Kapitza Resistance of Mercury between 1.1 and 2.1°K

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Between 1.1 and 2.1°K, measured values of the Kapitza resistance R of several Hg samples fall in the range $2.2 < RT^3 < 6.3$ deg^4 cm^2/W, where T is the absolute temperature. The temperature dependence of R can be accounted for by the Khalatnikov theory as modified to include the lattice specific heat of Hg. The observed magnitude of R is about one-fourth the predicted magnitude. R is different for the normal and superconducting states, showing that electronic processes do make a contribution to the thermal conductance of a Hg–He II interface.

INTRODUCTION

THE thermal resistance which occurs at an interface between a solid body and liquid helium is called the Kapitza resistance. Brief reviews of earlier work on this subject are given elsewhere.[1,2] Khalatnikov[3] showed that the heat exchange between a solid and liquid helium should take place mainly by radiation (or absorption) of phonons by the oscillating solid surface, and that the Kapitza resistance arises as a result of the acoustic mismatch of the solid to the liquid. The Khalatnikov theory predicts that the Kapitza resistance should vary as T^{-3}. Challis, Dransfeld, and Wilks[4] (CDW) considered the improved acoustic matching which should be provided by the compressed layers of helium near the surface; they predicted that the Kapitza resistance would be less than the Khalatnikov value and would vary as $T^{-4.2}$. Experimental values of the Kapitza resistance of many solids have been much smaller than predicted by Khalatnikov or CDW, and have displayed temperature dependences ranging from T^{-2} to $T^{-4.2}$. In fact, the only correlation between the available Kapitza resistance data and the acoustic transport theory is that the Kapitza resistance is generally larger for substances with higher Debye temperatures. However, the acoustic transport theory, when applied to a boundary between two solids,[5] agrees fairly well with the limited amount of available experimental data.[6,7] Thus, it seems that other mechanisms, in addition to acoustic transport, are responsible for heat transfer at a solid–liquid-helium boundary.

A calculation by Little[8] and a speculation attributed to Bloch[9] predicted that the conduction electrons of a metal would transfer energy to or from the phonons of the liquid helium. It was predicted that such processes would contribute to the boundary conductance amounts proportional to T^3 or T^5, and that such contributions would be greatly reduced in the superconducting state of a metal. To date, very little experimental evidence has been found in support of these theories. Little and Johnson[2] found that the Kapitza resistances of copper, tungsten, and gold exhibited none of the regularity predicted by the theories. In one instance,[10] the Kapitza resistance of tin was found unaffected by the superconducting transition; however, Gittleman and Bozowski[11] found the Kapitza resistance of tin to be increased by 10% in the superconducting state, and the Kapitza resistance of indium to be similarly increased by 5%. It is not entirely clear that these measurements were unaffected by the superconducting transition in the solder which mounted the specimens in the cryostat. Challis[1,12] found that the Kapitza resistance of the strong-coupling superconductor, lead,

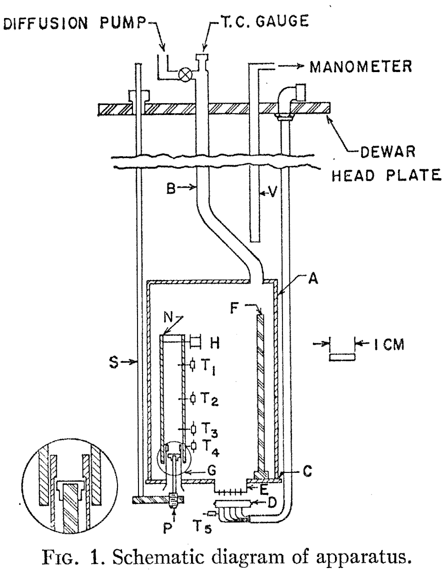

FIG. 1. Schematic diagram of apparatus.

