HEAT EXCHANGE BETWEEN A SOLID BODY AND LIQUID HELIUM BELOW 1°K

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The Kapitza thermal boundary resistance at the liquid-solid body interface was measured in He^3–He^4 solutions with molar concentrations of 1 and 5.5% and also in pure He^4 and He^3. The experiments were performed with four copper and stainless steel samples with either mechanically polished or rough surfaces. The temperature was varied between 1.3 and 0.1°K and the heat flux density between 17 and 1 erg/sec-cm^2 in He^3 and the solutions and between 200 and 1 erg/sec-cm^2 in He^4. In He^4 the $R \infty T^{-3}$ law is found to hold for the wall resistance in practically the whole temperature range investigated (0.1–1.3°K). The cubic temperature dependence applies to He^3 and the solutions only at temperatures below 0.3–0.4°K. The magnitude of the Kapitza resistance in this case depends weakly on the nature of the material and the state of the surface.

1. INTRODUCTION

$\mathbf{W}$HEN heat is transferred from one medium to another at low temperatures, a temperature difference is produced on the interface between any two media. This phenomenon was first observed in He II by Kapitza[1] and has been named in the literature the "thermal boundary resistance" or the "Kapitza jump."

It was shown in Kapitza's experiments that the thermal boundary resistance $R = \Delta T/Q$ is proportional to T^{-3}, and the jump ΔT itself occurs on the interface between the media (Q is the density of the heat flux through the interface).

A theory of this phenomenon was subsequently developed by Khalatnikov[2], Little[3], and Bekarevich and Khalatnikov[4]. For a copper-liquid He^4 interface (below T_λ) the theory gives

$$R \approx 500/T^3 \text{ [deg-cm}^{-2}/\text{W]}. \tag{1}$$

An analogous thermal resistance also occurs on the interface between a solid and liquid He^3 at not too low temperatures. The question of the temperature jump in He^3–He^4 solutions was not considered theoretically as such. It can be assumed, however, that the same theory can be applied to solutions at temperatures on the order of 1°K as to the pure isotopes.

In spite of the existence of a theory of heat exchange between two media at low temperatures, the experimental situation with the Kapitza jump is quite confusing. Most investigations in He^4 were performed at relatively high temperatures (1.3–2.1°K). In this temperature region, as a rule, the thermal boundary resistance depends little on the nature of the material and strongly on the state of the surface, namely its purity, stress, character of finish, etc. In numerous investigations performed by various authors, R varied approximately like AT^{-n}, but different values of A and n were obtained. We are unable to dwell here on the results of most investigations and refer the reader to the reviews[5,6].

It should be noted, however, that at high temperatures (T > 1°K), where the magnitude of the jump is relatively small, the temperature gradients in the solid are exceedingly important. If the purity of the metal and the finish of its surface are insufficient, an additional contribution is made to the thermal resistance of the boundary. In addition, at T > 1°K the wavelength of the phonons is negligible (~10^{-7} cm in helium and ~10^{-6} cm in the metal), and therefore the reflection of the phonons is quite sensitive to the state of the surface. Under these conditions the effective surface can be larger than the geometric one, and R is accordingly smaller. All this can apparently explain the large scatter in the experimental data in He^4.

As to the measurements in He^3, in the temperature region T > 0.5°K they are not sufficiently reliable, since the liquid has poor thermal conductivity and its resistance in a thin layer (100–200 μ) is much larger than or comparable with the Kapitza resistance. The corrections that are usually introduced for the temperature drop in the liquid lower the measurement accuracy. Analogous difficulties also arise, although to a lesser degree, in solutions because of their finite thermal conductivity.

It follows therefore that to compare theory with experiment, the temperature region most favorable for the investigation of the Kapitza resistance, both in He^4 and in He^3, is that considerably lower than 1°K. Such a possibility appeared with development of a new method of producing infralow temperatures by dissolving He^3 in He^4.

