

The Blocked Capillary Method for Determining Melting Pressures: The Melting Curve of Helium from 1.5° to 4°K

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(Received June 30, 1952)

The blocked capillary technique has been used to check previous piston displacement values for the helium melting pressure curve in the range from 1.5° to 4°K. In the course of these experiments, details of which are given, it was shown that melting pressure as given by the capillary blocking method are completely reliable, and can be very accurate. Variations of the capillary size or pressure difference across the capillary had no evident effect.

The behavior of the helium melting curve in the vicinity of the upper "triple point" is discussed in some detail. An analysis of the data indicates that it is in qualitative agreement with the assumption that the λ -transformation is of the second order.

I. INTRODUCTION

THE mechanical blocking of a fine capillary by the freezing of a liquid was first used by Kamerlingh Onnes at Leiden in his work on the melting pressure curve of hydrogen.^{1,2} The method has been used rather extensively, since in the low temperature region it requires a simple experimental set-up and relatively small quantities of the substance being investigated. The accuracy of this technique, as compared with others, has never been systematically studied, and discrepancies in the work of several experimentalists can be attributed either to impurities in the substance used, or to an effect which is inherent in the method. P. W. Bridgman has pointed out that the application of shearing stresses can lead to a lowering of the melting point of a solid, and that these stresses must exist across the block in the capillary. In some experiments on carbon dioxide, Michels reduced this pressure difference to less than an atmosphere, and obtained good agreement with melting pressures obtained by

Bridgman, who used the so-called "piston displacement" technique.³

In previously reported work, a modified form of the piston displacement method was used by the author to obtain melting pressures, latent heats of melting, and volume changes on melting for helium in the range from 1.6° to 4°K.⁴ The melting pressure data were believed to be accurate to better than one-tenth of one percent, although the data as a whole were thermodynamically inconsistent in the region of the upper "triple point" on the helium phase diagram. This made it advisable for the melting pressure curve to be redetermined using an independent method, and the blocked capillary technique was chosen as the most convenient, and most likely to give accurate results. The apparatus and procedure which were found to be capable of giving reproducible capillary blocking data, in agreement with the piston displacement data, are described below. An analysis of the composite melting pressure data for helium is also given, together with quite reliable values for the entropy differences between liquid and solid helium along the melting curve.

II. EXPERIMENTAL DETAILS

Figure 1 shows the simplest version of a capillary blocking set-up.² The coldest point of the capillary is held at a constant temperature T , while the pressure on the liquid in the capillary is slowly increased. P_1 and P_2 will remain equal and increase together as long as the capillary is below the freezing pressure corresponding to T . Above the freezing pressure, there will no longer be continuity between the two gauges, and P_2 will give a constant reading (roughly the freezing pressure) as P_1 is increased.

Freezing points were found to be quite difficult to work with, since supercooling was often observed. A second procedure, which turned out to be more reliable, was used to measure melting points. The temperature of the loop T , was slowly lowered until the capillary was

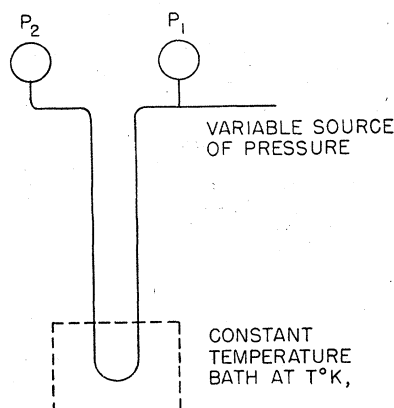


FIG. 1. A simple capillary blocking set-up.

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¹ H. Kamerlingh Onnes and W. van Gulik, *Leiden Comm.* 184a (1926).

² W. H. Keesom, *Helium* (Elsevier, Amsterdam, 1942), p. 180.

³ For a discussion, see P. W. Bridgman, *Revs. Modern Phys.* 18, 28 (1946).