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[1] L. J. Challis, Proc. Phys. Soc. (London) **80**, 759 (1962).
[2] R. C. Johnson and W. A. Little, Phys. Rev. **130**, 596 (1963).
[3] I. M. Khalatnikov, Zh. Eksperim. i Teor. Fiz. **22**, 687 (1952).
[4] L. J. Challis, K. Dransfeld, and J. Wilks, Proc. Roy. Soc. (London) **A260**, 31 (1961).
[5] W. A. Little, Can. J. Phys. **37**, 334 (1959).
[6] D. A. Neeper and J. R. Dillinger, Phys. Rev. **135**, A1028 (1964). The first author is grateful to A. H. Nethercot, Jr. for noting a numerical error in this reference. The theoretical value of the conductance of an indium-sapphire boundary should be in the range $4.3\times10^{-2}T^3$ to $4.9\times10^{-2}T^3$ W/cm^2 deg.
[7] R. J. von Gutfeld, A. H. Nethercot, Jr., and J. A. Armstrong, Phys. Rev. **142**, 436 (1966).
[8] W. A. Little, Phys. Rev. **123**, 435 (1961).
[9] See discussion in Ref. 2.
[10] Kuang Wey-yen, Zh. Eksperim. i Teor. Fiz. **42**, 921 (1962) [English transl.: Soviet Phys.—JETP **15**, 635 (1962)].
[11] J. I. Gittleman and S. Bozowski, Phys. Rev. **128**, 646 (1962).
[12] L. J. Challis and J. D. N. Cheeke, *Progress in Refrigeration Science and Technology* (Pergamon Press, Ltd., London, 1965), Vol. I, p. 227.

was greatly increased by the superconducting transition. The present paper describes an investigation of the Kapitza resistance of the other strong-coupling elemental superconductor, mercury.

EXPERIMENTAL TECHNIQUE

In these experiments, as in most others, the Kapitza resistance was determined by extrapolating the temperature gradient along a heated solid specimen to an end face exposed to the helium bath, and thereby inferring the temperature jump between the end face and the bath. This required that the mercury specimen be held at this end face by a superleak-tight seal of low thermal conductance. Other workers[13] mentioned that Hg would bond to an alloy of 90% Cu, 10% Ni. Trial proved this to be the best material for our purposes.

Experimental Apparatus

The experimental apparatus, which could be lowered into a stainless steel Dewar, is shown schematically in Fig. 1. The stainless steel cannister A, was supported from the Dewar head plate by the evacuation tube B. The base was attached to the cannister by the Wood's metal solder joint at C. Electrical leads were attached to two sockets represented by D, which plugged onto electrical feedthroughs represented by E. The wires leading from the pins of E were glued to the brass support post, F, and left F at suitable heights for attachment to the thermometers T_1-T_4, or the heater H. The mercury specimen was held by the 0.08-cm-wall nylon tube N. The heater and thermometers were thermally connected to the Hg by 0.076-cm platinum wires which tightly fit holes in N. The thermometers were nominally 47 Ω, $\frac{1}{10}$-W "Ohmite" resistors cemented into copper sleeves. The nylon tube N was forced onto the 0.013-cm wall cupronickel sleeve G. In various experiments, different cupronickel tubes of 0.64- or 0.94-cm diam were used at G. The length of cupronickel section exposed to the mercury varied from 0.076 to 0.23 cm. A brass plunger P was tipped with a Teflon cap which, when pressed against the lip in G, formed a seal impervious to liquid mercury. The plunger was threaded onto a rod S, which passed through an O-ring seal at the head plate. The 0.94-cm-diam open-ended vapor pressure tube V always extended below the liquid-helium level.

The dc heater and thermometer voltages were measured with L- and N-type K-3 potentiometers, with the current forward and reversed so as to compensate for small thermal emf's. The bath temperature was stabilized to better than 50 μdeg by an electronic regulator. Magnetic fields up to 3.5 kOe could be applied parallel or perpendicular to the axis of the mercury specimen by magnets outside the Dewar.

[13] R. T. Webber and D. A. Spohr, Phys. Rev. **106**, 927 (1957).

