

Kapitza resistance at the solid helium-copper boundary under heavy thermal loads

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Results are presented of three sets of measurement of the heat conduction at the interface between solid helium and a copper cold conductor in the 0.45-1.5°K temperature range. The heat flux through the boundary can satisfactorily be described by the expression $Q = A(T_{\text{He}}^4 - T_{\text{Cu}}^4)$, where the parameter A depends weakly on the temperature, i.e. the temperature dependence of the Kapitza resistance $R_K = (1/4 AT^3)$ is close to the cubic dependence predicted by the acoustic mismatch theory. On the other hand the quantity R_K is smaller by one or two orders of magnitude than that predicted by the theory and, in contrast to the theory, does not depend on the density or sound velocity in helium. A comparison of the resistances of the boundaries between helium and a copper single crystal and a polycrystalline copper sample shows that the quality of the crystal structure of the copper sample also does not affect R_K . The resistance at the helium-copper boundary is primarily determined by the properties of the surface layer of the copper sample. This follows unambiguously from the fact that R_K is independent of the helium impedance, which increases 5 times on going from He II to solid helium at 185 atm, and is also independent of the degree of perfection of the bulk structure of the copper sample and helium crystals, and also from the considerable change (up to 5 times) of R_K on substitution of the cold finger. Plots of the heat conductivity of He⁴ and He³ crystals prepared from technically pure gases at pressures between 40 and 150 atm are presented.

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1. The onset of a temperature jump on the boundary between superfluid helium and copper when a constant heat flux is passed through the boundary was first observed by Kapitza^[1] about four decades ago. Kapitza has established the following: a) the temperature jump ΔT_j is localized in the helium at distances $\sim 10^{-4}$ cm from the solid surface; b) at small fluxes Q the value of ΔT_j is proportional to Q , i. e., it can be represented in the form

$$\Delta T_j = R_K Q. \quad (1)$$

where R_K is the thermal resistance on the separation boundary between two media (now called the Kapitza resistance); c) R_K increases with decreasing temperature approximately in proportion to T^{-3} . Even at the present time, however, the investigations of these phenomena are being continued in many laboratories (see, e.g., the proceedings of the latest international conference on phonon scattering^[2]) since, first, the Kapitza resistance is determined by the maximum possible heat loads and the rate of establishment of thermal equilibrium in the liquid helium + solid system at $T \lesssim 1$ °K, and second, the study of the conditions of heat transfer at the separation boundary promises to become a powerful tool for the study of the properties of the solid surface itself.

In the acoustic mismatch theory proposed by Khalatnikov,^[3] the large thermal resistance on the interface between two media, exceeding the bulk resistance of the solid sample, is due to the large difference between the acoustic impedances of the liquid helium $\rho_{\text{He}} C_{\text{He}}$ and the usual solid $\rho_S C_S$ (ρ is the density and C is the average speed of sound in the medium): $(\rho_{\text{He}} C_{\text{He}} / \rho_S C_S) \ll 0.1$, so that even in the case of normal incidence of an acoustic wave on the interface the reflection coefficient is close to unity. Accordingly, if the principal thermal excita-

tion in each of the media is due to the phonons, then at arbitrary angles of incidence on the surface, the phonons cannot go over into the liquid from the volume of the solid and vice versa, since the ratio of the wave vectors of the thermal phonons is $q_S / q_{\text{He}} = C_{\text{He}} / C_S \gtrsim 10$ and it is impossible to satisfy simultaneously the energy and momentum conservation laws. In the absence of phonon absorption on the surface (we are considering an ideal atomically pure solid-crystal surface free of defects and stresses) the heat transfer through the separation boundary is by radiation of the energy from the vibrating surface. If the temperatures of the media on both sides of the boundary are designated by T_{He} and T_S , then the heat flux density through the boundary is

$$Q = A(T_{\text{He}}^4 - T_S^4), \quad (2)$$

where A is a parameter that depends on the acoustic impedances of the media. For a flat surface we have

$$A = \frac{\rho_{\text{He}}}{\rho_S} C_{\text{He}} \frac{4\pi^2 k^2 F}{15(2\pi\hbar C_T)^3}, \quad (3)$$

where C_T is the speed of the transverse sound in the solid, k and \hbar are the Boltzmann and Planck constants, and F is a parameter that varies little when the elastic properties of the solid are altered ($F \approx 2.5$ and ≈ 2.0 for glass and platinum, respectively).

