## Experiments on the Kapitza Resistance\*

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The Kapitza resistances of samples of copper, gold, tungsten, single-crystal lithium fluoride, and singlecrystal silicon have been measured in the temperature range from 1.25 to 2.10°K. We find that a clean copper surface prepared by machining under liquid helium exhibits a Kapitza resistance whose magnitude and temperature dependence are considerably greater than that which a dirty copper surface exhibits. The Kapitza resistances which these samples exhibited can be represented by a power law as follows: Au, 8 T<sup>-3</sup>; LiF, 23 T<sup>-3.75</sup>; Cu (clean), 19 T<sup>-3.6</sup>; Si (unetched, 7 dislocations/cm<sup>2</sup>), 21 T<sup>-3.2</sup>; Cu (dirty), 7 T<sup>-2.6</sup>; Si (etched, 7 dislocations/cm<sup>2</sup>), 35  $T^{-4.15}$ ; W, 39  $T^{-3.5}$ ; Si (etched, 800 dislocations/cm<sup>2</sup>), 35  $T^{-4.15}$  (units are deg W<sup>-1</sup> cm<sup>2</sup>). The interfacial surface of the lithium fluoride crystal was a (100) cleavage plane. The interfacial surfaces of the silicon crystals were (111) planes which were ground, polished, and in some cases, etched. We have considered these results in relation to the theories of Khalatnikov, Little, and a modification of the Khalatnikov theory by Challis, Dransfeld, and Wilks. These theories failed to predict both the absolute magnitude and temperature dependence of the Kapitza resistances and the relative magnitude of the resistances for different samples. In addition, we have found no evidence in our experiments on cleaned copper of the mechanism proposed by Bloch involving the conduction electrons.

N interesting thermal phenomenon was discovered A by Kapitza<sup>1</sup> in 1941. Accompanying the flow of heat across the interface between a heated body and liquid helium II, a discontinuous drop in temperature occurs at the interface. The thermal resistance which gives rise to this temperature jump is called the Kapitza resistance.

The first theories of the Kapitza resistance were those of Kronig and Thellung<sup>2</sup> and of Gorter, Taconis, and Beenakker.3 In both theories the temperature jump arose as a consequence of the superfluid properties of helium II. Later, however, Lee and Fairbank<sup>4</sup> observed the same phenomenon in the case of contact with liquid He<sup>3</sup>. We now believe that the superfluid mechanisms offer too little resistance in themselves to account for the entire phenomenon. In 1952 Khalatnikov<sup>5</sup> showed that for all temperatures below the  $\lambda$ -point heat exchange between a solid body and helium II should take place mainly by the emission or absorption of thermal vibrations by the surface of the solid. Hence, it was concluded that the Kapitza resistance arose from the severe acoustic mismatch of the solid to the liquid and was located at the interface itself. Experimental studies of the Khalatnikov theory have been reported by Dransfeld and Wilks<sup>6</sup> and by Challis, Dransfeld, and Wilks. The first of these papers reported that the Kapitza resistance was nearly independent of pressure up to 20 atm from about 1.3°K up to the lambda point, in contradiction to the Khalatnikov theory. They also found that the magnitudes of the resistances for copper, lead, and quartz were all of the same order, and approximately 1/50 to 1/20 of the predicted magnitudes. The second paper presented a modification of the Khalatnikov theory which leads to a dependence of the Kapitza resistance on the pressure which is qualitatively similar to the observed behavior. They considered the effect on the Kapitza resistance of the improvement in acoustic match afforded by the layer of helium at the interface which is compressed by the Van der Waals force of attraction to the solid body. In 1959, Little8 extended Khalatnikov's idea to the more general case of heat transfer across the interface between any two dissimilar bodies. His treatment arrived at the same result as Khalatnikov's. In addition, he suggested that the direct interaction of the conduction electrons of a metal with the surface disturbances of the totally reflected helium phonons might provide an important means for heat transfer. His prediction that there should be a considerable difference in the Kapitza resistance of a metal in the normal and superconducting states was confirmed by Challis9 who reported an increase in the magnitude and temperature dependence of the Kapitza resistance for lead in the superconducting state over its resistance in the normal state. In 1961, Little<sup>10</sup> presented a detailed calculation of the Kapitza resistance between helium II and metals. He obtained an expression for the heat flux which should arise from the interaction of the surface disturbances of the totally reflected helium phonons with the conduction electrons. The calculated magnitude

of this heat flux is about 300 times smaller than Challis

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<sup>1</sup> P. L. Kapitza, J. Phys. (U.S.S.R.) 4, 181 (1941).

<sup>2</sup> R. Kronig and A. Thellung, Physica 16, 678 (1950).

<sup>3</sup> C. J. Gorter, K. W. Taconis, and J. J. Beenakker, Physica 17, 86 (1951).

D. M. Lee and H. A. Fairbank, Phys. Rev. 116, 1359 (1959).

<sup>&</sup>lt;sup>5</sup> I. M. Khalatnikov, Zh. Eksperim. i. Teor. Fiz. 22, 687 (1952). <sup>6</sup> K. Dransfeld and J. Wilks, Low Temperature Physics and Chemistry (University of Wisconsin Press, Madison, Wisconsin,

<sup>1958),</sup> p. 39.

