Compressibility and Heat Transfer of Helium II^*

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It has been demonstrated that the thermomechanical effect operates in a closed system of helium II. This effect has been used to measure the compressibility of helium II. The results agree relatively well with the results of other investigators who used other methods. The agreement indicates that the method used, a packed column of rouge in a closed system, is a good one for obtaining the full thermomechanical effect. The heat transfer of helium II in the rouge column has been measured. The behavior found is, in general, similar to that found for the heat transfer through a slit between two optically polished surfaces. Evidence was found for a maximum in the heat transfer close to, but below, the λ point.

I. INTRODVCTION

 $\int T$ has been found¹ that when a sufficiently large heat current passes down through a closed system of helium II in channels in packed rouge, a sudden rise in the liquid level is produced. When the heat is turned on, a temperature difference is established and this, by virtue of the thermomechanical effect,² produces an underpressure in the helium in the rouge. This process may be thought of as a "relative stretch" of the two components—that is, a "Le Chatelier stretch." The relative concentration of normal to superfluid component is greater above than below because of the difference in temperature and by Le Chatelier's principle a compensation mechanism, which in this case is a pressure difference, results tending to equalize the concentration difference. The superfluid component moves with no viscosity, so the pressure is less below. This should result in a decreased density of the helium in the rouge. Although this was not definitely established because of the small differences in level before the sudden rise, other evidence presented here makes it reasonable.

As the heat current and temperature difference increase the underpressure increases until small bubbles are formed. These bubbles were not seen but their existence was inferred from a comparison of the underpressure necessary to form bubbles of the radius (2 microns) ot the rouge channels and of the underpressure calculated from London's thermomechanical equation.³ There was good agreement between these two quantities. It had been shown that London's equation could be applied approximately for these channel sizes by opening the system at the bottom and measuring the thermomechanical effect directly.

It is interesting to note that the heat current at the

rise was reproducible only when the apparatus was tapped. When the apparatus was not tapped the sudden rise was delayed until higher heat currents were reached. Calculations of the pressure in the channels while these high heat currents were passing through indicated that the liquid was under a negative pressure—that is, a tension.⁴

Since this work established that the thermomechanical effect operates in a closed system and can produce underpressures, the question arose as to whether the effect could be reversed and used for measuring the compressibility of helium II. Furthermore it previously had been reported by Allen and Misener⁵ that in the case of open systems and nonsteady-state conditions the gravitational flow of helium II through tightly packed rouge was identical with the flow in narrow smooth channels. Bowers, Chandrasekhar, and Mendelssohn⁶ and Chandrasekhar and Mendelssohn⁷ also found that the flow of helium II towards a heat source in compressed powder (in an open system) was the same as for flow through a slit. It would then appear that the thermal flow in packed rouge in a closed system should not be very different from thermal flow through a narrow slit. To check this, the heat transport property of helium II in the rouge channels was also investigated. The following sections describe these experiments.⁸

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¹ C. A. Reynolds, Phys. Rev. **93**, 1118 (1954).
² J. F. Allen and H. Jones, Nature 141, 243 (1938).
³ H. London, Proc. Roy. Soc. (London) 1**71,** 484 (1939).

⁴ L. Meyer and J. H. Mellink [Physica $13, 197$ (1947)] reporte the formation of bubbles in their heat transfer apparatus at times when conditions were not static and the fountain effect pressures could not be measured. The situation was such, though, that it was likely that the liquid was subjected to an underpressu brought about by the thermomechanical (fountain) effect. They assume that this underpressure must overcome the Van der Waals cohesion forces, which would amount to a stress of 40 atmospheres. It is more likely that a small sharp edge or a slight agitation of the apparatus started the bubble formation under the action of a smaller (relatively) thermomechanical effect.

⁵ J. F. Allen and H. D. Misener, Proc. Roy. Soc. (London) A172, 467 (1939).

⁶ Bowers, Chandrasekhar, and Mendelssohn, Phys. Rev. 80, 856 (1950).

⁷B. S. Chandrasekhar and K. Mendelssohn, Proc. Roy. Soc. (London) A218, 18 (1953).

Preliminary reports of this work have been given at the New York and Washington meetings of the American Physical Society LH. Forstat and C. A. Reynolds, Phys. Rev. 98, 1196(A) (1955) and 99, 669(A) (1955)].