At low heat fluxes, i. e., when the temperature jump on the boundary is small, $\Delta T_j = (T_{\text{He}} - T_S) \ll T_S$, formula (2) can be represented in a form analogous to (1). It follows from (2) and (3) that the theoretically predicted value of the Kapitza resistance is

$$R_K^{\text{theor}} = 1/4AT^3 \approx 1.2 \cdot 10^{-11} \frac{\rho_S C_T^3}{\rho_{\text{He}} C_{\text{He}}} \cdot T^{-3} \text{ (cm}^2 \cdot \text{K/W)}. \quad (4)$$

For example, on the boundary between superfluid helium ($\rho_{\text{He}} = 0.14 \text{ g/cm}^3$, $C_{\text{He}} = 0.24 \cdot 10^5 \text{ cm/sec}$ and copper ($\rho_{\text{Cu}} = 8.9 \text{ g/cm}^3$, $C_{\text{Cu}} = 2.3 \cdot 10^5 \text{ cm/sec}$) one should expect $R_K^{\text{heor}} \approx 3.8 \cdot 10^2 \cdot T^{-3} \text{ (cm}^2 \cdot \text{K/W)}$.

The experimentally observed values of R_K are usually lower than the theoretical ones by one or two orders of magnitude. The temperature dependence of $R_K(T)$ can also differ noticeably from the theoretically predicted^[3] cubic dependence and change from sample to sample even in investigations of the behavior of the thermal resistance on the interface between superfluid helium and samples of the same material.^[4] A comparison of the accumulated experimental data with the modifications of the theory,^[3] which take into account in one manner or another the influence exerted on the thermal resistance R_K in liquid helium by the properties of real solid surfaces, can be found in the reviews^[5,6] and in the Proceedings of the Conference on Phonon Scattering.^[2]

2. In this paper we present the results of systematic measurements of the heat transfer at the interface boundary between solid helium-4 and helium-3 and a copper cold finger at temperatures 0.4–1.5 °K, at a heat-flux density in the range $(0.2\text{--}50) \times 10^{-4} \text{ W/cm}^2$.

The point is that in principle the acoustic mismatch theory is suitable for the description of heat transport through the interface between any media at sufficiently low temperatures, when one can operate with the concept of quantum excitations, so that formulas such as (1)–(4) are suitable for the estimates of R_K at the boundary between liquid helium-3 and a solid at temperatures $T \geq 0.2 \text{ °K}$, where the principal role is played by phonons,^[7] and in the transition to liquid helium. Allowance for the inequality $(\rho_{\text{He}} C_{\text{He}} / \rho_S C_S) \ll 0.1$ in the formulas of Little,^[8] who considered the resistance on the boundary between two solids as a result of acoustic mismatch, leads to expression (4) with accuracy to a numerical factor close to unity.

When comparing experiment with theory, solid helium is even more convenient than liquid, since the heat is transported in it only by phonons (there is no need to introduce corrections for the contributions of the rotons and impurity excitations^[3] in superfluid helium-4 and zero-sound quanta in helium-3^[7]). By increasing the pressure in the experimental cell it is possible, without changing in practice the properties of the surface of the cold finger, to change by several times the acoustic impedance of the helium (by five times when the impedances of superfluid He⁴ and solid He⁴ are compared at 185 atm, and by one order of magnitude in comparison with liquid He³). This is particularly interesting, because the results of many experiments have so far not been able to determine unambiguously how R_K depends on the impedance of the sample in contact with the helium, although in accordance with formulas (2) and (4) one should expect an appreciable change of R_K (proportional to the depth and to the cube of the Debye temperature of the sample).

