid nitrogen temperatures down to liquid helium temperatures to one hour. Upon further cooling, activated charcoal 23 mounted on the first pellet adsorbed the exchange gas and guaranteed a good vacuum.

The magnetic field in the instrument was produced by an electromagnet which could be raised, lowered, and rolled away from the instrument. The position and shape of the magnet pole pieces

²⁾There was 0.15% He⁴ present in the original He³. The distillation column operated in the regime in which He³ was enriched from 40 to 99.985%.[⁶] Therefore, there should be $4 \times 10^{-5}\%$ He⁴ present in the purified He³. Since the vessels and connecting tubes in which the purification was carried out were rinsed six times with pure He³, one can guarantee that the concentration of He⁴ in the He³ used was in any case less than 0.001%.

of the electromagnet 24 in its upper position are indicated on Fig. 1 by solid lines.

An iron shield 25 was set up under the pole pieces to make the magnetic field fall off rapidly. The shape of the shield is evident from the cross section shown in the upper right, and the shape of the pole pieces is indicated by the cross section shown in the lower right. Dashed lines indicate the lower position of the magnet and shield. The dependence of the magnetic field at the center of the instrument on height, for the upper position of the magnet, is given at the right; the maximum value of the field is about 11,000 Oe.

When the sphere operated as a calorimeter, it was necessary to guarantee uniform generation of heat throughout the entire volume; this was achieved by gamma-ray irradiation of the sphere. In the first experiments, a Cs^{137} gamma source of activity 4.9 mg equivalent, ³⁾ enclosed in a thinwalled ampoule, was used. Later on two Cs^{137} sources were used: the first with an activity 4.9 and the second with 9.2 mg equivalent. The sources were fastened in 8 mm diameter tubes with little windows cut out to the side of the instrument. The tubes moved freely in holes bored in a 50 mm thick lead plate. The plate, in the form of a horseshoe, was brought close to the instrument on a special stand in such a position that the sphere was in the center of the horseshoe. The holes in the plate were bored horizontally and parallel: one-for the first source-at a distance of 46 mm, the other-for the second source-at a distance of 62 mm. Slots were made in the plate from the bored holes to the center so that, when the tubes were completely pushed in, the sources irradiated the entire sphere with gamma rays, without any of them being absorbed in the lead. The tubes were connected to a strap drawn away from the plate by a spring; the tubes containing the sources were thus placed in a position in which the sphere turned out to be shielded from the sources by a lead layer 5 cm thick. When the tubes were completely pushed in, a relay was actuated, holding the strap in a clamped position, and a contact was closed, sending pulses from a generator whose frequency was stabilized by quartz to the scaler circuit. The scaler circuit counted 10,000 signals and then sent a pulse to the relay, the strap was released, and within a time interval of less than 0.1 sec the sources moved out. Thus, by varying the frequency of the generator, it was possible to

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<sup>3)</sup>The decay scheme is

Cs^{137} \xrightarrow{33} years \beta^{-} + Ba^{137} \xrightarrow{150 \text{ sec}} \gamma (0.663 \text{ MeV}).
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irradiate the sphere with gamma rays during identical (to within 0.1 sec) intervals of time. The irradiation, as a rule, was continued for 60, 80, 120, and 180 sec.

The same type of automatic switching scheme was used in connection with the generation of heat by a current. In the present case, the current and signal for the PS-100 scaler were fed through an RPS 20 relay which, after 10,000 signals, switched over to the second position. In the second position the current went through a parallel circuit of the same resistance, instead of going through the heating coil. Since the time required to actuate the relay was less than 0.05 sec, in this case the amount of heat generated in the heater was the same, to within the accuracy of the current setting (0.3%).

PRELIMINARY EXPERIMENTS

The first experiments were carried out on the instrument with two-stage demagnetization, when a single large stage—about 100 g of ferric ammonium alum—was used instead of the two upper stages. The process of cooling the device in a magnetic field down to 0.35° K took from six to seven hours, then the magnet was lowered, and the sphere was cooled in the magnetic field for about two hours more. After this, the magnet current was gradually turned off and the magnet rolled away. With the aid of such a procedure, one is easily able to cool the empty sphere down to temperatures below 0.0032° K.