II. COMPRESSIBILITY

a. Apparatus

The apparatus used in the compressibility measurements is shown in Fig. 1. The compressed volume section was made from Pyrex glass tubing, 30.6-mm inside diameter and 87.7 mm in length. A carbon thermometer (1-watt Allen-Bradley resistor of 270 ohms) and a manganin wire heater (575 ohms) were mounted inside this volume. A fritted disk of medium porosity (hole sizes ranging from 10 to 15 microns) was sealed to the top of the compressed volume. This disk was used primarily as a backing against which jeweller's rouge (ferric oxide) was packed. A total of 5.95 ^g of rouge was packed into the glass tubing to a height of 8.0 cm. To prevent the rouge from separating under vacuum, a thin perforated copper disk was pressed against the top of the rouge column. To hold this in place, the disk was attached to a 2-inch length of copper tubing that was silver-soldered to the Kovar seal. The other end of the tubing was joined to another Kovar-glass seal, to which was added a Pyrex glass tube of uniform cross sectional area of 0.229 cm' which served as the viewing-section of the apparatus. Another carbon thermometer $(\frac{1}{2}$ -watt Allen-Bradley, 200 ohms) was placed at the bottom of the copper tubing (equivalent to being on top of the rouge column). The compressed volume was 67 cc and the volume above the fritted disk (from the fritted disk to the zero fiducial mark in the viewing section) was 6.1 cc.

b. Experimental Procedure

Helium gas was condensed into the apparatus from an external filling system until the level was at a convenient viewing height and then the apparatus was

FIG. 1. Compressibility apparatus.

FIG. 2. Height of liquid level versus temperature.

closed off. Starting slightly below the λ point, the thermometers were calibrated at the same time that a height reading of the liquid level in the experimental apparatus was taken. When the operating temperature was reached, power was introduced into the system through the heater in the compressed volume and both thermometers were read and height levels taken as a function of power.

The path for the heat current was up through the rouge column and out into the helium bath through the copper tube. In this manner, the compressed volume was put under the action of the thermomechanical pressure.

c. Results

Figure 2 shows the curves obtained by plotting the height levels as a function of temperature. The curve marked zero power curve (solid line) was the one obtained for zero power input. This is nothing more than the density curve for liquid helium except for a negligible correction for the variable amount of mass in the gaseous state. The dotted-line curve (marked power curve) shows the points obtained when the bottom heater was turned on. The heights here are plotted as a function of the temperature of the compressed volume. The differences between the height at the power points at a given temperature and the height of the zero power curve at the same temperature indicate that the liquid was compressed.

In order to get some indication of the consistency of the data and to get some estimate of the relative errors in the experiment, all the runs for zero power were normalized to the same height at one temperature. Although it is possible to fill the experimental apparatus with helium to the same level for each run, it is easier to make this normalization. Figure 3 shows the curve obtained from this normalization process and indicates that the data from the various runs are consistent. The power points were also reduced and the differences between them and the normalized curve were used in the compressibility calculation.

The usual expression for the compressibility, χ ,

$$
\chi = -\frac{1}{V} \frac{\Delta V}{\Delta P},\tag{1}
$$

rewritten in terms of height differences, becomes

$$
\chi = -\frac{A}{V} \frac{\Delta H}{\Delta P},\tag{2}
$$

where $A = \text{cross-sectional area of the viewing tube in}$ cm², $V=$ compressed volume in cm³, $\Delta H=$ corrected height difference in cm, and ΔP = corrected thermomechanical pressure in dynes/cm'.

Two corrections were applied to the height differences to obtain the final ΔH used in Eq. (2). First, a correction was made for the compression of the liquid in the rouge column. This was a subtractive correction, since the only volume of interest for this compressibility experiment was the volume below the rouge. In calculating the correction, it was assumed that the thermomechanical pressure varied linearly with the length of the rouge column and that the compressibility was constant. This correction amounted to approximately 1% .

Since the height readings with power, which were plotted as a function of the bottom (hot) temperature, were taken while the top portion was still at a colder temperature, it was necessary to correct these readings so that they would correspond to the condition of the entire liquid when all of it is at the bottom (hot) temperature. This was an additive correction because the specific volume of helium II, varying inversely with temperature, tends to lower the height of the level as the temperature rises and thus the differences with the zero power curve at a given temperature would be larger if all of the liquid were at the bottom temperature. This correction amounted to approximately 4% . Hence the over-all corrections to the height differences amounted to approximately 3% .