<sup>7</sup> L. J. Challis, K. Dransfeld, and J. Wilks, Proc. Roy. Soc. (London) **A260**, 31 (1961).

<sup>&</sup>lt;sup>8</sup> W. A. Little, Can. J. Phys. 37, 334 (1959).

<sup>&</sup>lt;sup>9</sup>L. J. Challis, in Seventh International Conference on Low-Temperature Physics, Toronto, 1960 (University of Toronto Press,

Toronto, 1961), p. 466.

10 W. A. Little, Phys. Rev. 123, 435 (1961),

had observed for lead. Little noted that the compressed layer of helium at the interface considered by Challis, Dransfeld, and Wilks would also assist this electron-phonon heat-transfer mechanism and also would modify its temperature dependence.

This was the situation at the beginning of our experiments. Observations had been made principally on copper and also on lead and quartz. The measured Kapitza resistances  $A\Delta T/\dot{Q}$  were of the order of  $10~T^{-n}$  deg W<sup>-1</sup> cm<sup>2</sup>, where A is the solid-helium interface area,  $\Delta T$  is the temperature jump,  $\dot{Q}$  is the heat current through the interface, and 2.5 < n < 4.0.

The purpose of the experiments reported here was to provide information for a comparison of the observed dependence of the Kapitza resistance on the nature of the solid with that predicted by the theories of Khalatnikov and Little and a theory developed by Bloch<sup>11</sup> during the course of this investigation. We have made measurements of the Kapitza resistance of samples of copper, gold, tungsten, single-crystal lithium fluoride, and single-crystal silicon in the temperature range from 1.25 to 2.1°K. We give first some experimental details to show how our measurements were made and then present our results. Following this we discuss these results in relation to the theories. Finally, we assess the extent of our present understanding of the Kapitza resistance.

#### **EXPERIMENTS**

In order to obtain information for a comparative study of the Kapitza resistance an experimental apparatus was required which was suitable for making measurements of sufficient accuracy on several different kinds of samples. We describe first our apparatus and then our information which it has provided.

## Technique

The experimental apparatus is shown in Fig. 1. The solid samples were all cylindrical in shape with an outside diameter of about 2 cm; some were about 1 cm long, others only 0.005 in. The interface area was typically about  $1\frac{1}{2}$  cm². The sample was pressed against a thin (<0.005 in.) indium O-ring seated on a flange which had been spun on the end of a  $\frac{5}{8}$  in. o.d., 0.010 in. wall, stainless steel tube. This tube, 3 cm long, served as the sample support. The sample, heater, and thermometers were sufficiently isolated from all their surroundings except the liquid helium at the interface by conventional low temperature techniques.

The temperature differences across the interface were of the order of 50 mdeg. It took several minutes to record all of the data for one value of heater power at one temperature. In order that  $\Delta T$  not drift more than 0.1% due to the drift of the helium bath temperature, the bath temperature must be held fixed to within about 10  $\mu$ deg for about 5 min. Such temperature

stability was achieved by an electronic system of temperature control whose concept has been outlined by Boyle and Brown, <sup>12</sup>

The temperature of the liquid helium at the solidhelium interface was assumed to be the same as the temperature of the liquid helium bath. This assumption introduces a systematic error in the value of  $\Delta T$  whose magnitude is never greater than 0.1% for the temperature range and heat fluxes of these experiments. The temperature of the helium bath was determined by the measurement of the vapor pressure of the liquid helium and the use of the 1958 He<sup>4</sup> scale of temperatures. The uncertainty inherent in the vapor pressure manometer readings amounted to  $\pm 0.0064$  mm of mercury. This corresponds to an absolute error in the temperature of the liquid helium at the interface of  $\pm 0.0010$  at 1.25°K down to  $\pm 0.0001$  at 2.10°K.

The temperature of the solid at the interface was determined from the carbon resistance thermometers. The resistance thermometers were carbon composition resistors of the  $\frac{1}{10}$ -W size manufactured by Allen-Bradley Company. For a parallel pair of nominally 67  $\Omega$  resistors we found that (-1/R)dR/dT was nearly constant and equal to 2.5 deg<sup>-1</sup> over the temperature range of 1.2 to 2.1°K. The resistance of these thermometers was measured in an ac bridge. The resistor was one arm of a Wheatstone bridge which was fed with an attenuated 150-cps signal. No more than  $2\times10^{-10}$  W were dissipated in the thermometers. The null setting of the bridge was detected with a high-gain

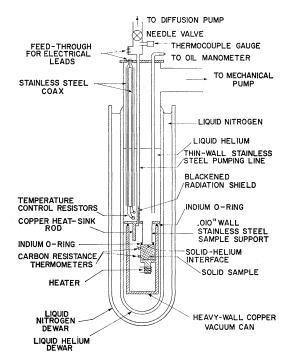


Fig. 1. A schematic drawing of the cryostat.

<sup>&</sup>lt;sup>11</sup> F. Bloch (private communication).