Heat Loss

Although only a small amount of heat would be shunted to the bath through the cupronickel mounting sleeve, this amount would be different for the superconducting and normal states of the mercury, since in the superconducting state an added thermal resistance would appear at the mercury-cupronickel interface.[14,15] Any shunted heat flux for mercury in the superconducting state was assumed to be negligible. To estimate the total shunted heat flux in the normal state, we numerically solved a boundary-value problem approximately representing the heat flow in the simple geometry shown in Fig. 2. The solid specimen was assumed to be isothermal, and any thermal resistance at the junction of the solid specimen to the sleeve was neglected. The results of the calculation, given in the dimensionless plot of Fig. 2, are valid for any situation in which the sleeve material has a conductivity much less than that of the specimen and the tube wall thickness is small compared to its diameter.

In separate experiments we found that between 1 and 2.1°K, the thermal conductivity λ of the cupronickel was approximately $(0.12+1.49T)$ mW/cm deg. The Kapitza resistance of the cupronickel was difficult to measure due to the high thermal resistivity of the material, but the data indicate a value of approximately $15T^{-3}$ deg cm^2/W. In the experiments with mercury, the estimated shunted heat flux amounted to 2.5% at 1.1°K and 0.7% at 2.1°K. The nylon tube carried a negligible heat flux.

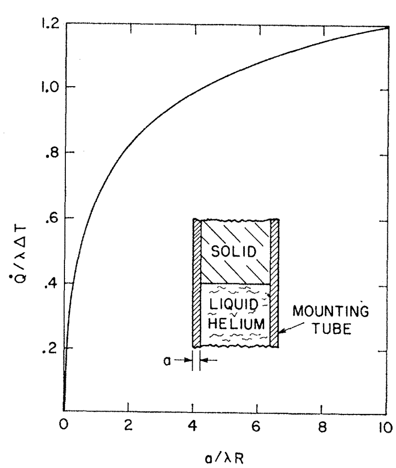

FIG. 2. Dimensionless plot of heat loss through the mounting tube as a function of tube properties. $\dot{Q}$ is the total heat current from the sleeve to the liquid helium per unit periphery of the sleeve, λ is the thermal conductivity of the tube material, ΔT is the temperature jump at the specimen-helium interface, a is the wall thickness of the tube, and R is the Kapitza resistance of the tube material.

[14] L. J. Barnes and J. R. Dillinger, Phys. Rev. **141**, 615 (1966).

[15] L. J. Challis and J. D. N. Cheeke, Phys. Letters **5**, 305 (1963).

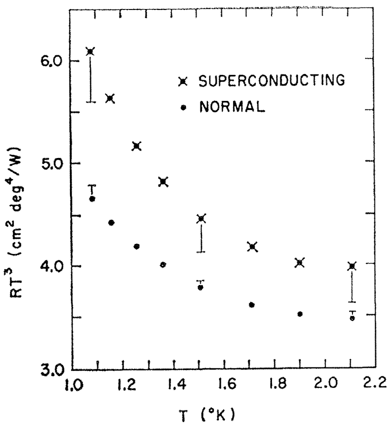

FIG. 3. The Kapitza resistance of sample 14 of mercury in the normal and superconducting states as a function of temperature. Each plotted point represents an unresolved pair of points obtained with different heat inputs. The ratio of the heat inputs for each measurement pair was about 2:1. For a discussion of error bars, see the text.

Experimental Procedures

In preparation for an experiment, the cupronickel sleeve which extended above the plunger was wet with mercury, using HNO_3 as a flux. The region was immediately flushed with NH_4OH, followed by distilled water. The contaminated mercury was removed and the shallow cup formed by the cupronickel tube above the plunger was flushed twice with pure mercury. The nylon tube was then pushed over the cupronickel sleeve and filled from the bottom with mercury. This was accomplished by using a hypodermic syringe, tipped with a long Teflon needle inserted down the axis of the nylon tube. Finally, the Teflon needle was withdrawn from the nylon tube, and the base was soldered into the bottom of the cannister with Wood's metal.

After the apparatus was lowered into the Dewar, the cannister was evacuated, and the pressure in the helium Dewar was reduced to 200 Torr. While the apparatus was being cooled by liquid nitrogen in the outer Dewar, the temperature of the mercury was monitored by the resistance of the heater H. After reaching the freezing temperature, the mercury solidified during a period of three minutes while 18 mW was being expended by the heater. The heating technique was employed to ensure that the sample would freeze from the bottom. After the mercury was frozen, the plunger was removed from the tube. Inspection of a solid specimen after an experiment indicated that solidification had indeed taken place from the bottom.