The value for ΔP in Eq. (2) was obtained by using the integrated form of London's thermomechanical equation' with more recent values for the density' and equation^s with more recent values for the density^s and
the specific entropy.¹⁰ The equation used was as follows

$$
\Delta P = (2.91/6.58)(T_b^{6.58} - T_t^{6.58}) \times 10^4 \text{ dynes/cm}^2, (3)
$$

Fro. 3. Normalized curve of zero poorer heights.

where T_b =temperature in the compressed volume in ${}^{\circ}$ K and T_t = temperature at the top of the rouge column in K . ΔP in Eq. (3) was corrected for the vapor pressure difference between the top and bottom of the rouge column. This correction amounted to approximately 5%. The corrected ΔP was then used in Eq. (2) to obtain the value for the compressibility.

Table I gives the results for the compressibility experiments. The last four rows were obtained with the apparatus used in the heat transfer experiments (to be discussed in the next section). The largest error arose in the height measurements. The height levels were measured with a cathetometer with an accuracy of ± 0.01 cm. Since it was necessary to record differences in height level, the error was increased to ± 0.02 cm. The height differences used in the calculations varied from 0.06 cm for small powers to 0.65 cm for large powers. This amounted to an average error of approximately 10%. The error of each determination of the compressibility is indicated in the last column of Table I.

In these experiments the accuracy was not sufhcient to show a dependence of compressibility on temperature. In any event, one would expect such a dependence to be small. Furthermore, no dependence on the thermomechanical pressure was found.

These measurements are in good agreement, within the experimental error, with results obtained by other

^{&#}x27; K. R. Atkins (private communication).

¹⁰ Kramers, Wasscher, and Gorter, Physica 18, 329 (1952).

$T.~\rm{^o K}$	ΔT . ^o K	ΔP	x	
bottom	(T_b-T_t)	(atmos)	$(atmos-1)$	
1.883	0.025	22×10^{-3}	$8 + 3 \times 10^{-3}$	
1.883	0.025	22	$8 + 3$	
1.969	0.111	110	$9 + 1$	
2.053	0.191	220	9 ± 1	
2.087	0.215	270	$9 + 1$	
2.089	0.214	270	$9 + 1$	
1.997	0.015	18	$11 + 4$	
2.021	0.038	50	$12 + 2$	
2.127	0.143	220	$10 + 1$	
2.163	0.175	280	$_{8\pm 1}$	
2.052	0.193	22	$9 + 1$	
2.052	0.193	22	9 ± 1	
2.052	0.193	22	9 ± 1	
2.042	0.181	20	$10 + 1$	
2.051	0.184	21	$10 + 1$	
2.052	0.172	20	$10 + 1$	
1.906	0.054	49	$10 + 2$	
1.953	0.097	95	$10 + 2$	
1.984	0.121	126	$12 + 1$	
1.985	0.119	125	13 ± 1	

TABLE I. Compressibility results.

investigators. Keesom and Clusius¹¹ measured the compressibility of liquid helium by a method involving the measurement of the amount of helium removed during a known pressure decrease. They obtained as a result, $\chi=11.5\times10^{-3}$ atmos⁻¹. Atkins and Chase¹² calculated the compressibility of liquid helium by measuring the velocity of first sound in liquid helium and applied these results to the expression,

$$
\chi = 1/\rho u_1^2,\tag{4}
$$

where ρ =density of liquid helium and u_1 =velocity of first sound. They obtained a value for the compressibility which was slightly temperature-dependent, but yielded, as an average value, 13.0×10^{-3} atmos⁻¹.

A confirmation of the bubble formation when the heat current is reversed, which was mentioned in the Introduction, was obtained in the latter part of the runs with the present apparatus. When the bath level dropped just below the copper section into the Kovar section (see Fig. 1), the liquid level in the viewing tube rose and a vapor block was found in the volume below. The conductivity of the Kovar is smaller than that of the copper so that the temperature of the liquid above the rouge was maintained closer to the temperature of the region outside of the copper. When the bath level dropped below the copper, there was a rise in temperature of the liquid above the rouge and a consequent flow of heat down into the compressed volume. By virtue of the thermomechanical effect, this resulted in a reduced pressure in the volume and the formation of a vapor block.