<sup>&</sup>lt;sup>12</sup> W. S. Boyle and J. B. Brown, Rev. Sci. Instr. 25, 359 (1954).

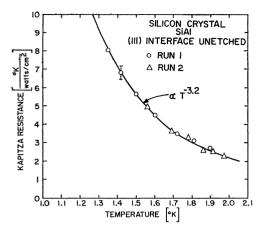


Fig. 2. The Kapitza resistance of silicon sample SiA1 whose surface was left unetched after being polished. The error bars shown do not include the error in magnitude of the interface area (see text). It will be noted that there are no error bars at the higher temperatures. This is so because the percentage error has diminished to about  $\pm 1\frac{1}{2}\%$  and cannot be conveniently displayed.

voltage amplifier and lock-in detector.<sup>13</sup> We were able to ascertain the resistance of the thermometer to within one part in 1000. In view of the value of (1/R)dR/dT, the error in  $\Delta T$  incurred by this uncertainty was about  $\pm 1$  to  $\pm 3\%$ . The temperature corresponding to any value of resistance was determined with the formula given by Clement.<sup>14</sup> This formula, which we found sufficiently precise and quite easy to handle, is  $\log R = T(a + b \log R)^2$ .

The nichrome wire heater was supplied with current from dry cells. The heater power was set to give heat fluxes between 2 and 49 mW/cm<sup>2</sup>. Mallory mercury reference batteries used in conjunction with a potentiometer gave the heater power with an error of  $\pm 1\%$ .

For the experiments on copper a tool was devised for machining the surface while immersed in liquid helium. This permitted us to make an experiment on the cleaned surface, allow the surface to be contaminated in some desired fashion, and then make a second experiment on the contaminated surface which was still unchanged with respect to the large scale crystalline imperfections present. The cutter was a ½-in.-diam two-flute end mill which had been drilled and tapped to fit on the end of a tool steel shaft. This shaft passed through a brass sleeve bearing and extended from the sample nearly to the cover plate of the helium Dewar. A fitting on the upper end of the shaft was made to accept a keyed rod. After the helium had been transferred one inserted the keyed rod into the fitting through the open helium transfer hole, pushed down until the cutter pressed against the sample surface, and then turned the cutter by hand slowly and firmly with a knurled knob attached to the keyed rod. About twenty turns seemed to suffice.

<sup>13</sup> H. L. Cox, Rev. Sci. Instr. 24, 307 (1953).
 <sup>14</sup> J. R. Clement, *Temperature* (Reinhold Publishing Corporation, New York, New York, 1955), Vol. II, p. 380.

We found that a premachining of the sample before installation in the Dewar made the operation much easier and more assured of success.

The interface area was measured with a linear scale with an error of  $\pm 3\%$ . When the relative magnitudes of the Kapitza resistance are considered in those cases where the interface area is known to have been constant we need not account for this error. In particular, this was the case for the pairs of runs on cleaned copper. It is also true for any single run, i.e., the evaluation of the temperature dependence of the Kapitza resistance for any run does not involve this error.

We can summarize the sources and magnitudes of the errors. They are  $\pm 1\frac{1}{2}\%$  of the heat current  $\dot{Q}$ (including heat leaks),  $\pm 1$  to  $\pm 3\%$  of the temperature jump  $\Delta T$ , and (where it applies)  $\pm 3\%$  of the interface area A.

Finally, we have corrected the experimental values of  $A\Delta T/Q$  to account for the second-order dependence of the heat current O on the temperature jump  $\Delta T$ . Thus, the results presented below give the Kapitza resistance in the limit  $\Delta T \rightarrow 0$ .

#### The Results

## Single Crystal Silicon

We felt that it would be useful to compare the Kapitza resistances of samples of a solid which had different dislocation densities present. When the surface disturbances of the helium phonons encounter the elastic strain fields of such large-scale crystalline imperfections as dislocations and grain boundaries they should suffer scattering. As Little<sup>8</sup> had suggested, such a mechanism would provide an avenue for the exchange of thermal energy across the interface. The fact that this mechanism should be present in nonmetals as well as metals was particularly important; Dransfeld and Wilks had found that quartz exhibited a Kapitza resistance of the same order of magnitude as copper

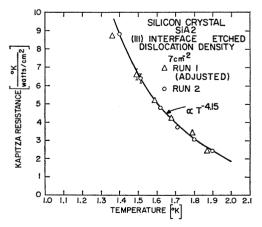


Fig. 3. The Kapitza resistance of silicon sample  $\mathrm{Si}A2$ whose surface was etched.

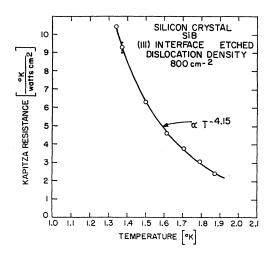


Fig. 4. The Kapitza resistance of silicon sample SiB whose surface was etched.

and lead. Crystals of silicon are easily obtained with widely different dislocation densities and there exist reliable techniques for revealing the dislocations and estimating their density.<sup>15</sup>

Two crystals of silicon were obtained from Knapic Electro-Physics, Inc., of Palo Alto, California. They are referred to as SiA1, SiA2, and SiB and their specifications follow:

SiA1 Single crystal grown in the [111] direction; p-type; resistivity greater than  $1000 \Omega$ -cm; impurity concentration less than  $1.3 \times 10^{13}$  atoms/cm³; dislocation density 7 cm<sup>-2</sup>; the interfacial plane was ground and polished normal to the growth axis and left unetched.