During heating caused by heat influx from without or with the aid of the gamma source, the magnetic moment of the sphere at first increased, reached a maximum, and then started to decrease. Since such a dependence for the magnetic moment of CMN was observed earlier by Daniels and Robinson,^[14] and according to their determination the maximum value of the magnetic moment corresponds to a temperature of 0.0032°K, in what follows we took the value of the maximum as a reference point and used a magnetic temperature scale, that is, we assumed $T = A/M = B/(\varphi - \varphi_0)$, where A and B are constants chosen such that the maximum value of M or φ corresponded to T = 0.0032° K. Usually φ_0 was determined from the deflection of the ballistic galvanometer at a moment when the ballistic coils were already at the temperature of liquid helium, but the sphere had not yet been cooled. The measurement magnetization current was then up to ten times larger than at very low temperatures. Usually the calculated φ_0 was of the order of 2 mm, and the total

deflection was about 140 to 250 mm. Such a temperature scale, although it must differ appreciably from the thermodynamic scale in the region $T < 0.006^{\circ}$ K, is most convenient when a sphere of CMN crystals serves as a thermometer. In what follows, all of the data is given in this temperature scale.

We were not able to cool the sphere completely filled with He³ down to temperatures below 0.008° in the two-stage demagnetization instrument. Therefore, experiments were carried out with the empty sphere, with it filled with only 0.05 cm^3 of liquid He³, that is, filled with an amount of liquid He³ sufficient to cover all of the CMN crystals with a liquid film of thickness (3 to 6) $\times 10^{-6}$ cm, and with the sphere 60% filled, that is, 0.725 cm³ of liquid He³. Heating curves for the sphere, continuously irradiated by a single 4.7 mg equivalent source, are shown in Fig. 2 for the three cases cited above. It is evident from the Fig. that the presence of helium at temperatures below 0.0055° leads not to a slower but to a more rapid heating of the CMN. The ratio of the additional heat flow ΔW , entering the CMN crystals in the presence of the helium, to the flow W_{γ} absorbed by the crystals in the empty sphere, is also shown on Fig. 2.

It should be noted that, as one would expect, a gamma counter placed behind the sphere indicated, to within experimental errors (2%), that absorption of gamma rays by the empty sphere and by the sphere filled with He³ was the same. According to an estimate, He³ should increase the absorption by 0.7%.

Thus, the first experiments already led to the conclusion that, at $T \sim 0.005^{\circ}$ He³ transmits heat very effectively from the copper components of the sphere and from its envelope to the CMN crystals. However, the heating curves do not give a definite



FIG. 2. Curves showing the heating of the sphere when it is continuously irradiated by gamma rays: 1 – sphere empty; 2 – 0.05 cm³ of liquid He³ in the sphere; 3 – 0.725 cm³ of liquid He³ (60% of the volume) in the sphere; 2' – $\Delta W/W_{\gamma}$ calculated from curves 1 and 2; 3' – $\Delta W/W_{\gamma}$ calculated from curves 1 and 3.

answer with regard to the existence in He^3 of a transition into a superfluid phase. Since we were not able to cool the sphere completely filled with He^3 with the two-stage demagnetization instrument, the instrument described above, with three stages of demagnetization, was built.

EXPERIMENTS WITH A SINGLE GAMMA SOURCE OF 9.2 mg EQUIVALENT

The experiment was carried out in the following manner on the three-stage demagnetization instrument. Liquid He⁴ was poured into the apparatus, cooled down to liquid nitrogen temperatures, and then right away, before the sphere cooled, a determination of the zero ballistic deflection associated with incomplete compensation of the bucking coils was carried out. The null deflection remained practically constant. Then the magnet was rolled in and set up in its upper position (see Fig. 1) and current was supplied to it. After this, the He^3 was condensed in the dewar and pumped on down to 0.03° K, then a definite amount of He³ was supplied to the sphere, and the instrument was cooled for seven to nine hours. After this time interval, all three stages reached a temperature close to 0.3



FIG. 3. Dependence of the heat capacity of the sphere on temperature according to measurements with a single gamma source: $O = 0.017 \text{ cm}^3$ of liquid He³ in the sphere; $\times -$ the values of O multiplied by 0.6; \bullet , \blacksquare , $\blacktriangle -$ different experiments with the sphere completely filled with liquid He³ (1.12 cm³). The heavy curve is obtained by calculation from the solid and dashed curves. The solid straight line at the bottom corresponds to C = 24T J/deg-mole for He³.

or 0.4° K (which was monitored by the readings of thermometers 19 and 22). Then the magnet was slowly lowered, reaching after 2 to 2.5 hours the position indicated by the dashed lines in Fig. 1. During this time, the upper pellet—the first stage of demagnetization—managed to demagnetize and to cool the second and partially also the third stage. The second stage was also demagnetized, cooling the third stage down to T ~ 0.04 to 0.05^{\circ}. After this, the magnet current was gradually reduced for 40 to 50 minutes, and the magnet was rolled away.