3. Three series of experiments were performed. In the first series, the cold finger was a copper rod of 1.8 mm diameter, soldered to a depth of 7 mm into the low-

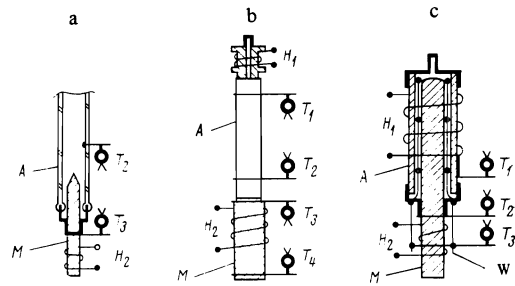


FIG. 1. Schematic diagrams of the cells used in the investigations: a) first, b) second, c) third series of experiments. C—supply capillary; $H_{1,2}$ —wire-wound heaters; A—ampoule (cell); M—coldfinger joining the cell with the He³ reservoir; $T_{1,2,3,4}$ —carbon thermometers. Teflon beads strung over the copper wires W prevented the wires from touching the cold finger or the cell in the third series of experiments.

er cover of a glass cell where the helium crystals were grown (Fig. 1a), while in the second series the cold finger was a rod of 4 mm diameter with a flat surface, soldered-in flush into the base of a thin-wall stainless-steel cube (Fig. 1b). After working on a lathe, this rod, in contrast to the first one, was first annealed at a temperature $\sim 850\text{--}900 \text{ °C}$, after which its surface was chemically polished. The soldering was with a low-melting point alloy (POS-60) in a helium atmosphere. In a third series of experiments, which were more qualitative in character and were intended to compare in a single experiment the behavior of R_K at the interface between helium and a perfect single-crystal surface and a polycrystalline surface with a large defect concentration, the cold finger was a copper crystal drawn from the melt by the Czochralski method in a vacuum of 10^{-3} Torr. Its average diameter was 2.7 mm, and the cell employed was a segment of ordinary copper tubing with inside diameter 5.0 mm and length 30 mm (Fig. 1c). Four copper wires of 0.4 mm diameter and 30 mm length were silver-soldered beforehand into the lower stainless-steel cover, and were subsequently soldered to a thermometer (T_3) for the measurement of the helium temperature in the space between the tube and the crystal. The lower cover and the copper crystal was also soldered in a helium atmosphere with low-melting-temperature solder.

The temperature distribution along the helium crystal and on the cold finger in the first two series of experiments and of the average temperatures of the cell, the helium, and the copper crystal in the third series, were monitored with carbon thermometers made of Allen-Bradley 10- Ω , 0.5-W resistors in accordance with the description of the preceding paper.^[9] The apparatus for measurements down to 0.4 °K and the procedure for growing helium crystals were similar to those used earlier.^[10]

Simultaneous measurements of the temperature at several points (Figs. 1a and 1b) has made it possible to calculate the thermal-conductivity coefficients of the helium crystal κ_{He} and of the cold finger κ_{Cu} , and then calculate the helium and copper temperatures T_{He} and T_{Cu} on both sides of the separation boundary, taking into ac-

count the experimentally obtained temperature dependences of $\kappa_{\text{He}}(T)$ and $\kappa_{\text{Cu}}(T)$. The microcalculations were performed with a "Nairi"-type computer. In the first phase we have neglected in the calculation of T_{He} and T_{Cu} the temperature gradient along the separation boundary, a procedure permissible only if ΔT_j is much larger than the gradient along the helium crystal, i. e., so long as $R_K > (l/\kappa_{\text{He}})$, where l is a characteristic dimension, say the length of the section of the cold finger situated inside the cell. This called for crystals with high thermal conductivity in the working temperature interval, so that we confine ourselves to perfect He^4 crystals grown at pressures $P > 60$ atm (the high thermal conductivity of such crystals could be assessed from the diagrams presented in the preceding paper^[10]). The use of a flat surface in the second series of experiments (Fig. 1b) made it possible to increase greatly the accuracy of the calculations of T_{He} and T_{Cu} on both sides of the separation boundary, and therefore in addition to the He^4 crystals we were able to investigate here also the resistance on the boundary between the copper and the He^3 crystals, the thermal conductivity of which was on the average much lower (we used commercially pure gas containing, according to the specifications, 0.2% He^4). It was assumed in the calculation that the magnitude and the temperature dependence of the thermal conductivity κ_{He} on the section from the separation boundary between the media and the upper thermometer were the same (i. e., the quality and orientation of the helium crystal were assumed to remain constant along the cell). The temperature gradients along the cell in measurements of κ_{He} should amount to ≥ 1 mK/cm, so that the maximum values of ΔT_j turned out to be approximately 0.1 °K—in fact, it was ΔT_j which determined the maximum permissible fluxes Q in the case of measurements near the thermal-conductivity maximum.