SiA2 This sample was obtained by etching the interfacial surface of the crystal called SiA1.

SiB Single crystal grown in the [111] direction; p-type resistivity greater than 1000Ω-cm; impurity concentration less than 1.3×10<sup>13</sup> atoms/cm³; dislocation density 800 cm<sup>-2</sup>; the interfacial plane was ground and polished normal to the growth axis and etched.

These crystals initially were rods 2 in. in length and  $\frac{7}{8}$  in. in diameter Centerless grinding reduced the diameter of  $1\frac{1}{4}$  in. of their length to 0.575 in. so that they would fit into our sample support.

Two experimental runs were made on the unetched silicon crystal with low dislocation density, SiA1. The results are displayed graphically in Fig. 2. The results may be closely represented by the expression  $A\Delta T/\dot{Q}=21~T^{-3.2}$  deg W<sup>-1</sup> cm². Two runs were made on the etched silicon crystal with low dislocation density, SiA2. The results are shown in Fig. 3, and they are well represented by the expression  $A\Delta T/\dot{Q}=36~T^{-4.15}$  deg W<sup>-1</sup> cm². A run was made on the etched silicon crystal with the higher dislocation density, SiB. The results are shown in Fig. 4, and they are represented by  $A\Delta T/\dot{Q}=34~T^{-4.15}$  deg W<sup>-1</sup> cm².

We can make a comparison between the results for SiB and SiA2. The experimental environment and

surface treatment for these two crystals were the same. The dislocation density was greater by a factor of 100 in SiB than that in SiA2. Within the limits of experimental error the Kapitza resistances which they exhibited were idential, approximately 35  $T^{-4.15}$  deg W<sup>-1</sup> cm². Furthermore, it appeared that etching away the surface damage present on SiA1 both substantially increased the magnitude of the Kapitza resistance and increased the temperature dependence by a factor of  $T^{-1}$ . The results obtained for the SiA2 sample have considerable intrinsic value because of the high degree of perfection of this crystal.

## Polycrystalline Gold

Interest in the possibility that the electron-phonon interactions in the vicinity of the interface might be mainly responsible, in metals, for the heat transfer across the interface led us to experiment on gold. Gold is quite inert and forms no stable oxides. Hence, its surface should be clean except for a layer of adsorbed gas atoms.

Two experimental runs were made on gold sample AuA and one run on AuC. Both samples were circular disks 2 cm o.d. cut from 0.005 in. thick, 99.99% pure, gold sheet. The results are shown in Fig. 5. The results for AuA can be represented by  $A\Delta T/\dot{Q} = 7.8 \ T^{-3}$  deg W<sup>-1</sup> cm<sup>2</sup>, and those for AuC by  $8.3 \ T^{-3}$  deg W<sup>-1</sup> cm<sup>2</sup>.

## Polycrystalline Copper

We measured next the Kapitza resistance of a clean metal surface. The stimulus for this arose out of the interesting idea of Bloch. He calculated what the heat transfer should be if the helium atoms could interact directly with the conduction electrons which leak through a clean metal surface. From this estimate the expectation was that the Kapitza resistance of a clean copper surface would be less than that of a dirty

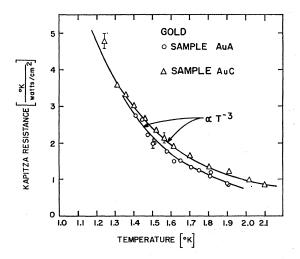


Fig. 5. The Kapitza resistance of gold samples AuA and AuC.

<sup>&</sup>lt;sup>15</sup> W. C. Dash, J. Appl. Phys. 27, 1193 (1956).

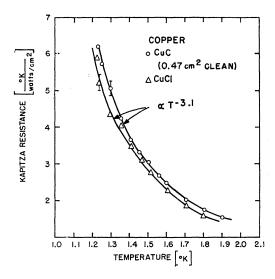


Fig. 6. The Kapitza resistance of copper samples CuC and CuC1.

surface and would exhibit a dependence on the temperature to the inverse fifth power. We say more about this theory below.

In these experiments we cleaned the interface under liquid helium with our cutting tool and then measured its Kapitza resistance. Then the sample was exposed to the atmosphere for about a week at which time we repeated the measurements for the oxidized surface. Two pairs of such experiments were carried out on samples of OFHC copper. The samples are designated as follows:

CuC 0.47 of 1.61 cm<sup>2</sup> were cleaned.

CuC1 This is sample CuC after exposure to air.

CuD 0.85 of 1.54 cm<sup>2</sup> were cleaned.

CuD1 This is sample CuD after exposure to air.

The samples were the same shape and about the same size as the silicon samples. Their interfacial surfaces were initially cut on a lathe. Subsequently, they were cleaned chemically and rinsed in water and ethanol before being installed in the cryostat. The results are shown in Figs. 6 and 7.