In this way, we managed to obtain a (magnetic) temperature on the order of 0.0033 to 0.0034° in the sphere completely filled with He³. A temperature calibration was carried out according to the maximum value of the magnetic moment of the sphere filled with a small amount of He³. The temperature during the time of the experiment was determined from the ballistic deflection, which was regularly recorded every minute.

After carrying out measurements for 10 minutes and checking that the heating took place slowly, the gamma source was brought up to the instrument and was automatically shut off after a given interval of time. Abrupt jumps were then observed on the curves of deflection vs. time, indicating that the time required for the establishment of equilibrium is less than a minute. The curves obtained were similar to curves 1 and 2 on Fig. 4 (see below).

For a temperature of about 0.025° in the sphere completely filled with He³, calibration of the gamma source was carried out with the aid of the electric heater 20. The heater was turned on for 120 sec

and 380 ergs was released; abrupt jumps with very small "tails," on the order of several minutes, were than observed on the curves of deflection vs. time. The energy absorbed by the sphere during a given time interval of irradiation was determined from measurements of the heating by the gamma source, by current, and again by the gamma source.

The temperature dependence of the heat capacity for the cases when the sphere is filled with 0.017 cm^3 of liquid He³ and when it is completely filled with He^3 is shown in Fig. 3. An experiment with a small amount of He³ was carried out in order to eliminate the influence of surface layers of helium and to improve the heat transfer during cooling of the sphere. As is evident from the data, the reproducibility of the results in three different experiments with the filled sphere is rather good, and, as in the previous experiments, the apparent heat capacity of the sphere containing a small amount of He^3 is 40% larger than that of the filled sphere. It is clear that the heat capacity of the filled sphere can only be larger than that of the empty sphere and, consequently, the presence of He³ leads to an additional heat flow from the copper components and casing of the sphere to the CMN crystals. If we assume that the CMN, in the almost empty sphere, obtains only 60% of the energy which it obtains when the sphere is filled, then the heat capacity in the region $\,T < 0.004^\circ\,$ will be almost the same, and one can ascribe the appearance of differences between them at higher temperatures to the heat capacity of He^3 . The difference between these two heat capacities is represented on Fig. 3 by a heavy curve. The straight line segment at the bottom corresponds to the specific heat

> FIG. 4. Record of the deflections of the ballistic galvanometer (0) and of the galvanometer measuring the resistance of the carbon thermometer (x) (the deflections of the ballistic galvanometer were measured at one minute intervals). $I = 0.058 \text{ cm}^3$ of liquid He³ (× on Fig. 5), heating by gamma sources at periods of 120 sec each; II – 1.12 cm³ of He³ in the sphere (∇ on Fig. 5), heating by gamma sources at periods of 120 sec each; III - a new sphere completely filled (1.05 cm³ of He³; Δ on Fig. 6), heating by current at periods of 80 sec each; IV - the empty sphere, heating by gamma sources (γ) and by current (I), with 60 sec periods; IV' - variation of R of the carbon thermometer plotted simultaneously with ballistic galvanometer measurements (IV); V corresponds to an experiment on the calibration by current of the gamma sources during measurements with the sphere completely filled. The time for heat generation is indicated by arrows.



of He³ according to the measurements of Anderson, Salinger, Steyert, and Wheatley: [15] C = 24T J/mole-deg, and the dashed line is its extrapolation according to a linear law.

As is evident from Fig. 3, in the region between 0.005 and 0.006° the dependence of the specific heat of He³ on temperature undergoes a substantial change, which can be explained as the transition of He³ into a superfluid state; however, the conditions of the experiment did not guarantee uniformity of the temperature over the entire volume of the sphere.

During the performance of the experiments described, the sphere was enclosed in a shield which was connected not with the second stage as shown in Fig. 1, but with the first stage; therefore, the "parasitic" heat flow to the sphere was on the order of 10 to 20 erg/min.