Although the accuracy of the measurement of compressibility here is not as good as the accuracy of the more conventional methods, it could be substantially improved by using a viewing tube of smaller crosssectional area. Also, this method could be "reversed" and used to measure the thermomechanical pressure in a closed system, that is, by taking a value for the compressibility from other measurements and measuring the compression produced, the pressure could be calculated.

The agreement of the compressibility value arrived at here with the values obtained by more conventional methods appears to justify the assumption that the full thermomechanical effect is operative across the rouge column in a closed system. It has been shown¹³ that, at a given temperature, there is a proportionality between the thermomechanical pressure and the heat current through narrow channels. It was of interest, therefore, to determine the heat transport properties of the helium II in the rouge channels.

III. HEAT TRANSFER EXPERIMENT

a. Apparatus

The apparatus used in the heat transport experiments was essentially the same in design as that used in the compressibility experiments. An additional feature was a glass vacuum jacket which enclosed the entire volume of the compression section from a point slightly above the top of the rouge column down to the bottom. This jacket was covered with aluminum foil in order to reduce heat radiation. In addition, two pieces of flat copper braid were soft soldered to the copper tube (see Fig. 1). These hung down in the helium bath and hence permitted better thermal contact between the bath and the copper tube when the helium bath. fell below the copper tube. A radiation shield, made of copper sheeting bent into a tube with two viewing slits cut along opposite sides of a diameter, was mounted above the adiabatic jacket and extended to a point slightly above the viewing section. The inner (compressed) volume of this apparatus was 10.6 cc and the rouge column was 4.43 cm high, containing 6.0 g of rouge.

b. Experimenta1 Procedure

The same procedure was followed here as in the compressibility measurements. The thermometers were calibrated, and then power was introduced into the system. The temperatures at both ends of the rouge column were read and the liquid level recorded.

As in the earlier experiments, mentioned in the Introduction, an average rouge channel size was determined by allowing air to pass through the rouge. (A finer rouge was used here than in the experiments mentioned in the Introduction.) If the flow of the

¹¹ W. H. Keesom and K. Clusius, Leiden Comm., No. 219f

 (1932) .
¹² K. R. Atkins and C. Chase, Proc. Roy. Soc. (London) A64, 826 (1931).

[»] J. F. Allen and J. Reekie, Proc. Cambridge Phil. Soc. 35, 114 (1939).

normal component of helium II through the rouge is of a Poiseuille type (see Sec. III. c.) and the air flow is also Poiseuille then the Poiseuille geometrical constant, K , measured by air flow may be used in Eq. (8) without any guesses as to the actual geometrical configuration in the rouge column. However, as will be mentioned in a subsequent paragraph, the air flow was not Poiseuille. Thus, not only for the estimation of an average channel size, but also for a calculation of K for the heat transport Eq. (8), it was necessary to make a guess as to the geometry of the channels in the rouge.

It was assumed that the rouge column is made up of N homogeneous cylindrical channels in parallel. This is undoubtedly not the case but the K calculated from this assumption is probably not far from the true value. For this assumption, the Poiseuille geometry factor, K , then is

$$
K = N\pi a^4/8L,\tag{5}
$$

where a =radius of the channels and L =length of the channels. The value of $Na⁴$ could be calculated from the (air) measured K . Using the density and mass of the rouge and the volume of the rouge column, Na^2 could be calculated. The combination of these yields the rouge channel radius, a. The value obtained for the channel size was 0.25 micron.

Since the mean free path for air at room temperature
of the same order of magnitude as the channel size,¹⁴ is of the same order of magnitude as the channel size, the channel size was recalculated assuming a Knudsentype flow. The value obtained for the channel size was 0.10 micron. From the criterion¹⁵ for flow conditions, it was concluded that the fiow was practically all Knudsentype and so the value 0.10 micron was used for the channel size.