The surface, part of which has been cleaned, consistently exhibits a larger Kapitza resistance than does the surface which is entirely oxidized. We conclude that the clean region of the copper surface offers a considerably higher Kapitza resistance than the oxidized surface. We have done other experiments on copper which also give qualitative support to this conclusion. Our results can be represented as follows:

Sample	Kapitza resistance	Sample	Kapitza resistance
CuC	11 T <sup>-3.1</sup>	$\mathrm{Cu}D$	11 $T^{-3.0}$
CuC1	$10 \ T^{-3.1}$	CuD1	$7 T^{-2.6}$
(Units	are deg W <sup>-1</sup> cm <sup>2</sup> )		

From the results for CuD and CuD1 we estimated the Kapitza resistance of a clean copper surface and found

that it could be represented by  $A\Delta T/\dot{Q}=19~T^{-3.6}$  deg W<sup>-1</sup> cm<sup>2</sup>. The results for CuC can be predicted to within 4% by using our estimated resistance for a clean surface in conjunction with the results for CuC1.

It is well to note that the pieces of copper that were cut from the surface lay on the interface throughout the run. We were careful not to disturb the system during the exposure of the surface to the atmosphere. The second run was carried out with the chips still lying on the interface and the cutting tool in place where it had been left after being withdrawn from the cut surface. Examination of the cutting residue showed it to consist of a few shavings of copper in the form of curls. The thermal contact of such a residue with the solid sample surely would have been so poor that its effect on the Kapitza resistance was considered to have been negligible.

### Single Crystal Lithium Fluoride

Lithium fluoride is a solid in which there is no possibility for heat transport by charge carriers at liquid-helium temperatures. An interface which is exceedingly smooth may be obtained by utilizing a cleavage surface of such a crystal. Our lithium fluoride sample was a 99.99% pure crystal made by the Harshaw Chemical Company, Cleveland, Ohio. The sample was in the shape of a solid rod 2 cm o.d. and  $\frac{1}{2}$  in. long with (100) cleavage plane ends. Initially we experienced difficulty from cracks which developed in the crystal which was subjected to stresses applied by the sample support assembly as the temperature was lowered. Success was finally achieved by replacing the indium wire usually employed as an O-ring with a stack of three thin annular rings of indium made by flattening wire O-rings. This apparently reduced the stress on the crystal face at the contact between the crystal and the O-ring sufficiently so that no cracks developed. The results are presented in Fig. 8. They may be represented by  $A\Delta T/\dot{Q}=23~T^{-3.75}~{\rm deg~W^{-1}~cm^2}$ .

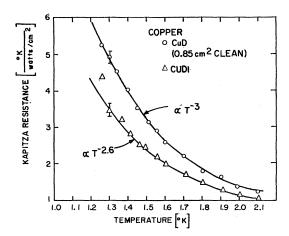


Fig. 7. The Kapitza resistance of copper samples CuD and CuD1.

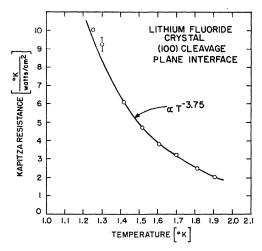


Fig. 8. The Kapitza resistance of lithium fluoride.

## Polycrystalline Tungsten

Tungsten is a solid for which the Khalatnikov theory predicts a very large Kapitza resistance. Also, the electron-phonon interaction for this metal is several times larger than for copper or gold. This fact makes tungsten interesting from the point of view of the theory of Little.

Our sample was 99.95% pure tungsten, and its dimensions were the same as those of the gold sample. The results are shown in Fig. 9 and they are represented by  $A\Delta T/\dot{Q}=39~T^{-3.5}$  deg W<sup>-1</sup> cm<sup>2</sup>.

## DISCUSSION

We consider here these results in relation to the heat transfer mechanisms of Khalatnikov, Little, and Bloch. A tabulation of experimental results and the corresponding theoretical predictions is given in Table I. For reasons of comparison involving other mechanisms we have calculated in addition the van der Waals interaction energies between samples in these experiments. These are also presented in Table I.

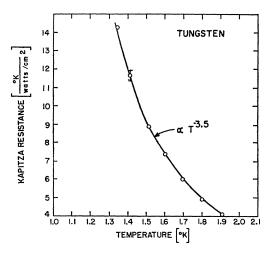


Fig. 9. The Kapitza resistance of tungsten.

#### The Acoustic Transfer of Heat

The principal mechanism for the transfer of heat between a solid body and liquid helium II was shown by Khalatnikov<sup>5</sup> to be the emission or absorption of thermal vibrations from the vibrating surface of the solid which is in contact with the helium. His expression for the Kapitza resistance for a solid of density  $\rho$  and transverse velocity of sound  $c_t$  in contact with liquid helium of density  $\rho_{\text{He}}$  and velocity of sound  $c_{\text{He}}$  is

$$\frac{\Delta T}{\dot{Q}/A} = \frac{15h^3}{16\pi^5 k^4 \rho_{\rm He} c_{\rm He} F} \frac{\rho c_t{}^3}{T^3} \frac{{\rm deg}}{{\rm W/cm^2}},$$

where F is a function of the ratio of transverse to longitudinal sound velocities and is equal to 1.5 for most solids. Upon substitution of the values

$$\rho_{\text{He}} = 0.145 \text{ g/cm}^2$$
,  $c_{\text{He}} = 2.4 \times 10^4 \text{ cm/sec}$ ,  $F = 1.5$ ,

into Khalatnikov's expression for the Kapitza resist-

Table I. The measured magnitudes and some predicted magnitudes of the Kapitza resistance. Some van der Waals interaction energies between a helium atom and solid surfaces.