⁴ C. A. Swenson, *Phys. Rev.* 86, 870 (1952).

found to be blocked. T was then held constant as P_1 was gradually decreased. P_2 remained unchanged until the solid blocking the capillary was melted at the value of P_1 corresponding to the melting pressure at T . At this melting pressure, P_2 dropped rapidly, and approached P_1 . It was necessary that P_2 be greater than P_1 , since melting occurred initially at the lower pressure. It also had to be established that the melting pressures observed for a given temperature were independent of the pressure differences across the capillary, $\Delta P = P_2 - P_1$.

The essential features of the apparatus used, then, were a source of variable pressure, a cryostat, in which the temperature of the loop was accurately known, and a means for indicating the pressure difference across the capillary when it was blocked. P_1 , the source of variable pressure, was the same pressure balance which was used in the piston displacement experiment, so that the absolute calibration of the balance was not of importance in comparing the two sets of data. Similar vapor pressure thermometers were also used in the two experiments, so that difficulties would not arise from the comparison of the temperatures.

The final design of the cryostat which was found to be satisfactory for this work is sketched in Fig. 2. It was constructed entirely from copper, and mounted in a vacuum jacket for thermal isolation. The base of the cryostat was immersed in a liquid helium bath, the temperature of which could be varied from 4° to about 1.4°K , and its top was held at a temperature near 4° . This insured that there was a small but definite temperature gradient along its length.

The capillary itself was suspended in the liquid of the vapor pressure thermometer (of about 1 cc capacity), so that the temperature of the capillary could be read accurately on a mercury or oil manometer. The ends of the capillary were connected to room temperature by $\frac{1}{2}$ mm o.d. stainless steel tubing, while the vapor pressure thermometer was connected to the manometer by a copper nickel tube, 1.5 mm i.d.

The lower heater, No. 1, was used to vary the temperature of the vapor pressure thermometer, and, hence, the temperature of the capillary, by sending a heat flow through the copper connection to the outer bath. Thus, if the outer bath was held at 1.5° , the capillary temperature could be raised as high as 1.9° by applying current to the heater. A fine control on this heater current was used to regulate the temperature of the capillary, and to compensate for slight changes in the bath temperature. Temperature fluctuations were kept to less than 0.0005 degree.

The upper heater was necessary to give reproducible results. It was found that if the temperature gradient (or heat flow) down the capillary was too small, a considerable length of time was needed for the heat of fusion to be supplied to the solid in the capillary. Checks were always made to insure that the melting point found was not a function of the heat input, or

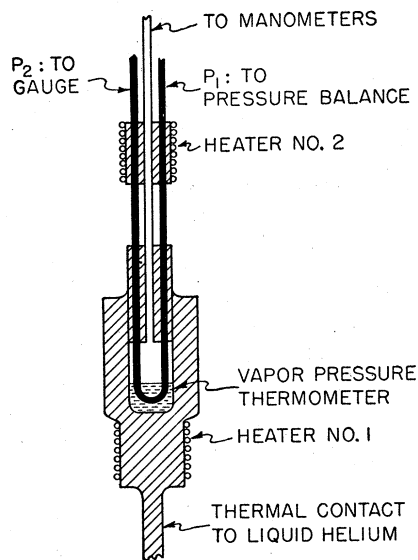


FIG. 2. The final version of the capillary blocking cryostat used.

of the rate of change of pressure. A minimum input of about 2 milliwatts seemed necessary to insure reproducibility. Below the λ -point, however, the thermal conductivity of helium II was found to be large enough so that this heater could be dispensed with.

With the experiment as set up finally, the requirements on the gauge for measuring P_2 were not very stringent. Any device that would show the disappearance of the pressure difference across the block would be satisfactory. In anticipation of other work, it was decided that the gauge would have to have a very small dead volume, and would have to be sensitive to variations in pressure of at least 0.1 atmosphere over the pressure range from 20 to 150 atmospheres. Differential monometers are usually not very rugged, and are easily blown out by careless handling. A Bourdon gauge is not quite sensitive enough, and is likely to stick within the requirements above. The final solution to the problem was simple and sturdy, and extremely sensitive.