EXPERIMENTS WITH THE SPHERE HEATED BY TWO GAMMA SOURCES AND BY CURRENT

In the previous experiments the gamma-ray source was located on one side of the sphere, at a distance of 46 mm from its center, and this caused considerable nonuniformity of heating. According to calculations, the part of the sphere close to the source absorbed an amount of heat per unit mass up to three times larger than the opposite part. Therefore, it was decided to obtain a smaller "parasitic" heat influx and to irradiate the sphere from two sides. The two available gamma sources of 4.9 and 9.2 mg equivalent intensity on hand were used. Upon placing them on different sides of the sphere, at distances of 46 and 62 mm from the center of the sphere, respectively, the nonuniformity in the absorption of heat by different sections of the sphere was less than 15%. In addition, the shield surrounding the sphere was fastened not to the first but to the second pellet, which enabled us, for a time interval on the order of one to two hours, until the second stage warmed up, to decrease the parasitic heat influx down to between 3 and 6 ergs per minute.

Cooling and demagnetization of the apparatus in these experiments was accomplished in the same way as for the experiments with a single gamma source. Experiments were carried out under the following conditions: I-0.058 cm³ of liquid He³ is condensed in the sphere, an amount sufficient to cover all of the CMN crystals with a film; II-1.12 cm³ of liquid He³ in the sphere at a pressure p = 28 mm Hg-completely filling the sphere; in both cases the heating was accomplished by the two gamma sources; III-1.12 cm³ of liquid He³ in the sphere at a pressure p = 0.5 mm Hg, heating produced by current with the aid of the heater 20; IV—empty sphere, and V—1 cm³ of liquid He³ in the sphere (p = 0), heating by two gamma sources.

In addition, a special experiment was carried out with the sphere completely filled with He³ $(1.12 \text{ cm}^3, \text{ p} = 28 \text{ mm Hg})$ in order to determine the power absorbed by the sphere during irradiation by two gamma sources. In this case, beginning with T = 0.0088° and continuing to T = 0.015°, the following steps were carried out in turn: heating for 120 sec by two gamma sources, then heating by current with release of 138 ergs during 120 sec, again heating by the gamma sources, by the current, and again by the two gamma sources. From this calibration it followed that the sphere filled with He³ absorbs, during irradiation by two sources, just as much heat as during the heating by current, that is, it absorbs 138 ± 6 ergs during 120 sec.

Some of the recordings of the ballistic galvanometer deflections, taken every minute, are presented in Fig. 4. As is evident from Fig. 4, the generation of heat by the gamma sources causes an abrupt change in the magnetic moment of the CMN crystals, and consequently also causes an abrupt change in their temperature. Since there are no "tails" of any kind, one can conclude that thermal equilibrium between CMN and liquid helium is established in a time interval of less than one minute. During the generation of heat in the sphere completely filled with He³ by a current, an abrupt change of the magnetic moment is also observed, but with small "tails" of around 2 to 3 min (curve III). It is evident from curve IV that heating by a current is accompanied by long "tails," that is, by clearly nonequilibrium processes. The simultaneously measured resistances of the carbon thermometer (19 on Fig. 1) indicate (curve IV' on Fig. 4) that the irradiation by gamma sources and especially the generation of heat by a current through the heater are accompanied by a considerable rise in the temperature of the copper components of the sphere: from thousandths of a degree up to between 0.029 and 0.043°. Such warming up was also observed in the sphere filled with He³, but it was appreciably less (curve V on Fig. 4), that is, the conditions were closer to equilibrium.

The results of five experiments on the measurement of the heat capacity of the sphere under different conditions are shown in Fig. 5a. The heat capacity C was calculated as the ratio of the heat absorbed, ΔQ , to the temperature difference, ΔT , corresponding to the jumps on the curves of deflection vs. time: $C = \Delta Q / \Delta T$.



FIG. 5. Dependence of the heat capacity of the sphere on temperature, according to measurements with two gamma sources and with heating by current: $\times -0.058 \text{ cm}^3$ of liquid He³ in the sphere, p = 0; $\nabla - 1.12 \text{ cm}^3$ of liquid He³ in the sphere, p = 28 mm Hg, heating by two gamma sources; $\Delta - 1.12 \text{ cm}^3$ of liquid He³ in the sphere, p = 28 mm Hg, heating by current; • - the empty sphere; $\Box - 1 \text{ cm}^3$ of liquid He⁴ in the sphere, p = 0.

For the sphere completely filled with He³ and irradiated by gamma sources (∇ on Fig. 5a), the value $\Delta Q = 138$ erg was determined as the product of the power absorbed by the sphere (taken from a calibration with respect to current, as described above) times the time of irradiation. For experiments with the generation of heat by current (Δ) , it was considered that at temperatures below 0.01° only 66 ergs out of the 68 generated in the heater pass into the sphere, and 2 ergs were transmitted to the second stage of demagnetization. The reason for so thinking was the fact that small "tails" on the order of 2 to 3 min were observed on the deflection vs. time curves after switching on the current, when the sphere filled with He³ was heated with current. In this case the values for the heat capacity, determined from heating by current and by gamma sources, coincide at $T = 0.0035^{\circ}$.