Solid	Experiment, Kapitza resistance $\left[\frac{\deg}{(W/\mathrm{cm}^2)}\right]$	Khalatnikov, Kapitza resistance $\left[\frac{\deg}{(W/\text{cm}^2)}\right]$	Van der Waals interaction energy $W$ . Helium atom distance $D_0$ ( $\mathring{\mathbb{A}}$ s) from surface $[eV]$	CDW input impedance (approximate magnitude)	$\begin{bmatrix} \text{CDW} \\ \text{Kapitza} \\ \text{resistance} \\ \text{(approximate)} \\ \begin{bmatrix} \frac{\deg}{(\text{W/cm}^2)} \end{bmatrix} \end{bmatrix}$	Little theory for metals, Kapitza resistance (relative magnitudes)
Silicon crystal Gold	35 T <sup>-4.15</sup> (mean) 8.0 T <sup>-3</sup> (mean)	1130 T <sup>-3</sup> 158 T <sup>-3</sup>	$0.068 D_0^{-3} \ 0.24 D_0^{-3}$	$6.5 \times 10^{3}$ $9.4 \times 10^{3}$	$\begin{array}{c} 490 \ T^{-4.2} \\ 47 \ T^{-4.2} \end{array}$	1
Copper	•••	498 T-3	•••	•••		• • • •
$\dot{ ext{Cu}}D$	11 $T^{-3}$		• • •	• • •	• • •	• • •
$\mathrm{Cu}D1$	$6.9 T^{-2.6}$	• • •	$0.040 D_0^{-3}$	$5.5 \times 10^{3}$	$254\ T^{-4.2}$	>0.3
clean	19 $T^{-3.6}$	• • •	$0.26 \ D_0^{-3}$	$9.5 \times 10^{3}$	$147 T^{-4.2}$	0.3
Lithium fluoride crystal	$23 T^{-3.75}$	$1130 \ T^{-3}$	$0.085 D_0^{-3}$	$7.0 \times 10^{3}$	$450 T^{-4.2}$	• • •
Tungsten	$39\ T^{-3.5}$	2040 T-3	•••	•••	•••	•••

ance, we get

$$\frac{\Delta T}{\dot{Q}/A} = \frac{4.5 \times 10^{-15} \rho c_t^3}{T^3} \frac{\text{deg}}{\text{W/cm}^2}.$$

In Table I the column labeled Khalatnikov has the Kapitza resistance as calculated from Khalatnikov's formula where appropriate sound velocities were used. As Challis, Dransfield, and Wilks<sup>7</sup> have already observed in the cases of copper, lead, and quartz, we note the following from Table I. The experimental values of the resistance are 20 to 50 times smaller in magnitude than predicted, and the exact dependence of the resistance of the inverse third power of the temperature is not observed. Also, the resistance is not proportional to  $(\rho c_t^3)$  for the solid.

In an effort to explain their experimental results Challis, Dransfield, and Wilks<sup>7</sup> (hereafter referred to as CDW) took into account the fact that van der Waals interaction between the solid surface and the helium atoms should compress the liquid helium in the vicinity of the interface. This compression should raise the helium density and velocity of sound to values approximately the same as for solid helium within a thin layer of helium at the interface. CDW modified the result of Khalatnikov's theory by replacing the quantity  $(\rho c)_{\text{helium}}$  (the acoustic impedance of the liquid) in Khalatnikov's expression for the heat flux with their approximation to the acoustic impedance of the liquid as seen by the solid through the acoustic matching unit formed by this thin compressed layer.

In order to evaluate the merit of the CDW modification one needs to know the magnitude of the van der Waals force field acting on the liquid helium. We have calculated the van der Waals interaction energies between a helium atom and the surfaces of most of the solid samples. The interaction energy W between a helium atom and a metal surface was calculated with the Margenau-Pollard16 formula. The interaction energy  $\langle \Delta E \rangle_{\rm av}$  between an insulator atom and a helium atom separated by a distance R was calculated from the formula in Margenau's17 review paper. A formula, which is easily derived,18 gives the total interaction energy W between an insulator surface and a helium atom as follows:

$$W = (\pi N/6D_0^3)(\langle \Delta E \rangle_{\rm av} R^6)$$
 ergs,

where N is the number of interacting insulator atoms per unit volume,  $D_0$  is the distance between the insulator surface and the helium atom, and  $(\langle \Delta E \rangle_{av} R^6)$  is a constant independent of R for the van der Waal's interaction between a given pair of atoms. These interaction energies are listed in Table I.