We present below the results of measurement of R on copper and stainless steel in contact with liquid He^4, He^3, and two weak solutions in the temperature interval from 1.3 to 0.05–0.1°K. Besides their theoretical significance, these measurements are of practical interest from the point of view of choosing the material and the construction of heat exchangers in dissolution cryostats.

2. EXPERIMENTAL TECHNIQUE

The measurements were performed in a dissolution cryostat[7] that produces a limiting temperature 2.5×10^{-2}°K in the circulation regime and 1×10^{-2}°K in the one-shot regime. Figure 1 shows the dissolution bath 1 with volume 30 cm^3, made of stainless steel, inside of which were placed a chamber with the investi-
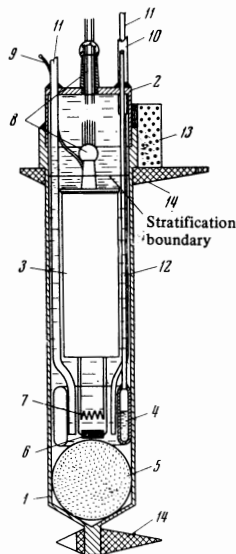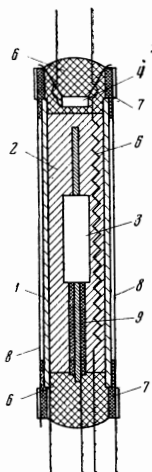
FIG. 1   FIG. 2

FIG. 1. Dissolution bath: 1—thin-walled cylinder of stainless steel, 2—copper cover, 3—measuring chamber, 4—He³ condensation thermometer, 5—CMN salt, 6—Speer carbon resistance thermometer, 7—heater, 8—platinum-glass seal for electric leads, 9—capillary for condensation of the investigated liquid, 10, 11—heat-exchanger tubes, 12—condensation-thermometer tube, 13—carbon adsorber, 14—clamps.

FIG. 2, Sample: 1—cylinder made of investigated material, 2—In, 3, 4—Speer carbon thermometers, 5—heater of insulated constantan wire, 6—epoxy resin with quartz, 7—teflon rings with h = 5 mm and diameters 5 and 4.6 mm, 8—insulated copper wires of 50μ diameter (100 pieces), 9—teflon insulation.

gated liquid and samples 3. The amount of He³ in the cryostat was chosen such that the stratification boundary would lie above the measuring chamber, so as to ensure uniform temperature over the height of the chamber.

The temperature of the liquid was determined from the magnetic susceptibility of the CMN salt 5 by the ballistic-galvanometer method. The salt was pressed from powder into a sphere of 10 mm radius. In the temperature region from 1.3 to $0.5°$K, the CMN salt was calibrated against the vapor tension of He³ of the condensation thermometer 4 and against standards of Al and Cd. A Speer carbon resistance thermometer 6 and a wire heater 7 were also placed in the liquid.

The measuring chamber was made of one piece of technically pure oxygen-free copper. Four holes of 30 mm length and 6 mm diameter were drilled in the copper to accommodate the investigated samples. The radial gap between the sample and the chamber was $d \sim 0.5$ mm. A copper cover with electric leads 8 and a capillary 9 for the condensation were hermetically soldered to the chamber.

We investigated four samples made of two different materials: technical oxygen-free copper (99.90) and stainless steel (1Kh18N9T). Two samples made of different materials had a smooth mechanically polished surface and two of them had a rough surface obtained by diffusion welding of the main sample to a thin layer ($\Delta d \approx 150-200$ μ) of sintered powder (particles with dimensions 50×70 μ). The sintering was in hydrogen
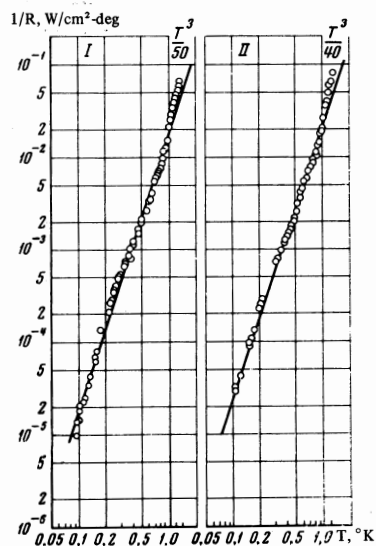
FIG. 3. Boundary conductivity in He⁴ for copper samples.