In all experiments, data were taken with the mercury first superconducting, then normal in a 500-Oe axial field, and finally in other axial or transverse fields. It was expected that ring solder joints in the cannister might prevent use of axial fields; however, a sharp, total transition in the mercury was observed at the appropriate value of the applied axial field.

Data were taken with the heater on and off in the manner described in Ref. 6. The thermometers were calibrated against the vapor pressure of helium at about 12 temperatures between 1.25 and 2.15°K. For each thermometer, a single least-squares fit of the calibration data to the three-constant formula[16] exhibited a maximum deviation of ± 0.5 mdeg.

EXPERIMENTAL RESULTS

In experiments such as these, a cold-worked layer near the surface of a machined or polished specimen may add a significant thermal resistance indistinguishable from the true Kapitza resistance. This may account for the lack of reproducibility in many previous measurements. In the present experiments it was hoped to avoid this problem by using the specimen as cast from the melt. Nonetheless, there was a problem in reliable extrapolation of the thermal gradient due to diffusion of copper from the cupronickel into the mercury near the end of the specimen.

The supplier[17] quoted the mercury as having a total impurity content of one part per million, and indeed the thermal conductivity displayed a higher peak at a lower temperature than that of previously reported specimens.[13] The thermal conductivity of the segments of Hg held by the nylon tube generally exhibited a maximum of 2.8 W/cm deg at 1.35°K in the superconducting state, and a maximum greater than 30 W/cm deg below 1.1°K in a 500-Oe axial field. It was noticed that the conductivity between thermometers T_3 and T_4 consistently indicated a higher impurity content than the upper section of the specimen, indicating that copper from the mounting sleeve was not rising to the top surface of the specimen during solidification. Subsequently, an experiment was conducted in which all of the specimen below T_2 was encased in a cupronickel sleeve (sample 12). The resulting conductivity between T_1 and T_2 was characteristic of previous data, but the section encased by the cupronickel displayed a conductivity maximum of only 1.3 W/cm deg at 2°K when superconducting, in agreement with the data of Webber and Spohr.[13] The values for the conductivity of contaminated mercury allowed an estimate of the upper limits of the systematic extrapolation error in the other data.

Systematic errors in the measured Kapitza resistance of 1 to 2% were estimated to result from uncertainties in thermometer placement and temperature calibration. In the normal state, the temperature drop in the sample below T_4 never exceeded 1% of the total temperature difference between T_4 and the helium bath. In the superconducting state, the temperature drop in the sample below T_4 was 5 to 25% of the total temperature drop between T_4 and the bath. Copper impurities in the sample below T_4 introduced an uncertainty into the temperature extrapolation for the superconducting state.

Measured values of RT^3, where R is the Kapitza resistance and T the temperature, are plotted for a

[16] G. K. White, *Experimental Techniques in Low Temperature Physics* (Oxford University Press, London, 1959), p. 127.

[17] Lytess Metal and Chemical Corporation, New York, New York.

typical sample as a function of temperature in Fig. 3. The error bars on the normal-state curve derive mainly from the heat shunted by the cupronickel tube. The error bars on the curve for the superconducting state arise from uncertainties in the extrapolation of the temperature below T_4. The data points shown were determined by assuming the thermal conductivity to be the same as that measured above T_4, and the lower limits of the error bars were determined by assuming the thermal conductivity to be the same as for the deliberately contaminated sample.

Variations from sample to sample of the normal-state Kapitza resistance were much larger than any estimated experimental uncertainty. These variations may be due to different crystallographic orientations of the samples. The superconducting-normal difference was always greater than the experimental uncertainty and therefore can be regarded as a real effect. For all samples the measured Kapitza resistance could be described by an empirical equation of the form $1/R=\alpha T^3-\beta T$, where α and β are constants for a given sample. Values of these constants for several samples are listed in Table I, although no physical interpretation can be given to the negative quantity. R did not vary as any single power of T.