A sketch of the gauge used is shown in Fig. 3. A steel plate, 2 mm thick, was fastened by means of six steel bolts to a brass block by a steel ring, so that a diaphragm 2 cm in diameter was formed. A pressure inlet of small diameter was made in the block under the center of the diaphragm. The constants of the diaphragm were chosen so as to give its center a rise of about 0.1 mm when a pressure of 120 atmospheres was applied to it. This rise was magnified by means of the 100:1 lever arrangement shown, and finally detected by an electrical contact between the micrometer and the end of the lever. A thyatron detection circuit of high internal resistance, attached to an indicator light, made it possible to reproduce settings to about 0.005 mm on the enlarged micrometer head, and changes of this order of magnitude were easily detected.

This meant that changes in pressure of less than 0.1 atmosphere could be observed over the usable range. It is interesting to note that the change in the height of the diaphragm caused by the change in pressure of 0.1 atmosphere is less than one-fifth of a wavelength of visible light.

The lever arrangement was first suggested by Professor K. T. Bainbridge, and was based on an original idea by Dr. D. W. Dye.⁵ The center of the diaphragm made contact with a ball bearing by means of a post. The center of this ball bearing was 1 mm in front of a line through the centers of two outer ball bearings, both of which rested on a hard (glass) surface. The distance between these bearings and the electrical contact on the end of the lever was roughly 10 cm. The springs at the outer ends of the center bar were to insure that the lever pivoted on the two outer bearings, and was also in stable equilibrium with the center bearing. The counterbalance was used to supplement the springs. The combination of a mechanical lever arm, micrometer, and electrical contact, rather than the original optical arm, led to a much more convenient piece of apparatus for this purpose.

This gauge was very sensitive, and extremely rugged. Its chief usefulness comes when it can be calibrated at one point (as when the capillary was free) and used to measure changes in pressure from this value. In the absence of vibration, it was quite stable, and retained a setting during leak testing, for instance, within ± 0.005 mm on the scale for twenty-four hours or longer. Because of its sensitivity, it was not reproducible to this amount for large pressure differences (twenty or thirty atmospheres), although the drift or hysteresis

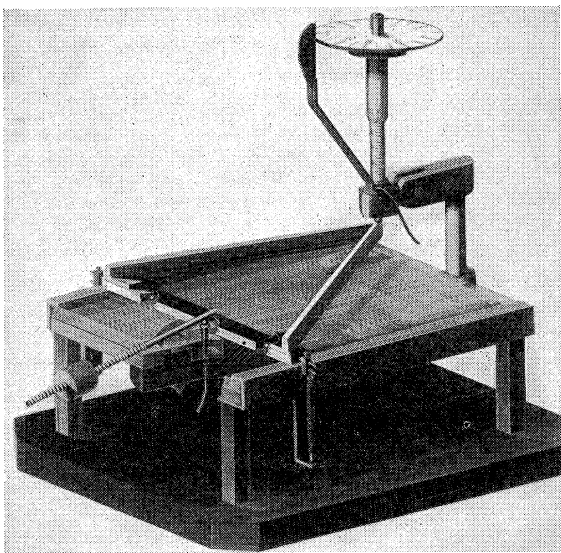


FIG. 3. The pressure gauge used to measure P_2 . See the text for a description and details.

⁵ F. H. Rolt, *Gauges and Fine Measurements* (MacMillan and Company, Ltd., London, 1929), p. 339.

was not large. The chief source of error was most likely very small irregularities either on the bearings or on the glass.

The pressure difference across the capillary ΔP was varied by altering the temperature of the blocked side of the capillary after it had emerged from the cryostat. A loop of this capillary was immersed in liquid nitrogen while the capillary was free. After the block was formed, P_2 could be increased at will by varying the length of the capillary remaining in the liquid nitrogen. This procedure was used to verify that, during a run, P_2 was always greater than P_1 . It was also possible to show that the melting point at a given temperature was independent of the pressure difference across the capillary. This appeared to hold, in general, even for a pressure difference of 25 atmospheres at a melting pressure of 50 atmospheres, so no precautions were taken to insure that ΔP was any special value in the following experiments. It was usually of the order of five or six atmospheres.