c. Theory

The expression for the heat transport is

$$
\dot{Q} = \dot{V}_{n}\rho T s,\tag{6}
$$

where \dot{V}_n =volume rate of flow of the normal component of helium II in cm³/sec, ρ = density of the liquid in g/cm^3 , and $s=$ specific entropy of the liquid in cal/g K . If laminar flow is assumed, Poiseuille's equation holds and Eq. (6) may be written as

$$
\dot{Q} = \rho s T (K/\eta_n) \Delta P, \qquad (7)
$$

where η_n is the normal-component viscosity. When ρ_s of the Leiden¹⁸
=3X10⁴T^{s.6} cgs units, $X = (T/T_\lambda)^{5.6}$, and $T_\lambda = 2.186^{\circ}\text{K}$. The London and
are substituted, Eq. (7) becomes,¹⁶ are substituted, Eq. (7) becomes,

$$
\dot{Q} = 3 \times 10^4 T_{\lambda}^{5.6} (K/\eta_n) X T \Delta P. \tag{8}
$$

Equation (8) was used in the heat transfer calculations. The factor K , given by Eq. (5), was calculated

TABLE II. Heat transfer results.

Tb . °K (bottom)	ΔT , °K (T_b-T_t)	ΔP (dynes/cm ²)	Ò $-$ (exp) ΔT (mw/°K)	Q (exp) (mw)	O (theo) (mw)	Q (exp) Q (theo)
1.683	0.001	0.05×10^{4}	1.22×10^{2}	0.11	0.006	18.3
1.692	0.006	0.35	0.64	0.41	0.04	10.3
1.712	0.027	1.49	0.61	1.63	0.21	7.8
1.743	0.054	3.13	0.68	3.68	0.50	7.4
1.743	0.054	3.12	0.69	3.68	0.50	7.4
1.781	0.089	5.65	0.74	6.61	1.02	6.5
1.781	0.089	5.68	0.74	6.61	1.03	6.4
1.821	0.129	8.75	0.80	10.4	1.71	6.1
1.853	0.006	0.54	1.47	0.88	0.01	8.8
1.906	0.054	5.33	1.85	10.1	1.42	7.1
1.953	0.097	10.3	1.93	18.7	3.17	5.9
1.984	0.121	13.6	2.10	25.5	4.34	5.9
2.011	0.096	12.0	2.82	27.0	4.20	6.4
2.169	0.002	0.50	5.35	1.23	0.16	7.7
2.175	0.008	1.82	5.17	4.29	0.60	7.2
2.178	0.002	0.51	4.00	0.92	0.18	5.1

using $a=0.10$ micron. The pressure difference, ΔP , between the ends of the rouge column was calculated by substituting the measured temperature differences in the thermomechanical Eq. (3). The values for the normal-component viscosity, η_n , were obtained from the data of Hollis-Hallet.¹⁷

d. Results and Discussion

The results of the experimental and theoretical determinations of the heat transfer are given in Table II. The first column in the table gives the bath temperature; the second column gives the temperature difference between the top and bottom of the rouge column; in the third is the calculated thermomechanical pressure; in the fourth is the ratio of the observed heat current to the observed temperature difference; the fifth column gives the observed heat current; in the sixth column are the values of the heat current calculated from Eq. (8); and in the last column are the ratios of the experimental heat currents (column 5) to the theoretical heat currents (column 6).

The first eight entries are all from one run, taken at the same bath temperature. The last eight were taken at random from the data of five other runs.

The results in column 7 show that the calculated values of the heat current are smaller than the experimental values by factors ranging from 5 to 18. This disagreement, however, is smaller than the disagreement observed by London and Zilsel¹⁶ in their analysis of the Leiden¹⁸ data for a comparable channel size. The London and Zilsel analysis gave discrepancies as high as a factor of 260 for the region of small temperature differences and small channel sizes. It should be kept in mind that, because of the different geometry used, the comparison of the experiments reported here with those of the Leiden group can only be made through the theory. The experimental difhculties in

¹⁴ We thank Professor Lars Onsager of Yale University for pointing this out to us.

pointing this out to us.
¹⁵ S. Dushman, *Scientific Foundations of Vacuum Technique*
(John Wiley and Sons, Inc., New York, 1949).
¹⁶ This is essentially the equation arrived at by F. London and
P. R. Zilsel [Phys. Rev.

¹⁷ A. C. Hollis-Hallet, Proc. Roy. Soc. (London) A210, 404 (1952).

[»] W. H. Keesom and G. Duyckaerts, Physica 13, ¹⁵³ (1947); also: J. H. Mellink, Physica 13, ¹⁸⁰ (1947); L. Meyer and J. H. Mellink, Physics 13, 197 (1947).

determining the channel size both in these experiments and those of Leiden might account for the difference in the comparisons to theory. In both sets of experiments, however, the observed heat currents are larger than
predicted.¹⁹ predicted.