at an approximate temperature $1,000°$C for copper and $1300°$C for stainless steel, and lasted for several hours. The powder was deposited with the aid of organic adhesive, which volatilized during the sintering. The increase of the effective surface after the sintering was $\Delta S/S \approx 8$ at $S = 3.4$ cm².

All the samples were made in identical fashion (Fig. 2) in the form of hollow cylinders of outside diameter 5 mm and height 22 mm. The thickness of the cylinder walls was 0.5 mm for copper and 0.1 mm for stainless steel. For better contact between the thermometer and the sample and for more uniform radiation of the heat from a heater, the sample was filled with pure molten indium ($r/\Delta r = 2000$).

The measurements were made both in a field $H > H_{cr}$ and without a field. There was no noticeable difference in R. In the presence of a field (H ~ 300—500 Oe), the equilibrium was established somewhat more rapidly at low temperatures.

The carbon resistance thermometers 3 and 4 (Fig. 3) were calibrated against the susceptibility of the salt CMN with the power turned off. The thermometer 3 made it possible to measure the temperature inside the sample, and thermometer 4, one end of which was connected to the bundle of insulated copper wires stretched along the sample, made it possible to follow the temperature of the liquid near the boundary. The wires 8 (100 pieces of 50 μ diameter) were uniformly stretched along the surface of the sample at distances $l \leq 200$ μ.

The measurement method consisted of determining the heat rise of the sample ΔT when the power was turned on. The measurements were made alternately on all four samples. The temperature rise inside the sample after turning the power on was determined directly on the scale of the double potentiometer R-308 with automatic compensation. The potentiometer made it possible to follow simultaneously the readings of thermomenters 3 and 4. The power released in the samples was set in such a way that when the samples were connected to the circuit the temperature rise of the liquid near the boundary did not exceed 5% of ΔT.
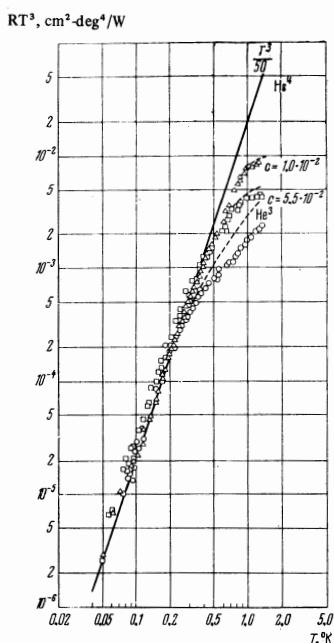
FIG. 4

FIG. 5

FIG. 4. Boundary conductivity of copper with smooth surface in He^3 and in solutions with molar concentrations $c_1 = 1.0 \times 10^{-2}$ and $c_2 = 5.5 \times 10^{-2}$. Straight line—$1/R$ for pure He^4.

FIG. 5. Boundary conductivity for sample of stainless steel with smooth surface in He^3 and in two solutions: $c_1 = 1.0 \times 10^{-2}$ and $c_2 = 5.5 \times 10^{-2}$. Straight line—$1/R$ in He^4.
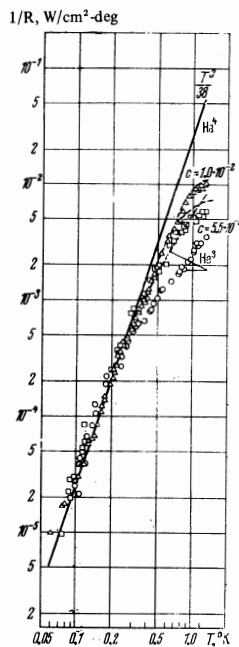
This is accomplished by keeping the power in the measuring chamber on during the entire time of measurement of R, and merely switching it from one sample to the other.