The experimental procedure may now be summarized briefly. With about two milliwatts of power being supplied to the top heater, and a constant pressure in the capillary, the lower heater current was slowly decreased until there was no longer contact between the pressure balance and the pressure gauge. ΔP was increased to about five atmospheres, and the temperature of the vapor pressure thermometer regulated as closely as possible. Weights were removed from the pressure balance at the rate of about 0.01 atmosphere/minute until the pressure gauge changed suddenly, showing that the block had been broken. At the same time, a slight increase was usually noted in the vapor pressure, resulting from the sudden passage of the "hot" gas through the thermometer. The melting pressure was calculated from the constants of the balance and barometric pressure, and the temperature from the 1949 Cambridge vapor pressure scale, using a conversion factor of 15.62 between the density of mercury and Apiezon A oil.

III. RESULTS

The apparatus was first tested by making a determination of the melting curve of nitrogen, using the two techniques. Nitrogen was chosen as a convenient substance to work with, since it could be obtained relatively pure, and its triple point was easily reached experimentally. Absolute temperatures were calculated from the work of Keesom and Bijl.⁶ It was found that the piston displacement and capillary blocking data were indistinguishable between 5 and 140 atmospheres, and could be represented by the equation,

$$P + 882.5 = 0.0024703T^{3.084305} \text{ atmos.}$$

This curve is parallel to the earlier curve of Keesom and Lissman,⁷ although it is displaced downwards by roughly $1\frac{1}{2}$ atmospheres.

⁶ W. H. Keesom and A. Bijl, *Physica* 4, 305 (1937).

⁷ W. H. Keesom and J. H. C. Lissman, *Leiden Comm.* 232b (1934).

A preliminary check with helium showed that the two methods agreed between 2.5° and 4°K to within a few tenths of an atmosphere, so the remainder of the work reported here was concentrated in the region of the upper "triple point" on the helium phase diagram. Previous work, using the piston displacement method, had given consistent results which are shown as the solid curve in Fig. 4.⁴ The melting pressures obtained by using the capillary blocking technique are shown as points, and it can be seen that the agreement between the two methods is quite satisfactory.

The effect of capillary size was also investigated, since it is possible, especially in liquid helium II, that size effects (small ratios of volume to area) will become important. Three separate capillaries are represented in the data of Fig. 4 (not indicated individually), with approximately equal numbers of the points due to each. The capillaries were, in order of size, (a) copper, 0.026-inch i.d., volume/area=0.165 mm; (b) copper, 0.013-inch i.d., with a 0.009 inch steel wire down its center, volume/area=0.025 mm; (c) a copper tube, drawn down over sixty-six 0.005-inch piano wires, volume/area = 0.002 mm.⁸ No size effect was found, even in helium II, which is not surprising, since the spaces in the finest capillary were of the order of one hundred helium II film thicknesses. It would be of great interest to go to still smaller capillaries, but the time taken to obtain equilibrium across the finest capillary used was of the order of a minute, and this would increase considerably if a decrease of another factor of ten were attempted in the ratio of volume to area.

It was quite necessary to make the capillaries from copper because of its relatively high thermal conductivity. Initially, when stainless steel and copper-nickel capillaries were used, deviations from the piston displacement curve were observed below 1.764°. These deviations could be roughly predicted as the result of the fact that the thermal conductivity of liquid helium II under pressure was so much greater than that of the capillary material that appreciable temperature drops (of the order of a few thousandths of a degree) could appear across the walls of the capillary. These deviations, always giving melting pressures that were too high, were consistent, and seemed to disappear above 1.764°. If this effect were the result of superfluid helium II, then the temperature of the upper "triple point" given in reference 4 (1.743°) could not be correct. To check this, a more accurate determination of the λ point-pressure curve was made.