Starting from the same considerations, in the case of the sphere filled with a small amount of He^3 it was assumed that out of 140 ergs absorbed by all parts of the sphere, 14 ergs were transmitted to the second stage. The results of this calculation are marked on Fig. 5a with the symbol \times . A curve showing the dependence of C on T is drawn through the points obtained. The difference of heat capacity between this curve and the points corresponding to experiments with the sphere completely filled with He³ is marked at the bottom of Fig. 5a by the same symbols as used for the heat capacity of the entire sphere. The depend-ence of the specific heat of He³ on temperature, C = 24 T joule/mole-deg, corresponding to the measurements of Anderson, Salinger, Steyert, and Wheatley, ^[15] is represented at the bottom by the heavy straight line segment. The dashed curve plots the dependence of the specific heat of He³ on temperature, corresponding to the calculations of Soda and Vasudevan, ^[16] if the lower curve is extrapolated into the region of low temperatures assuming $T_{\lambda} = 0.0055^{\circ}$.

The results of measurements of the heat capacity of the empty sphere (\bullet) and of the sphere filled with 1 cm^3 of liquid He^4 (\Box) are also presented in Fig. 5a. For the calculations it was assumed that 130 ergs was absorbed in the sphere during 120 sec of irradiation by the gamma sources. As expected, because of the very small specific heat and thermal conductivity of He⁴ in the range of temperatures investigated, the results of the experiments for the empty sphere and for the sphere filled with He⁴ coincide. However, for $T < 0.005^{\circ}$ the apparent heat capacity of the empty sphere turns out to be appreciably larger than when it is filled with He³; hence it follows that at such temperatures liquid He³ possesses the ability to easily transmit the additional heat absorbed by the copper components and envelope of the sphere to the CMN crystals.

It is clear from Fig. 5a that there is a systematic discrepancy between the results of the measurements with heating by gamma sources and heating by current; therefore, on Fig. 5b the same measurements are plotted under several other assumptions. It was assumed that all of the heat generated by the heater remains in the sphere so that the gamma sources release 145 ergs in the sphere filled with He³ in 120 sec, and they release 120 ergs in the sphere containing 0.058 cm³ of He³. In this case, the values for the heat capacity of the sphere in the region from 0.006 to 0.011° deduced from measurements with heating by a current and by gamma sources agree. The points down to 0.015° are plotted from the calibration experiment. At lower temperatures a substantial deviation remains, and the point for measurements with gamma sources at T = 0.004° drops appreciably; however, the character of the specific heat curves becomes more similar to that expected theoretically for superfluid He³.

Analysis of the results obtained leads to the conclusion that a singularity in the temperature dependence of the specific heat of He^3 occurs in the neighborhood of 0.0055°, which one would expect for the transition of He^3 into a superfluid state. Since all of the previous experiments were carried out with a single sphere, and in spite of the fact that characteristic changes in the temperature dependence of the heat capacity are clearly associated with the presence of He^3 in the sphere, it was decided to prepare a second sphere in order to eliminate the possibility that random paramagnetic impurities, with Curie point in the vicinity of 0.0055°, were present.

MEASUREMENTS WITH THE NEW SPHERE

The new sphere also contained 14.1 g of CMN crystals, with average dimension 10μ , but the 640 copper 50 micron wires of total weight 0.6 g were soldered directly to the capillary and to the tin wires (of length 70 mm, diameter 0.18 mm) which served as a superconducting switch. A heater made out of 50 micron constantan wire of resistance 119 ohms (at liquid helium temperatures) was placed inside the sphere, in the immediate vicinity of the place of soldering of the copper and tin wires. Vacuum tightness of the sphere was ensured, as in the previous case, with the aid of polymerized sheath of epoxy resin mixed with powdered quartz, through which the leads from the heater also passed. The weight of this sheath was 2.6 g. A carbon film thermometer was consolidated directly on the sheath of the sphere. Irradiation by the gamma sources heated the thermometer to 0.04°, but when heated by a current, its resistance remained unchanged, that is, the warming up was less than 0.01 to 0.02°.

The volume of the pores in the sphere was 1.05 cm^3 . Cooling and measurement processes

were carried out in the same way as in the previous experiments. The difference associated with heating by current consisted in the fact that even small "tails," on the order of a few minutes, were not observed on the curves of deflection vs. time (as seen in Fig. 4, curve III).