This information was then applied to the work of CDW as follows. They calculated the real part of the input impedance to the helium as a function of frequency and presented this graphically, but only for the specific case of copper for which they used an interaction energy,  $W_c = 0.54 \times 10^{-36} D_0^3$  ergs. We can make general use of their graph because the input impedance of a line of length x at the frequency  $\nu$  is a function only of the product  $(x\nu)$ . Since the line length x must be proportional to  $W^{1/3}$ , then we can find the input impedance at frequency  $\nu$  for our interaction energy W by locating the impedance on the CDW graph at  $\nu(W/W_c)^{1/3}$ . This was done for the five solids for which we have evaluated W at  $\nu = 5.9k(1.5)/h \text{ sec}^{-1}$ , the frequency at which the maximum energy is emitted at 1.5°K (see CDW). The results are listed in Table I. The Kapitza resistance which this scheme predicts is very nearly equal to 2.8×10<sup>3</sup> times the Khalatnikov value divided by the input impedance in cgs units, and this is presented in the table.

The oxidized copper is considered to give the same van der Waals interaction energy with a helium atom as would a sample of Cu<sub>2</sub>O. This is reasonable in view of the fact that the van der Waals interaction with the solid extends only a distance  $R_0$  of about 20 Å into the liquid and the thickness t of the oxide layer can be expected to be at least 100 Å. Then  $R_0^3 \ll t^3$ , and the interaction energy can safely be regarded as entirely due to the Cu<sub>2</sub>O.

In all cases under consideration with the exception of the cleaned copper surface we expect to have a monolayer of physically adsorbed nitrogen, oxygen, and water frozen onto the interface. Qualitatively, we can estimate the effect of this. We imagine that the surface layer of solid atoms is replaced by a layer of atoms of a molecular solid having about unit density and a velocity of sound of about 105 cm/sec. The van der Waals interaction energy between helium atoms and the atoms of this layer can be estimated by assuming the atomic polarizability of these atoms to be about 10<sup>-24</sup> cm<sup>3</sup> and their principal optical transition energy to be about 10 eV. It is found that the energy of interaction of a helium atom with this monolayer is greater than that of a helium atom with a Cu<sub>2</sub>O monolayer, less than with a LiF or Au monolayer, and about the same as with a Si monolayer. The effect of the monolayer should be to improve the acoustic match in the case of Cu<sub>2</sub>O, to worsen the match in the cases of LiF and Au, and to leave the case of Si about the

We see from Table I that, although considerations of matching do indeed bring the predicted magnitudes of the Kapitza resistance closer to the measured magnitudes, the predicted relative magnitudes for different solids fail to agree with experiment; in some cases the disagreement is more serious than with the simpler Khalatnikov theory. For example, experiment shows the Kapitza resistance of an oxidized copper surface to be

<sup>&</sup>lt;sup>16</sup> H. Margenau and W. G. Pollard, Phys. Rev. 60, 128 (1941), Eq. (12).

17 H. Margenau, Rev. Mod. Phys. 11, 1 (1939), Eq. (10).

18 S. Franchetti, Nuovo Cimento 4, 1504 (1956).

about half as large as that of the clean copper surface. The CDW picture says that an oxidized copper surface should have a resistance about 80% greater (diminished some by the presence of the frozen monolayer) than the clean copper surface because the van der Waals interaction energy for the Cu<sub>2</sub>O-He case is only one-sixth of that for the Cu-He case. The oxide layer itself might help match the copper to the helium but it is doubtful that it could affect the necessary increase in the input impedance to the helium. In another case, experiment shows that the resistance for a silicon surface is about 1.55 times that for LiF. The CDW theory says it should be 1.07 and the Khalatnikov theory says they should be the same (here the presence of a frozen monolayer would make the ratio less than 1.07 according to CDW). Furthermore, the CDW mechanism predicts a temperature dependence for the resistance of  $T^{-4.2}$  and the resistance is observed to depend on the temperature to powers from -4.15 to -2.5.

We must conclude that the acoustic mismatch theory modified to consider the compressed helium at the interface does not provide an explanation of the Kapitza resistance.

# The Interaction of the Metal Electrons with the Surface

Disturbances of Helium Phonons

Little<sup>10</sup> has considered the transfer of heat across an interface by means of the interaction of the metal electrons with the surface disturbances of the helium phonons which are totally reflected at the interface. The calculation gave a result which may be compared with our results for gold, copper, and tungsten. The result was as follows:

$$\frac{\Delta T}{\dot{Q}/A} = \frac{(2\pi)^5 \hbar^6 (\rho c^3)_{\rm He}}{8k^4 m^2 J_4(\infty) (eV_1)^2 F(\theta) T^3}$$