In the third series of experiments, the He^4 crystals

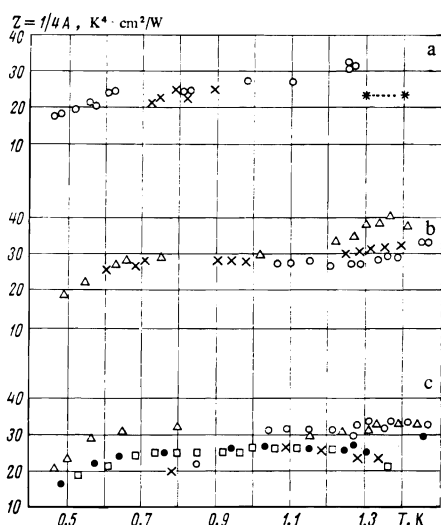


FIG. 2. Dependence of the parameter $Z \equiv (1/4A)$ on the temperature in the first series of experiments. a— $P=85$; b— $P=153$; c— $P=185$ atm. Different types of points correspond to different crystals; the asterisks correspond to measurements in superfluid helium.

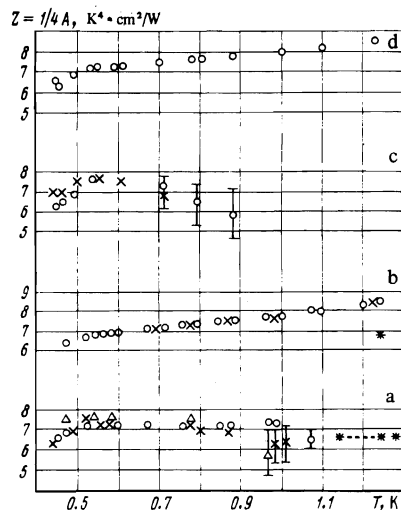


FIG. 3. Results of experiments in the second series: a— He^4 : Δ — $P=32$; \times —58; \circ —101 atm; b— He^4 : \circ , \times —152 atm; c— He^3 : \times —58; \circ —102 atm; d— He^3 —147 atm. The asterisks on a and b show the measurements of the heat transfer in the superfluid helium at 1.5 and 22 atm (*).

were grown with gradual decrease of the power delivered to a heater placed outside the copper tube (Fig. 1c). The temperature drops along and across the cell, across the solid-helium layer, and across the copper crystal (cold finger) were neglected in the calculation of ΔT_j (earlier measurements have shown that the smallest is the thermal conductivity of the helium crystal, but at $\kappa_{\text{He}} > 0.1$ W-cm/K the bulk resistance of the helium can be neglected in comparison with R_K), i. e., it was assumed that the thermometer readings correspond to the values of T_{He} and T_{Cu} at the corresponding boundaries.

4. Since the connection between ΔT_j and Q at large thermal loads was known to be nonlinear, it was natural to verify in the course of the reduction of the experimental data the suitability of formula (2) for the description of the measurement results. It turned out that in one experiment the values of the parameter $A = Q/(T_{\text{He}}^4 - T_{\text{Cu}}^4)$, pertaining to the same temperature $T = (T_{\text{He}} + T_{\text{Cu}})/2$, agree with each other on the average with accuracy better than 5% when Q is increased by a factor 2–4. Plots of $Z \equiv 1/4A = R_K T^3$ against the temperature are shown in Figs. 2, 3, and 5, while Fig. 4 shows the temperature dependence of the thermal conductivity of a number of He^4 and He^3 crystals grown without special precautions in a metallic cell in the second series of experiments. The asterisks on Figs. 2a and 3a show the results of measurements of the Kapitza resistance in superfluid helium at 1.2–1.4 °K, carried out periodically by way of control. The structure of all the He^4 crystals was hcp, and that of He^3 was bcc at $P < 110$ atm and hcp at higher pressures. Different points on the plots of Figs. 2 and 3 correspond to crystals grown at a given pressure in different days.