The thermal conductivity of normal mercury became less sensitive to impurity content with increasing transverse magnetic field becoming independent of impurity content above 2 kOe. The variation with magnetic field was in rough agreement with the observations of Webber and Spohr.[13] It was thought that, by varying the field, the form of the temperature gradient could be made to coincide with the form of the gradient for the superconducting state. However, since the conductivity became independent of impurities at higher fields, this was impossible.

The change in R due to magnetic fields varied positively or negatively from sample to sample but was never greater in magnitude than 6%. The thermometers exhibited negligible magnetoresistance.

DISCUSSION

Figure 4 displays $\Delta K/T^3$ as a function of T, where $\Delta K=(1/R\text{ normal}-1/R\text{ super})$ is the superconducting-

TABLE I. Values of the constants α and β for the empirical expression $1/R=\alpha T^3-\beta T$ W/cm² deg. Samples 10 and 14 were prepared with an assembly technique which produced a minimum contamination of the mercury. Sample 0 was analyzed as received from the supplier and sample 10 was destroyed in the cryostat.

Sample No.	Normal α	Normal β	Superconducting α	Superconducting β	Copper impurity (ppm)
10	0.518	0.283	0.476	0.290	···
12	0.412	0.137	0.409	0.237	300
14	0.322	0.130	0.292	0.155	50
0	···	···	···	···	1

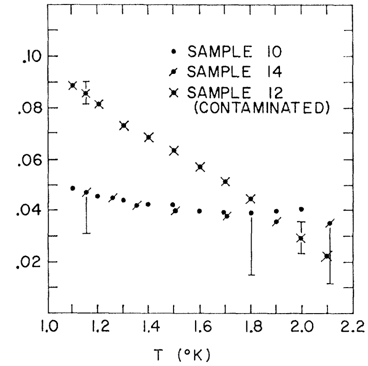

FIG. 4. $\Delta K/T^3$ as a function of temperature for the samples described in Table I. $\Delta K=(1/R\text{ normal}-1/R\text{ super})$ is the superconducting-normal difference in the boundary conductance. $\Delta K/T^3$ was computed from smoothed curves similar to that shown in Fig. 3. The error bars indicate uncertainty due to temperature extrapolation.

normal difference in the boundary conductance. Near 2.1°K, there is an appreciable density of normal electrons in the superconducting state, so the electronic contribution to the boundary conductance cannot simply be equated to ΔK. The T^3 dependence for the electronic boundary conductance predicted by Little[8] is in approximate agreement with the present results. It is to be noted that $\Delta K/T^3$ falls in the range of $2\text{–}8\times10^{-2}$ W/cm² deg⁴, which is about an order of magnitude larger than the rough numerical estimate by Little[8] for the electronic conductance and about the same magnitude as predicted by Andreev[18] for metals with long electronic mean free paths. In view of the substantial experimental uncertainties, further analysis of the curves of Fig. 3 was not attempted.

Khalatnikov[3] considered heat transport at a boundary between a solid and liquid helium to result from three processes: collision of helium phonons with the solid, collision of rotons with the solid and acoustic radiation by the vibrating solid surface. The first two processes are predicted to carry about 40% of the total heat current above 2°K and to become insignificant between 1 and 2°K.

To calculate the contribution of the third process, Khalatnikov assumed that a solid surface vibrating in contact with the low-density fluid is equivalent to a solid surface vibrating in free space. For this process alone the boundary conductance is given by

$$K_3=\frac{\rho_l c_l}{\rho_s}\pi^5\frac{16}{15}\frac{k^4T^3}{h^3c_t^3}F, \qquad (1)$$

in which ρ_l and c_l are the density and sound velocity in the liquid, ρ_s is the density of the solid, c_t is the transverse sound velocity in the solid, k is Boltzmann's constant, h is Planck's constant, and F is a constant of the solid presumed to be about 1.5.[19] Thus one would expect RT^3 to be independent of temperature near 1°K. Since the experimental values of RT^3 do not become

[18] A. F. Andreev, Zh. Eksperim. i Teor. Fiz. **43**, 1535 (1962) [English transl.: Soviet Phys.—JETP **16**, 1084 (1963)].