The results of the experiments are presented on Fig. 6. For calculations of the specific heat, it was assumed that all of the heat generated by the current in the heater is transmitted to the sphere when irradiated by gamma sources, the sphere completely filled with He³ absorbs 1.14×10^{-7} W,



FIG. 6. Dependence of the heat capacity of the new sphere on temperature: 0 - 0.057 cm³ of liquid He³ in the sphere, p = 0, heating by two gamma sources with 120 sec period. $\Delta Q = 100$ erg; $\Box - 1.05$ cm³ of liquid He in the sphere, p = 60mm Hg, heating by two gamma sources at 80 sec periods, $\Delta Q = 91$ erg; $\Delta = 1.05$ cm³ of liquid He³ in the sphere, p = 14 mm Hg, heating by current at 80 sec periods, $\Delta Q = 104$ erg; $\nabla - 1.02$ cm³ of liquid He³ in the sphere, p = 0, heating by two gamma sources at 80 sec periods, $\Delta Q = 91$ erg; $\bullet - 0.062$ cm³ of liquid He^3 in the sphere, p = 0, heating by two gamma sources at 80 sec periods, $\Delta Q = 67$ erg. Curve 1 – the difference between the upper and lower curves, corresponding to the heat capacity of 1 cm3 of liquid He3; curve 2 - the same difference in the case when the values of the lower curve are decreased by 10%; curve 3 - the same difference in the case when the values of the lower curve are decreaed by 20%: curve 4 – the value for the heat capacity of 1 cm^3 of He³ according to the measurements of Anderson et al.^[15] The dashed curve is an extrapolation of curve 4 based on the calculations by Soda and Vasudevan^[16] with $T_{\lambda} = 0.0055^{\circ}$. The dot-dashed curve corresponds to the heat capacity of the filled sphere in the case when the specific heat of He³ varies according to a linear law for $T < 0.015^{\circ}$.

and the sphere containing CMN crystals covered only with a liquid helium film absorbs 8.3×10^{-8} W.

At the bottom of Fig. 6 are marked with corresponding symbols the difference between the heat capacity obtained in an experiment with the completely filled sphere, and its values taken from a curve drawn through the points obtained from measurements with the sphere containing a small amount of He³-that is, values of the heat capacity for 1 cm³ of liquid He³. These results correspond to curve 1. If we consider that the sphere containing CMN crystals covered only by a liquid He³ film absorbs during irradiation by the gamma sources not 8.3×10^{-8} W, but 10% less, then curve 2 is obtained from curve 1, but if the amount absorbed is 20% less, then curve 3 is obtained. We see that such recalculations very strongly influence the value obtained for the specific heat of He^3 at T ~ 0.004°, but have practically no effect in the determination of C for $T > 0.01^{\circ}$.

On Fig. 6 the dot-dashed curve indicates the heat capacity of the sphere filled with He³ for a specific heat of He^3 in the form C = 24 T, that is, for a linear temperature dependence. It is clear that the experimental data can in no way be arranged to fit such a curve. Since the present method does not enable us to confidently determine the absolute value of the specific heat of He³ in the temperature range near 0.005°, we can only assert, comparing curves 1 through 4 of Fig. 6, that the behavior of the temperature dependence of the specific heat of He^3 in the region between 5 and 6 thousandths of a degree undergoes a substantial change. But, since in order to guarantee the required accuracy of the measurements, it was necessary during one stage to heat the sphere by several thousandths of a degree and complete uniformity of the temperature throughout the inside of the sphere could not be guaranteed, all sharp variations in the behavior of the specific heat must be strongly smeared out, and in these cases the properties of He³ described by curve 4 of Fig. 6 and its dashed extrapolation must give results similar to curve 1.

DISCUSSION OF THE RESULTS OBTAINED

The determination of the specific heat was carried out in the present experiments on the basis of measuring the difference ΔT in the temperature of the sphere after releasing a definite amount of heat ΔQ in it: $C = \Delta Q/\Delta T$. Unfortunately, neither of these quantities was measured directly.