5. Let us discuss the results of the experiments.

a) As seen from the diagrams, the heat flux through the boundary between solid helium and copper, at large thermal loads and in the temperature interval 0.45–

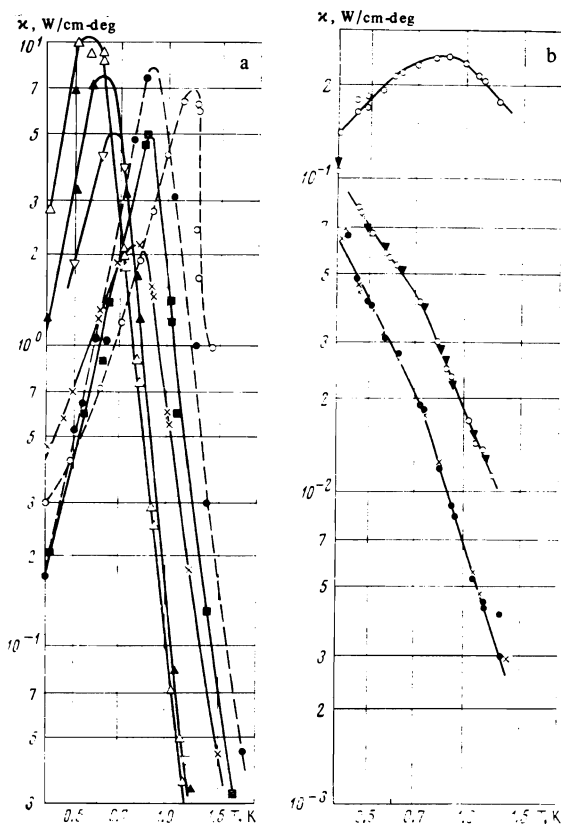


FIG. 4. Thermal conductivity of helium crystals drawn at different pressures, in the second series: a—He⁴: Δ , \blacktriangle , ∇ — $P=32$, \times , \blacksquare — $P=58$, \bullet — $P=101$, \circ — $P=152$ atm; b—He³+0.2% He⁴: \times , \bullet — $P=58$, \circ , \blacktriangledown — $P=102$, \circ — $P=147$ atm.

1.5 °K, is well described by expression (2), where T_{He} and T_{Cu} are the temperatures of the media on both sides of the separation boundary, calculated from the known values of $\nu_{He}(T)$ and $\nu_{Cu}(T)$.

b) The parameter A depends little on the temperature; on the average we can write $[A(T) - A(0.5)] \sim T^{-(0.3-0.1)}$, where $A(0.5)$ is the value of the parameter at $T=0.5$ °K. This means that, just as in the case of superfluid helium,^[11] the temperature dependence of the Kapitza resistance at an interface with a copper sample whose surface was worked in air by the standard methods is close to cubic: $R_K \sim (AT^3)^{-1} \sim T^{-(2.7-2.9)}$, as predicted by the acoustic mismatch theory.^[3]

c) Although the plot of $Z(T) = R_K T^3$ can shift along the ordinate axis by $\pm 10\%$ from experiment to experiment, the curves pertaining to the crystals He⁴ and He³ grown at different pressures, differ also in quality and in crystal structure within the limits of the indicated scatter, being practically superimposed one on the other in one series of experiments, and coincide with the resistance $R_K T^3$ in superfluid helium at 1.2–1.4 °K. The acoustic impedance of the He⁴ crystal at 185 atm pressure is about 5 times higher than the impedance of superfluid helium and ~ 3 times higher than the impedance of solid He³ at 60 atm. Thus, in contrast to the theory of^[3,8], the resistances at the boundaries of the investigated copper surfaces do not depend on the acoustic impedance of the helium.

d) The values of the parameter A , and accordingly R_K , can vary by several times when the cold finger is changed, apparently as a result of the change in the properties of the copper surface, but in all cases R_K is lower by at least one order of magnitude than the estimates given in the theory.^[3,8] One might attempt to ascribe the small value of R_K , in comparison with the theoretical value, to the influence of the large defect concentrations on the copper surface. We were unable, however, to observe substantial differences in the behavior of the Kapitza resistance on the boundary between helium and a copper single crystal drawn from the melt (free surface with small number of defects), and on the boundary with a copper tube (minute-crystallite structure, with chemical purity and quality of the crystal lattice known to be low) in a single experiment. Moreover, R_K for a polycrystal turned out to be higher than on the boundary of helium with a copper single crystal.

A comparison of our results with the data, e.g., of Zinov'eva,^[11] who measured the resistance at the boundary of copper samples and samples of stainless steel with liquid He⁴ and He³ in the interval 0.05–1.2 °K, show that both the values of R_K and the observed temperature dependences are close to each other: in our case $R_K \approx 30 \cdot T^{-2.8}$ in the first series, $\approx 7 \cdot T^{-2.8}$ cm² °K/W in the second, and $\approx (20-50)T^{-3}$ in the third series of experiments, while Zinov'eva obtained on the average $R_K \approx 50 T^{-3}$ cm² °K/W regardless of the surface quality. Replacement of He⁴ by He³ at $T \leq 0.7$ °K also did not influence the value of R_K in her experiments, although the impedances of the liquids differ by a factor of two.