For He^3 and the solutions, the heat flux densities ranged from 17 to 1 erg/sec-cm^2 as the temperature changed from 1.3 to 0.1°K. The Kapitza jump varied in the range 10^{-3}–10^{-2}°K. In He^4, the maximum heat-flux density was 200 erg/sec-cm^2 at 1.3°K and decreased to 1 erg/sec-cm^2 at 0.1°K. The corresponding ΔT were 10^{-4}–10^{-2}°K.

During the course of the measurements, the temperature varied very slowly, so that the entire temperature interval was plotted during 4—5 days of helium experiments. The samples remained at helium temperature. However, the results showed good reproducibility even after heating on repeated cooling.

3. MEASUREMENT RESULTS

We present below the results of measurements of the temperature dependence of the wall conductivity $1/R = Q/\Delta T$ for four samples, designated as follows: I—copper with smooth surface, II—copper with surface of sintered powder; III—stainless steel with smooth surface, IV-stainless steel with surface of sintered powder. For all four samples, the surface area was assumed to be equal to the geometrical surface. No corrections for the temperature drop in the interior of the metal were introduced, since they are negligible in copper and indium in the entire temperature interval, and in thin-walled stainless steel they can become

comparable with the Kapitza jump only above 1°K, however, a comparison of the results obtained with copper and stainless steel in He^4 indicates that these corrections are small.

Figure 3 shows results of direct measurements of the boundary conductivity $1/R$ in copper as a function of the temperature in He^4 in the temperature interval 0.1—1.3°K. We see that below 1°K, within the limits of the measurement accuracy, the results fit well the straight line T^3/A. The rough surface on the copper decreases the thermal resistance only slightly (~20%), as against the expected decrease by a factor of 8. In measurements on stainless steel, analogous results were obtained: $1/R \infty T^3/38$ for sample III and $1/R \infty T^3/45$ for sample IV.

Measurements of $1/R$ were also made in solutions with molar concentrations $c_1 = 1.0 \times 10^{-2}$ and $c_2 = 5.5 \times 10^{-2}$, as well as in pure He^3. The results of measurements on samples I and III are shown in Figs. 4 and 5. The straight lines on each diagram correspond to $1/R$ in He^4 for each sample. The dashed curves on the figures take into account the corrections for the thermal conductivity of the solutions, which was taken by us from [8-10]. Analogous relations also hold for samples II and IV.

Measurements in superfluid solutions, as always, are connected with the danger of disturbing the concentration equilibrium as a result of blowing away of the He^3 by the heat flux. Calculation shows, however, that the heat-flux densities used by us (1—17 erg/sec-cm^2) are so small that the temperature gradients produced in the solution and the associated concentration gradients are negligible: $\Delta T = 5 \times 10^{-6}$–$10^{-4}$°K and $\Delta c = 10^{-9}$–10^{-7} for d = 0.5 mm.

As seen from the figures, allowance for the thermal conductivity of the liquid brings the points closer to the straight line T^3/A, but it does not eliminate completely the deviations from this line at high temperatures.

It should be noted that the corrections were introduced approximately, since the exact distribution of the temperature near the boundary is unknown. One cannot exclude the possibilities that a more accurate determination of ΔT on the boundary might give better agreement with the law $1/R \infty T^3$ also at higher temperatures.

Quite unexpectedly, the boundary thermal resistances of a solid in He^3, He^4, and weak solutions turn out to be approximately the same below 0.3°K.

The total error in the determination of $1/R$ was on the average 15—20%, although, as shown by the measurements, random errors in individual points sometimes increase to 40—50%. It should also be noted that the scatter of the points in He^3 is much larger than in pure He^4 in the entire temperature interval, this being most probably due to the long time required to establish equilibrium. The cause of the increase of random errors at very low temperatures may be an increase of the inertia of the carbon thermometers.