Instead of the thermodynamic temperature, the magnetic temperature of a sphere containing small

CMN crystals was used in the experiments. According to the measurements of Daniels and Robinson, ^[14] a single-crystal CMN ellipsoid gives a magnetic temperature differing from the thermodynamic temperature at 0.005° by 10%, but at lower temperatures the difference is even greater. Conditions for a sphere containing randomly distributed, strongly anisotropic crystals are no better; however, one might think that the weak maximum value of the magnetic moment observed in this case corresponds to the correct referral of the magnetic temperature to 0.0032° K. It is clear that for $T < 0.006^{\circ}$ the specific heat, determined from the magnetic temperature, will differ from the thermodynamic value; however, a phase transition in He³ must give in this case as well a clear anomaly in the temperature dependence of the specific heat. According to the data of Daniels and Robinson, ^[14] the entropy and consequently also, in first approximation, the specific heat of CMN is inversely proportional to the square of the magnetic temperature, that is, $C_1 = A/T^2$. Since the magnetic moment of the sphere $M = M_0/T$ = $B\varphi$, then for a constant heat flux $C_1T = W = const$ we obtain $\dot{\varphi} = -M_0W/AB = -K$ and $\varphi = \varphi_0 - Kt$. Thus, the deflections of the ballistic galvanometer should decrease linearly with the time, which is actually observed experimentally (see Fig. 4), and enables us to extrapolate the curves of deflection vs. time with great accuracy. The error in the determination of magnetic temperature amounted to a fraction of a per cent, but ΔT was determined to within about 2 or 3%.

The problem of uniformity of the temperature throughout the volume of the sphere is more complicated. In the sphere completely filled with He³ the liquid occupies $\frac{1}{8}$ of the volume; therefore, one can assume that the effective thermal conductivity in the sphere is given by

$$\kappa_{\rm eff} = 1/8 \kappa_{\rm He^3} \approx 6 \cdot 10^{-7} T^{-1} \, {\rm W/cm-deg}.$$

At T = 0.005° K, the heat capacity per unit volume of the sphere is, according to measurements, given by C \approx 0.003 J/cm³-deg; therefore the time required for the establishment of equilibrium is

$$\tau = CD^2 / \pi^2 \varkappa_{eff} = 14 \text{ sec}$$

(D = 2.4 cm is the diameter of the sphere), that is, equilibrium should manage to be established. However, in addition to the thermal resistance because of the finite thermal conductivity of He³, there is also a temperature discontinuity at the boundary between liquid helium and solids. If we assume that the discontinuity between CMN and He³ is the same as between He³ and copper in isolation, ^[17] that is, $R = 70 \text{ T}^{-3} \text{ deg-cm}^2/\text{W}$, and the specific heat of CMN is $C = 1.35 \times 10^{-7} \text{ T}^{-2} \text{ J/deg-cm}^3$, then for a crystal with volume V and surface area S

$$\mathbf{r}_1 = VCR \,/\, S = 10^{-5} V \,/\, ST^5.$$

For $T = 0.005^{\circ}$ and a cube of CMN with edge length 10^{-3} cm, the time $\tau_1 = 8$ min, that is, it will take a long time for equilibrium to be established. But the process of establishment of thermal equilibrium between the CMN crystals will not be noticed during measurements of the magnetic temperature. In fact, for all of the crystals one can write $\Sigma C_i \dot{T}_i = 0$, where C_i $= A_i / T_i^2$, that is,

$$\sum A_i \frac{T_i}{T_i^2} = \frac{d}{dt} \sum -\frac{A_i}{T_i} = 0,$$

or $\Sigma A_i / T_i = \text{const, but}$

$$\sum A_i/T_i = \sum aM_i = aM.$$

Thus, in the process of establishment of equilibrium, M = const and consequently the magnetic temperature is also constant. In this connection, liquid He³ will take on the average temperature after a time interval τ_2 which will be up to seven times smaller because of the fact that the volume of helium is up to seven times smaller than the volume of CMN, and up to $C(He^3)/C(CMN)$ times smaller because its specific heat is smaller. Therefore, at $T = 0.005^{\circ}$ the value of $\tau_2 = \tau_1/70 = 7$ sec. In this connection, τ_2 increases with decreasing temperature more slowly than τ_1 : $\tau_2 \sim 1/T^2$ [if C(He³) ~ T], that is, at 0.003° one would expect the value $\tau_2 = 20$ sec. As is evident from Fig. 4, no "tails" of any kind associated with the process of establishment of equilibrium were actually observed, and the jumps of the magnetic temperature are very clear.

At the same time, it is impossible to expect establishment of thermal equilibrium between all of the CMN crystals. Nonequilibrium must manifest itself in a smearing of the curve for the specific heat of He^3 as a function of temperature, measured by the method explained, and this forces us to strive as far as possible for uniform heating throughout the entire volume of the sphere, that is, to use two gamma sources. A comparison of the curves on Fig. 4 and Figs. 5 and 6 confirms the arguments cited: The anomaly in the behavior of the specific heat curve for He^3 is more sharply expressed during irradiation by two gamma sources than during irradiation by a single source. For CMN crystals of larger size, the ratio of surface area to volume would be even less favorable, and the anomaly in the behavior of the specific heat as a function of temperature would be practically unobservable.