The method used was essentially a modification of that used by Schmidt and Keesom,⁹ and more recently, Long and Meyer.¹⁰ Two copper vapor pressure thermometers were connected to opposite sides of a differen-

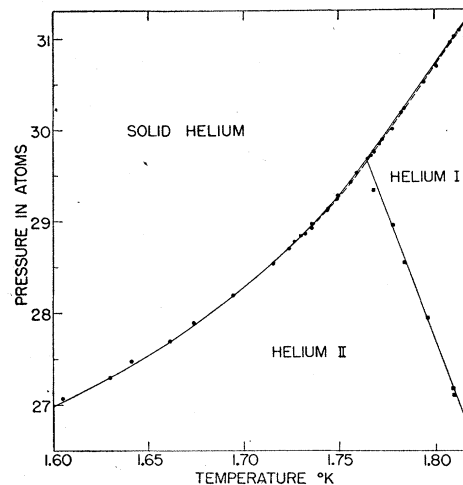


FIG. 4. The melting pressure data in the vicinity of the upper "triple point." The individual points represent capillary blocking data, while the solid line represents previous piston displacement data.

tial oil manometer, and were joined by a 3-inch copper capillary (0.026 inch i.d.), containing liquid helium under pressure, and soldered to the sides of the thermometers. The temperature of the upper thermometer, as measured by means of an absolute oil manometer, was held constant while the bottom thermometer was kept at a slightly higher temperature by means of a heater. The pressure on the helium in the capillary was then slowly decreased by removing weights from the pressure balance until at the λ -point, the temperature difference between the gas thermometers, as shown by the differential manometer, tended to decrease. The accuracy of this method is limited because the thermal conductivity of helium II has a finite temperature coefficient at the λ -point,¹¹ but the data of Fig. 5 (tabulated in Table I) are probably accurate to $\pm 0.002^\circ$. The coincidence of the upper λ -point as obtained by an extrapolation of these data (Fig. 4), and as deduced from the conductivity effect, seems to verify this assumption. The coordinates of this "triple point" can now be given with considerable assurance as

$$P_\lambda = 29.64(\pm 0.03) \text{ atmospheres,}$$

$$T_\lambda = 1.764(\pm 0.003)^\circ\text{K.}$$

This is in better agreement with the original data of Keesom and Clusius.¹²

The λ point-pressure curve as determined earlier and the kink in the piston displacement melting pressure curve at 1.743° seem to be closely connected.⁴ The capillary blocking data lie slightly below this curve above 1.743°, and seem to indicate that the kink does not actually exist. It is most likely that when the

⁸ For a discussion of the method used for the calibration of this capillary, see J. F. Allen and A. D. Misener, Proc. Roy. Soc. (London) A172, 467 (1939).

⁹ G. Schmidt and W. H. Keesom, Physica 4, 963 (1937).

¹⁰ E. A. Long and L. Meyer, Phys. Rev. 83, 860 (1951).

¹¹ J. F. Allen and E. Ganz, Proc. Roy. Soc. (London) A171, (1939).

¹² See reference 2, p. 225.

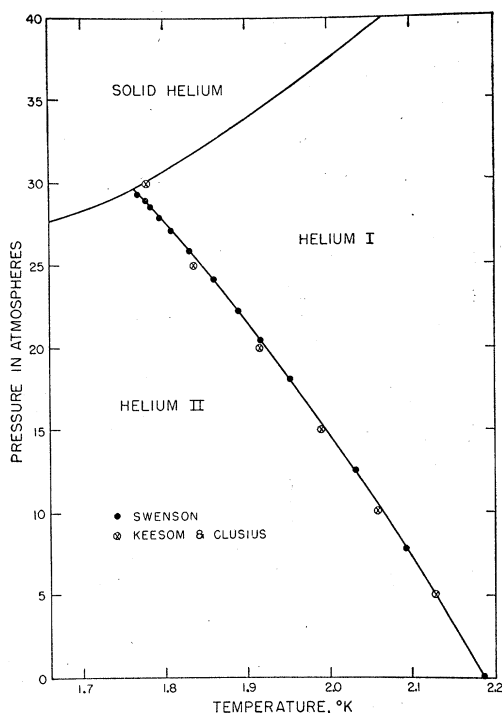


Fig. 5. The λ point-pressure curve.

thermal conductivity of the helium II became sufficiently great, a small temperature inhomogeneity inside the piston displacement cryostat was evened out. The crude λ point-pressure technique used previously could, presumably, detect only when the thermal conductivity of the helium was "sufficiently large," and not when it began to change rapidly.