4. DISCUSSION OF RESULTS

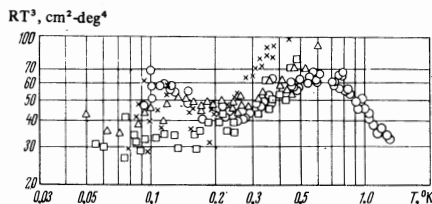In order to establish how well the law $R \infty T^{-3}$ is satisfied and the degree to which R is the same for

FIG. 6. The quantity $A = RT^3$ as a function of the temperature for sample I: $\bigcirc$—He^4, $\times$—He^3, $\triangle$—c = 1 mol. % He^3, $\square$—c = 5.5 mol.% He^3.

He^3 and He^4, it is convenient to plot the results in co-ordinates RT^3 against T. Figure 6 shows such a dependence for sample I in He^4, He^3, and in solutions, with allowance for the corrections for the thermal conductivity of the liquids. Similar relationships also hold for the other samples.

It is seen from the figure that in He^4 in the entire investigated interval, and in He^3 and in solutions below 0.5°K, the value of $A = RT^3$ does not remain strictly constant, but ranges from 30 to 70 with a mean value 40—50. In the region above 0.7—0.8°K, there is a noticeable tendency towards decreasing A in He^4. The reason for such a decrease remains unclear. In He^3 and in solutions above 0.3—0.5°K, a deviation from the cubic dependence is observed (possibly because of the low accuracy of the corrections for the thermal conductivity of the liquid). At low temperatures, for all the investigated liquids, the points are subject to wide scatter, but there is no apparent tendency towards differentiation of A as a function of the acoustic resistance of the liquid. We note that in the investigated interval the temperature changes approximately by a factor of 20, and the boundary resistance by a factor 10^4, and therefore the coefficient A can be regarded as constant with good accuracy.

In Figs. 7 and 8, our results are compared with the the published data for the Kapitza resistance at $T < 1°K$ on the boundary between copper and liquid helium. In He^4 (Fig. 7), according to the data of Kuang Wei-Yen[11], $R_4 = 21/T^{2.6}$ (0.57—2.2°K); according to the data of Fairbank and Wilks[12], $R_4 = 45/T^2$ (0.3—0.9°K), and according to the data of Anderson et al.[13], $R_4 \approx 12/T^4$ (0.075—0.85°K). As seen from Fig. 7, our experimental data for He^4 are closest to the T^{-3} dependence, but, like all other results, they lie below the Khalatnikov theoretical curve[2].

For the boundary between copper and liquid He^3 (Fig. 8), Lee and Fairbank[14] obtained $R_3 = 130/T^2$ (0.24—1.5°K), and Anderson and co-authors[15] obtained for electrically polished copper $R_3 = 200/T^3$ (0.05—0.1°K) and a stronger temperature dependence ($R_3 \infty T^{-4}$) in the interval $0.1° < T < 0.8°K$. Our results for He^3 below 0.3°K are in better agreement with the data of Anderson et al.[10] for insulated copper wires: $R_3 = 70/T^3$ (0.02—0.04°K) than for data for an electrically polished surface[15]. It should be noted, incidentally, that the resistance of the He^3-"epibond"[16] boundary, $R_3 = 35/T^3$, likewise differs little from our results for copper.

For the boundary between copper and a saturated He^3—He^4 solution there are only the data of Wheatley and co-authors[17]: R_{3-4} (60—70)/T^3, with which our results are in full agreement.