where  $(eV_1)$  is the electron-phonon interaction energy,  $J_4(\infty)$  is a definite integral about equal to 26, and  $F(\theta)$ is a definite integral proportional to  $(\rho^2 c^3)_{\text{He}}/(\rho^2 c^3)_{\text{solid}}$ . We see that  $\Delta T/(Q/A)$  is proportional to  $(\rho^2 c^3)_{\text{solid}}/$  $\rho_{\rm He}(eV_1)^2$ . We can compute the ratio of the Kapitza resistance offered by this mechanism for two metals. Estimates of this ratio appear in Table I under the column headed Little theory for metals. It is evident that the data do not support this theory either. The acoustic properties of copper and the large electronphonon interaction in tungsten should, respectively, serve to make the Kapitza resistances of copper and tungsten less than that of gold. Just the reverse is observed. The results for clean copper and for gold are a particularly good test for this theory because neither had a surface oxide layer. This mechanism would be expected to be less effective when an oxide layer is present on the surface, but the oxidized copper surface was observed to have a lower resistance than the clean surface. Finally, the predicted magnitudes of the resistances for this mechanism in the cases of copper and gold are about 100 times the value,  $1.85 \times 10^3$  deg W<sup>-1</sup> cm², which Little calculated for lead. These magnitudes are much greater than any observed. We must conclude that this mechanism may exist but is so much weaker than the mechanism principally responsible for the heat transfer that its presence has not been observed in our experiments.

# The Direct Interaction of Metal Electrons with the Helium Atoms

In an independent consideration of the problem of the Kapitza resistance, Bloch<sup>11</sup> has made the important observation that one must look for a mechanism where the high inertia  $(\rho_{\text{solid}}\gg\rho_{\text{He}})$  and the high rigidity of the solid  $(c_{\text{solid}}\gg c_{\text{He}})$  are not important, i.e., where one may even consider the lattice as rigid and yet obtain an energy transfer.

This led him to consider the conduction electrons and the fact that they leak out of the metal and could interact directly with the helium atoms. The interaction between the system of electrons of the metal and the phonons of the liquid causes the creation or annihilation of helium phonons with corresponding jumps down or up in energy states of metal electrons. Bloch has calculated the heat transfer by this mechanism. The final result is

Heat flux = 
$$\frac{8}{3} \left( \frac{E_F}{W} \right)^3 \left[ \frac{c_{\text{He}}}{V_F} \frac{\rho_{\text{He}}}{\rho_e} \left( \frac{mV_F a}{h} \right)^2 \right] \times \frac{1}{c_{\text{He}^2}} \left( \frac{kT}{Mc_{\text{He}^2}} \right)^2 \frac{(kT)^4}{h^3} I_e,$$

where  $E_F$  and  $V_F$  are the Fermi energy and velocity, respectively; W is the work function of the metal;  $c_{\text{He}}$  is the velocity of sound in helium;  $\rho_e$  is the mass density of electrons; a is about equal to the atomic radius of helium; m and M are the mass of the electron and helium atom, respectively;  $I_e$  is a definite integral, and  $\rho_{\text{He}}$  is the mass density of the liquid helium.

Bloch has estimated the ratio of the heat flux for his mechanism to the heat flux for the acoustic mechanism of Khalatnikov and found it to be equal to

(a quantity of order unity) 
$$\frac{(\rho_{\text{metal}}c_{\text{metal}}^3)}{(\rho_{\text{He}}c_{\text{He}}^3)} \left(\frac{kT}{Mc_{\text{He}}^2}\right)^2$$
,

where  $c_{\text{metal}}$  is the velocity of sound in the metal and  $\rho_{\text{metal}}$  is the mass density of the metal. We find that this is equal to about 10  $T^2$ .

With this in mind we considered the results for copper. No evidence of this mechanism was found. First, it was clearly demonstrated that the clean copper

surface exhibited a higher Kapitza resistance than did the oxidized surface. Since the cleaning of the copper should have uncovered this avenue for the heat transfer and reduced the Kapitza resistance, it must be that this mechanism is weaker than estimated and its presence is not observable in our experiments. Second, we observed no temperature dependence of the Kapitza resistance as great as the inverse fifth power. This could also be understood if the mechanism is too weak to be seen. We must conclude that, within the limits of experimental error, the mechanism of Bloch for the transfer of heat across a metal-helium interface either is too weak to be observed or is rendered ineffective by some factor which has been neglected.

#### CONCLUSION

The information provided by these experiments has improved the perspective of our view of the Kapitza resistance. Our results, combined with those obtained by other experimenters, show that none of the mechanisms which so far have been considered in theories provide an explanation of the Kapitza resistance. Either the mechanisms are too weak to provide an avenue of sufficiently low thermal resistance, or something is missing from the models chosen to represent the thermal contact at the interface. We see that theoretical progress on this problem is likely only if

two points are kept in mind. First, the mechanism mainly responsible for the Kapitza resistance is probably very different from, and not merely a modification of, those weak mechanisms which have been considered in the past. Second, the factors which are missing from a strong mechanism, such as the model of Bloch, must be found and successfully incorporated in the theory.

Part of the answer to this problem may lie in considering the adsorption and desorption of helium atoms from the solid surface—a process which has not been taken into account in previous theories. An atom adsorbed on the solid surface would have initially an energy appropriate to the temperature of the liquid; however, after a characteristic time, depending upon its coupling to the solid, its energy would approach that appropriate to the temperature of the solid and upon desorption would carry heat from the solid to the liquid or vice versa. Such a process gives promise of explaining some of the present difficulties, but will be left to a later publication for its further development.

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