The deviation of the capillary blocking points above 1.75° is slightly outside experimental error (about 0.001°). It is consistent enough, however, under various conditions, so that a fair amount of weight should be placed on it when deciding on the final smoothed melting pressure curve. The curve finally decided on is indicated by the dashed line in Fig. 4.

There also seems to be a tendency below 1.7° for the capillary blocking points to deviate towards higher melting pressures than those given by the piston displacement curve. These deviations are towards better agreement with earlier data obtained by Keesom and Miss Keesom¹³ and the author.¹⁴ It is perhaps significant that the data from all sources are in quantitative agreement from 1.75° to 1.8° , so that discrepancies at lower temperatures are most likely the result of differences in experimental technique. The maximum deviation of 0.15 atmosphere at 1.6° is between the present work and the earlier capillary blocking data obtained by the author.¹⁴ The set-up which was used in that work was such that the thermal conductivity

effect, which was mentioned previously, may be responsible for the discrepancy. The agreement between the various sources seems to become better as the temperature is lowered.

The consolidation of these differing but seemingly accurate sets of data into a reliable melting pressure curve is quite difficult. The present capillary blocking data confirm the piston displacement experiments so well that a smoothed curve incorporating the two sets of data was taken as the most likely to be correct. This curve is given in column 2, Table II, for temperatures between 1.4° and 4°K . It is not likely to be in error by more than two-tenths of one percent at any given point, and is internally consistent to a greater accuracy than that near 1.75° .

The slope of the melting pressure curve, $(dP/dT)_M$, is actually of more interest than the curve itself, since it can be combined with measurements of the change in molar volume on melting $((\Delta V)_M = V_L - V_S)$ to obtain the entropy difference between the solid and the liquid along the melting curve. The derivatives were obtained by using the first and second differences of the smoothed curve at small temperature intervals.¹⁵ This procedure is straightforward as long as the second differences are small, but between 1.7° and 1.8° the slope of the melting pressure curve is changing very rapidly. A certain amount of arbitrariness is introduced in the way that the smoothed curve is drawn through the experimental points, and this is reflected in the $(dP/dT)_M$ curve. An obvious criterion which must be satisfied by any $(dP/dT)_M$ curve is that the area under the curve between any two temperatures must equal the experimentally observed difference in melting pressure.

The final smoothed values of $(dP/dT)_M$ are summarized in column 3 of Table II, and the data in the vicinity of the λ -point are plotted in Fig. 6. The shape of this curve resembles very closely the expected curve for a second-order transformation, with a discontinuity in its slope occurring at the upper "triple point." This discontinuity in $(d^2P/dT^2)_M$ is finite, and is of the order of -500 atmospheres/deg². It can also be calculated from other melting curve data by using the Ehrenfest relations for a second-order phase

TABLE I. Smoothed values of the λ point-pressure curve.

T_λ ($^\circ\text{K}$)	P_λ (atoms)
2.186	0.02
2.15	3.18
2.10	7.26
2.05	11.10
2.00	14.72
1.95	18.18
1.90	21.50
1.85	24.62
1.80	27.62
1.764	29.64

¹³ W. H. Keesom and A. P. Keesom, *Leiden Comm.* **224e** (1933).

¹⁴ C. A. Swenson, *Phys. Rev.* **79**, 626 (1950).

¹⁵ E. T. Whittaker and G. Robinson, *The Calculus of Observations* (Blackie and Son, Ltd., London, 1932), p. 62.

transformation.⁴ If the new values for $(dP/dT)_\lambda = -54.5$ atmospheres/deg and $(dP/dT)_M = 27.5$ atmospheres/deg are used, $\Delta_{I,II}(d^2P/dT^2)_M$ can be computed as -220 atmospheres/deg².

The argument between the two figures is such that no real decision can be made about the applicability of the Ehrenfest relations in this case. An error of a factor of two might be expected from the experimental data, since the second derivative of the melting pressure curve has been taken, and it is varying quite rapidly. The slope of the $(\Delta V)_M$ curve at the λ -point also is used, and could be in error in spite of a seeming consistency in the data.