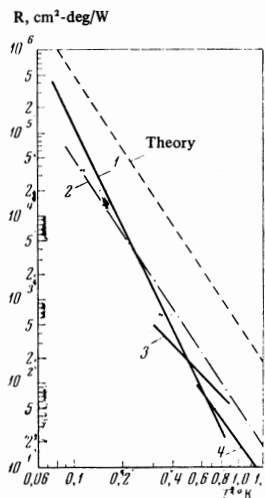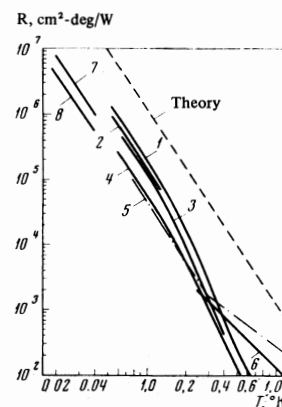
FIG. 7   FIG. 8

FIG. 7. Boundary resistance R for the copper-liquid He^4 interface in accordance with data by various authors: 1—Anderson, Connolly, Wheatley [13], electrically polished surface, $R \infty 12/T^4$; 2—present paper, sample I, $R \infty 50/T^3$; 3—Fairbank, Wilks [12], $R \infty 45/T^2$; 4—Kuang Wei-yan [11], $R \infty 21/T^{2.6}$. Dashed—Khalatnikov's theory [2], $R \infty 500/T^3$.

FIG. 8. Boundary resistance R for copper-liquid He^3 interface in accordance with data by various authors: 1—Anderson, Connolly, Vilches, Wheatley [15], electrically polished surface, p = 0.07 atm, $R \infty 200/T^3$ (0.05—0.1°K); 2—after [15], p = 6.5 atm, 3—after [15], p = 26.9 atm; 4—after [15], copper—solid He^3; 5—present work, sample I, $R \infty 50/T^3$ (0.08—0.3°K); 6—Lee and Fairbank [17], $R \infty 130/T^2$; 7—Anderson, Salinger, Steyert, Wheatley [16], copper—"epibond," $R \infty 35/T^3$. Dashed—Khalatnikov's theory [2], $R \infty 1200/T^3$.

The theoretical values of R lie much above the experimental ones both in He^4 and in He^3. Many hypotheses have been advanced concerning the causes of the strong discrepancy between experiment and theory, but so far none can be regarded as reliable. In particular, the idea was advanced that the boundary "is made transmitting" as a result of possible formation of a monolayer of solid helium on the surface[18], or as a result of contamination of the surface of the solid by sorbed gases[19], or else as a result of diffusion of the helium atoms into the solid[20]. The existence of a transmitting layer with acoustic resistance in the interval between the acoustic resistances of the solid and the liquid leads to an increase of the energy transfer from one medium to another. At the same time, ideas were also advanced concerning the possible existence of energy-transmission mechanisms other than phonons, for example with the aid of normal electrons in metals[21,22]. Calculations and experiments show, however, that although all the indicated mechanisms do improve the agreement with the theory, nonetheless they cannot fully explain such a large deviation from it.

At the same time, the question arises as to the causes of the disparity between individual experimental results.

The numerous data available for the region 1—2°K do not provide definitive answers for the foregoing questions and are in part contradictory. Thus, for example, most measurements at temperatures 1--2°K indicate that heat exchange increases on pure surfaces

and pure substances, including single crystals, as compared with heat exchange on contaminated surfaces[6]. However, a comparison of our data for mechanically polished samples with the data of Anderson et al.[13,15] on electrically polished samples at $T \sim 0.1°K$ gives, as it were, the opposite result. Another question also arises: Why do the coefficients of T^{-3} increase by almost one order of magnitude in the experiments at low temperatures?

It will obviously be possible to answer these questions only after a systematic investigation is made of the Kapitza jump at low temperatures. Many factors that are important at $T \sim 1°K$ become unimportant, namely the resistance of the sample itself, the small roughness, thin oxide films not exceeding the phonon wavelength. To compare the experimental data, it is very important to use samples with well known data on their purity, structure, and surface state. There is no doubt at present that the $R \propto T^{-3}$ law is satisfied at low temperatures. What remains unclear is the quantitative difference.

In conclusion, I am deeply grateful to P. L. Kapitza for interest in the work, to V. P. Peshkov for a discussion of the results and much valuable advice, and to V. N. Krutikhin for help with the experiments.

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