[19] L. J. Challis, in *Proceedings of The Seventh International Conference on Low Temperature Physics, 1960*, edited by G. M. Graham and A. C. Hollis (University of Toronto Press, Toronto, 1961), p. 467.

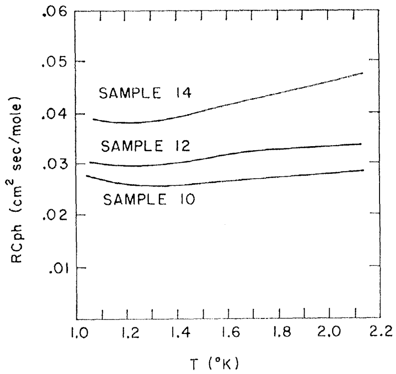

FIG. 5. RC_{ph} as a function of temperature for the mercury samples described in Table I. R is the Kapitza resistance for the superconducting state, and C_{ph} is the lattice specific heat.

temperature-independent, one may conclude that the predicted phonon- and roton-collision processes, if operative at all, do not cause the observed temperature dependence of RT^3. Table II lists the values of RT^3 predicted by Eq. (1) for a set of possible values of c_t calculated from the elastic constants of mercury,[20] and also lists some experimental values for the superconducting state. The published values[21] of the Debye Θ of mercury imply an average c_t of about 5×10^4 cm/sec. It can be seen from Table II that the experimental values of RT^3 fall near that corresponding to the lowest plausible c_t. In no case did we observe a Kapitza resistance greater than that of sample 14. Thus, although the sample-to-sample variations might be due to various crystallographic orientations of the surface, it can be concluded that the magnitude of R is about $\frac{1}{4}$ of the Khalatnikov value. Part of this discrepancy may be due to the fact that the microscopic surface area is generally greater than that implied from the sample diameter.

TABLE II. Comparison of experimental values for RT^3 in deg^4 cm^2/W with theoretical values for various c_t in cm/sec. Experimental values are given as the range observed for the superconducting state.

	$c_t=9\times10^4$	$c_t=5\times10^4$	$c_t=4\times10^4$	Sample 10	Sample 12	Sample 14
RT^3	58	17	5.1	2.4–4.4	2.7–4.7	4.0–6.2

[20] E. Gruneisen and H. Hoyer, Ann. Physik **22**, 663 (1935).
[21] B. J. C. van der Hoeven, Jr., and P. A. Keesom, Phys. Rev. **135**, A631 (1964).

The T^3 dependence of K_3 in Eq. (1) results from an implicit assumption of a T^3 lattice specific heat for the solid. In general, K_3 should be proportional to the lattice specific heat if one assumes that all phonon modes of the solid follow the same dispersion relation and if surface-wave contributions are relatively unimportant.[22] Figure 5 shows RC_{ph} as a function of T, where C_{ph} is the lattice specific heat of mercury obtained from Ref. 21. It can be seen that indeed most of the observed temperature dependence of R can be accounted for by the temperature dependence of the lattice specific heat, despite the stringent foregoing assumption.

CONCLUSIONS

The observed Kapitza resistance of mercury qualitatively supports the acoustic-radiation theory in the sense that (a) mercury has the lowest Kapitza resistance yet reported as would be expected from its low Debye Θ of about 60°K; and (b) the product of the Kapitza resistance and lattice specific heat is nearly independent of temperature. Experiments extending to below 0.7°K, where the lattice specific heat becomes cubic in temperature, would investigate condition (b) more completely. The Kapitza resistance of mercury is less than the Khalatnikov value, as is the case for other solids.

The decreased Kapitza resistance in the normal state shows that electronic processes do contribute to heat transfer at a helium-mercury surface, but such contributions amount to less than 30% of the total heat transfer at temperatures above 1°K.

ACKNOWLEDGMENTS

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[22] W. A. Little, in *Proceedings of The Seventh International Conference on Low Temperature Physics, 1960*, edited by G. M. Graham and A. C. Hollis (University of Toronto Press, Toronto, 1961), p. 482.