One possibility exists which is not usually considered. If the specific heat, thermal expansion, and compressibility have infinite discontinuities at the λ -point, then $(d\Delta S/dT)_M$ would be infinite also. Previous data are so crude (except for the specific heat data) that this could have been overlooked. The ordinary equations which govern the second-order phase change would be indeterminate, with the λ point-pressure curve still existing. The values for the entropy difference between liquid and solid helium have been calculated, using previous ΔV values, and are tabulated in column 5 of Table II. The slope of the entropy difference curve in helium II at 1.764° is rather large (about 16 cal/mol-deg), but not infinite. These calculations involve the $(\Delta V)_M$ curve, however, and are, therefore, open to suspicion.

It is possible to calculate the slope of this curve directly from the observed values of $\Delta_{I,II}(d^2P/dT^2)$, $(dP/dT)_\lambda$, $(dP/dT)_M$, $(\Delta V)_M$, and the temperature at the upper λ point, using the Ehrenfest relations. The

TABLE II. Smoothed values for the melting pressure, P , the slope of the melting pressure curve, $(dP/dT)_M$, the molar change in volume on melting, $(\Delta V)_M = V_L - V_S$, and the molar entropy difference between liquid and solid helium, $(\Delta S)_M = S_L - S_S$.

T (°K)	P (atmos)	$(dP/dT)_M$ (atmos/deg)	$(\Delta V)_M^4$ (cc/mol-deg)	$(\Delta S)_M$ (cal/mol-deg)
1.40	(25.81)	4.2	2.06	0.21
1.50	26.22	6.3	2.00	0.30 ⁶
1.55	26.56	7.7	1.95	0.36 ⁶
1.60	26.99	9.6	1.89	0.44
1.65	27.54	12.3 ⁵	1.76	0.52 ⁵
1.70	28.27	16.3 ⁵	1.60	0.63 ⁵
1.72	28.63	18.2 ⁵	1.54	0.68
1.74	29.03	20.7	1.49	0.74 ⁵
1.75	29.26 ⁶	22.4	1.48	0.80 ⁶
1.764	29.63	27.5	1.47	0.98
1.80	30.70	29.2	1.45	1.02 ⁵
1.85	32.21	31.0	1.44	1.08
1.90	33.80	32.2	1.43	1.11 ⁵
1.95	35.45	33.2	1.42	1.14
2.00	37.08	34.3	1.40	1.17
2.10	40.56	35.8	1.38	1.20
2.20	44.21	37.2	1.35	1.21 ⁵
2.30	47.98	38.6	1.32 ⁵	1.24
2.50	56.3	41.0	1.28	1.27
2.75	66.7	43.6	1.22 ⁵	1.30
3.00	78.1	46.2	1.19 ⁵	1.34
3.50	102.0	51.0	1.10	1.36
4.00	128.6	56.0	1.03	1.40

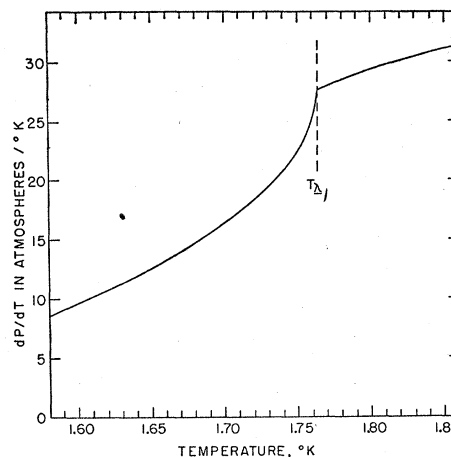


FIG. 6. The slope of the melting pressure curve $[(dP/dT)_M]$ in the vicinity of the upper "triple point."