Column 4 of Table II shows that for the first set of readings (those for a single run) the Q_{exp} are very nearly proportional to ΔT , as predicted by the linear theory. It should also be noted that there is an apparent dependence of \dot{Q} on the pressure difference. There is a decrease in $\dot{Q}_{\rm exp}/\dot{Q}_{\rm theo}$ of a factor of about 3 for an increase in ΔP of a factor of about 200. The reasonableness of the compressibility results discussed in Sec. II would indicate that this dependence is on ΔP rather than of ΔP on the temperatures (top and bottom).²⁰ than of ΔP on the temperatures (top and bottom).²⁰

It is also of interest to compare the values of $\dot{Q}/\Delta T$ (column 4) as a function of temperature with a similar quantity which was plotted by Keesom and Duyckaerts. '8 They showed that the heat current density at constant temperature-difference plotted as a function of the temperature exhibited a maximum below the λ point. The maximum decreased and moved closer to the λ point with decreasing channel size. The smallest channel size which showed a maximum on this plot was 9.3 microns for which the maximum occurred at $2.0\textdegree K$. The linear theory does not predict a falloff of the curve to a small value at the λ point.

From an examination of column 4, Table II, it can be seen that the values of $\dot{Q}/\Delta T$ increases with increasing temperature (except for the first and last entries), and no real maximum is observed. From an extrapolation of the temperatures of the peaks of the curves obtained by Keesom and Duyckaerts for different channel sizes, it was found that the peak for the present data would be extremely close to the λ point. In an effort to locate the maximum, measurements were made near the λ point.

While making such measurements, it was found that the liquid level in the viewing section at zero power input continued to fall after the bath temperature had been increased from a previous value and then held

constant. A decrease in the liquid level was expected, since by raising the temperature (while below the λ point) the density of the liquid is increased. The drift in liquid level could be interpreted as being a long equilibrium situation usually associated with poor heat transfer and that the heat conductivity peak below the λ point had been located. On the other hand, if the temperature below the rouge column was above the λ point, all one would be observing would be the poor heat transfer above the λ point. However, the liquid level, as mentioned, continued to decrease rather than increase, indicating that the temperature was not above the λ point. In contradiction to this was the measurement of the temperature of the liquid below the rouge which gave a value just above the λ point. However, a slight error in the absolute calibration of the thermometer would permit the possibility of an actual temperature just below the λ point.

An effort was made to account for the fall in level by calculating the variation in density of the liquid above the rouge due to a possible variation in the bath temperature. This calculation yielded a level decrease of only 0.002 cm, whereas the observed change was 0.29 cm. Another calculation was made on the assumption that the temperature of the liquid below the rouge was above the λ point but that the full thermomechanical effect was operative across the rouge, compressing the liquid below. This yielded a value of 0.07 cm for a possible level decrease, still too small to account for the observed change. Thus, it was concluded that the level decrease indicated that the liquid below the rouge was still below the λ point.

It is a possibility that the observed drift $2¹$ of the liquid level indicates a region of poor heat transfer below the λ point. On the basis of the measurements that were made, the peak in the heat transfer would occur in a region of approximately 1 to 2 millidegrees below the λ point.

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 19 C. J. Gorter and J. H. Mellink [Physica 15, 285 (1949)] suggested that the discrepancy between the Leiden data for small slits (reference 18) and the theory (reference 16) is caused by a mean free path effect. They estimated that the mean free path is of the order of 1 micron. However, later results LWinkle, Van Groenor, and Gorter, Physica 21, 345 (1955)] indicate that there is no mean free path effect down to about 1 micron. Our estimate of a mean free path is (roughly) 0.07 micron, which is suggestively,

at least, near to twice the film thickness (0.02 micron).
²⁰ The recent Leiden results [P. Winkle *et al.*¹⁹] show directly that for a slit width of 6 microns there is a falloff in the heat current as the thermomechanical pressure increases.

²¹ K. R. Atkins has informed us that a similar effect was observed, coming up in temperature from below the λ point, during measurements on the coefficient of expansion of helium II. [See K. R. Atkins and Edwards, Phys. Rev. 97, 1429 (1955).]