6. It follows therefore from our experiments that, in contrast to the predictions of the theory (formulas (2) and (3)), a fivefold increase of the impedance of helium has practically no influence on the value of R_K , whereas the boundary Kapitza resistance can change by more than 5 times (from 7 to 50 cm² K/W at 1 °K) when the copper cold finger is replaced. The degree of perfection of the crystal structure of the helium crystals and of the copper cold finger likewise do not affect the value of R_K . As indicated above, although the measurements of R_K in liquid helium have been the subject of several dozen studies, it is likewise still impossible to determine unambiguously the dependence of the Kapitza resistance on

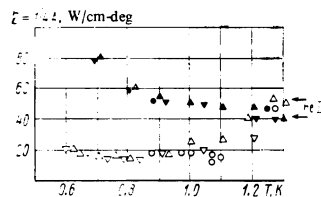


FIG. 5. Dependence of the parameter $Z = (1/4A)$ on the temperature in the third series of experiments: the dark points correspond to the boundary between the copper tube and helium, while the light points correspond to the boundary between helium and a copper crystal; $P=48$ atm (triangles), 102 atm (circles), and 152 atm (inverted triangles). The arrows indicate the results of the measurement at the boundaries with the superfluid helium ($P=1.5$ and 22 atm; the points coincide fully).

the impedance of the material in contact with the helium: the scatter of the values of R_K for one and the same substance is of the order of the predicted change of $R_K^{\text{theor}}(\rho_S C_S)$ for different substances. To explain the experimental results we can either assume that the decisive role is played by the properties of the surface rather than by the bulk impedance of the material, or else attempt to find additional more effective heat-transfer mechanisms at the separation boundary. Furthering the first assumption is the agreement, observed at large heat fluxes, between the theoretically predicted^[5] and experimentally observed laws governing the heat transfer at the interface, and the resultant close to cubic dependence of R_K on the temperature. In other theoretical models (see the review^[5] and the Proceedings of^[2]), the temperature dependence of R_K^{theor} differs substantially from cubic. The strong diffusion of the helium atoms at room temperature through oxide films that cover the surface of the actually investigated samples, prepared by ordinary methods and stored in air (for example, in alkali-halide crystals helium penetrates to a depth on the order of several microns; moreover, one observes penetration of the helium along the dislocations even in the case of crystals deformed in the liquid helium medium,^[13] as well as an increase of R_K when the surface is stored for a long time in a vacuum of $\sim 10^{-9}$ Torr^[14]), and the increase, compared with the average density of the helium, near the separation boundary as a result of attraction to the surface, can greatly decrease the differences between the impedances, namely, smear out the boundary over distances comparable with or exceeding the wavelengths of the thermal phonons in the investigated temperature interval ($\lambda_T \approx (40/T)$ Å in He II and increases in proportion to the speed of sound, i.e., $\approx (130/T)$ Å in solid He⁴ at 185 atm and $\approx (400/T)$ Å in copper). Allowance for the change of the impedances near the surface can lead not only to a decrease of R_K^{theor} , but can also influence the dependence on the density and on the speed of sound in helium. It would be interesting to solve the problem of the oscillations of the smeared-out solid surface in a medium with a given compressibility.

Saslow^[12] has noted that a comparison of the experiments at $T \leq 1$ °K in liquid helium and in theory may turn out to be incorrect because the transport mean free paths of the excitations in helium becomes comparable with the

characteristic dimensions of the experimental cells, and has proposed a separate model for the calculation of R_K^{theor} under these conditions. It follows from our measurements, however, that the correction necessitated by the change in the angular distribution of the phonons incident on the surface and the direct fly-through of the phonons in the case of large mean free paths is apparently small: the obtained values of R_K at $T = 0.5 - 0.7$ °K are close to one another for the different crystals. This is true even though the effective mean free paths of the phonons in helium, judging from the thermal conductivity (Fig. 4), could differ by three orders of magnitude (the maximum free path is approximately a millimeter).

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