An anomaly in the behavior of the specific heat curve was also observed^[18] during the separation of a mixture of He³ with He⁴ into two phases. At temperatures around 5 to 6 thousandths of a degree, a very small amount of He⁴ is sufficient for this phase separation to take place; however, the thermal effect associated with this separation is insignificant. The He³ used in the present experiments was subjected to special purification from He⁴ in a distillation column; according to estimates, there was less than 0.001% He⁴ present in it. The heat of mixing in the phase separation region was measured by Ouboter et al.^[18] down to $T = 0.5^{\circ}$ K; it rapidly decreases with decreasing temperature. If we assume, for an estimate, that the heat of mixing is proportional to T^2 , that is, $H = 0.6 (T/0.5)^2 J/mole$, and assume that there was 0.01% He⁴ in the He³, and that it was completely separated at 0.005° K, then the thermal effect in 1 cm³ of liquid helium altogether amounts to $\Delta Q = 2 \times 10^{-3}$ erg. A more accurate thermodynamic calculation gives a value which is more than three orders of magnitude smaller; therefore, the presence of He⁴ impurities in liquid He³ at temperatures on the order of thousandths of a degree cannot lead to a measureable anomaly in the specific heat curve.

The question about the amount of heat absorbed by the sphere during its irradiation by gamma sources is also complicated in the present measurements. If we compare the heat absorbed by the sphere, completely filled with He³, with that absorbed by the sphere containing CMN crystals covered only by a He³ film, then one can explain the difference in the heat absorption by the fact that the heat absorbed by the envelope and by the copper components is, in one case transmitted through the liquid He³ to the CMN crystals, but in the other case it is not. However, in the case of the empty and filled spheres, in order to obtain a reasonable value for the heat capacity, it is necessary to assume that a double difference in the heat absorption is obtained, which is impossible. The explanation lies in the fact that a nonequilibrium distribution of temperatures is observed in the empty sphere. Whereas well cooled crystals were demagnetized down to very low temperatures, other crystals were demagnetized only down to 0.0032°. Upon heating, the magnetic moment of the first crystals does not change for a long time, until $T = 0.0032^\circ$, whereas the second class of

crystals rapidly reaches $T = 0.0032^{\circ}$ and their moment begins to change. This is recorded as an increase in the temperature of the entire sphere, although the heat absorbed only by that portion of the crystals whose moment changes is registered. which leads to an apparent increase of the specific heat. The same effect may occur in the sphere partially filled with He³. In connection with this, and also since a magnetic and not a thermodynamic scale of temperatures was used, it is impossible to talk about a measurement of the specific heat of He³ in the thermodynamic sense. However, the so determined specific heat should not differ markedly from the thermodynamic; in addition, for the sphere completely filled with He³, the temperature dependence of the specific heat in the region from 0.005 to 0.006° has a clearly visible irregularity. There is no such irregularity for the empty sphere or for the sphere filled with He³ film. Therefore, one must recognize that the anomaly in the behavior of the heat capacity of the sphere is associated with a characteristic change in the specific heat of He³, which must occur if liquid He³ goes over into a superfluid phase.

The observed temperature of the transition, T ~ 0.0055°, lies within the limits of the region from 0.008 to 0.002° K, obtained theoretically by Gor'kov and Pitaevskiĭ^[19] on the basis of known physical properties of He³, and the results of the experiment are in agreement with the basic theoretical ideas of a number of authors.^[16,20-24]

It should be noted that observation of superfluidity in He³ by an abrupt increase of the heat transfer, which is easy to observe in He^4 at the lambda-point, is difficult to observe in He³ because, when the temperature decreases the viscosity increases like $\eta = 10^{-6} \text{ T}^{-2}$ poise, but the thermal conductivity increases like $K = 5 \times 10^{-6}$ T^{-1} W/cm-deg and, according to an estimate, at $T = 0.005^{\circ}$ the convective heat transfer in He³ will exceed the thermal conductivity for capillaries with diameters greater than 0.3 mm. It will also be difficult to observe superfluidity by the "fountain-effect," since at $T = 0.005^{\circ}$ and for a difference of levels h = 0.3 cm, a temperature difference $\Delta T = 0.001^\circ$ would arise because of the thermo-mechanical effect.

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