agreement is satisfactory with the above figure. What is perhaps of more interest is to calculate the magnitude of the discontinuity in the specific heat which one would find in the vicinity of the upper triple point, if these data are assumed. Using the relations which were mentioned in the previous paper,⁴ the following expression for the specific heat discontinuity can be derived:

$$\Delta C_p = C_{pI} - C_{pII} = T(\Delta V)_M \Delta_{I,II}(d^2P/dT^2)_M / [1 - R(2 + R)],$$

where $R = (dP/dT)_M / (dP/dT)_\lambda$. If

$$(\Delta V)_M = 1.48 \text{ cc/mol}, \quad T_\lambda = 1.764^\circ,$$

$$(dP/dT)_M = 27.5 \text{ atmospheres/deg},$$

$$(dP/dT)_\lambda = -54.5 \text{ atmospheres/deg},$$

and

$$\Delta_{I,II}(d^2P/dT^2)_M = -500 \text{ atmospheres/deg}^2$$

are substituted into this expression, the calculated value for ΔC_p is -18 cal/mol-deg, or -4.5 cal/g-deg. This is reasonable, and of the same order of magnitude as the specific heat anomaly found in liquid helium under its saturated vapor pressure.

The final conclusions to be drawn from this melting pressure work are not clear-cut. The blocked capillary technique seems to be as capable of giving accurate results as the piston displacement method, and the two sets of data are in excellent agreement in the vicinity of the upper "triple point" for helium. The analysis of the results is more difficult, and only indications can be drawn from the experimental data. It is quite definite that the slope of the melting pressure curve is continuous at the λ -point, and that the data are in qualitative agreement with the Ehrenfest relations. If the behavior of the λ -transformation is the same at the lower "triple point" as at the upper,

then these data would lead one to expect that the discontinuities in the specific heat and thermal expansion would be finite at the normal λ -point. This is the present indication from direct measurements.¹⁶

¹⁶ See reference 2, pp. 206 and 245.

This work was made possible in part by a grant from the Harvard Foundation for Advanced Study and Research. I wish to thank Mr. G. Bjorklund and Mr. E. Wilkie for their assistance in constructing the apparatus used.

Z-Dependence of the Pair Production Cross Section at 1.33 and 2.62 Mev

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(Received October 21, 1952)

The Z -dependence of the pair production cross section has been measured at 1.33 Mev for Al, Cu, Sn, and Pb, and at 2.62 Mev for Be, C, Al, Cu, Sn, and Pb. A target is used which is thick for the positrons produced in it but thin for the incident gamma ray beam. The positrons stop in the target and are detected by observing their annihilation radiation with two NaI(Tl) scintillation counters in coincidence. At both energies the Z -dependence of the cross section is best represented by an equation of the form $aZ^2 + bZ^4$. If it is assumed that the Born approximation calculations of the cross section give the correct value in the limit of low Z , then in lead at 2.62 Mev the measured cross section is 23 percent higher than the value calculated from the Born approximation, and 104 percent higher at 1.33 Mev. All these results are in good agreement with the exact numerical calculations of Jaeger and Hulme.

The present measurement shows that near threshold the pair production cross section in lead is considerably higher than the Born approximation calculation, whereas at energies between 17 Mev and 280 Mev the value has been measured to be 10 percent to 15 percent lower than that calculated from the Born approximation. The crossover appears to occur at around six Mev.

I. INTRODUCTION

IN the years since the discovery of the production of electron-positron pairs by gamma-rays, this process has frequently been studied using the 2.62 Mev gamma-ray from ThC'' .¹ These experiments have served to establish the nature of the process and to give some information about the distribution of the particles in energy and angle. But since these measurements were based on at most a few hundred cloud-chamber pictures with one or perhaps two target materials, there is no accurate data on the total cross section for pair production, and until recently nothing on the Z -dependence of the total cross section at energies below 3 Mev.

The theory of pair production is summarized in Heitler.² It predicts that the total cross section for nuclear pair production will vary as Z^2 . At the energies involved here, it is not necessary to include the effect of screening of the nuclear Coulomb field by the atomic electrons. The theory is calculated using the Born approximation, which is expected to fail for high Z and low gamma-ray energy. Jaeger and Hulme have made a number of exact numerical calculations of the pair

production cross section.³⁻⁵ They find that in lead the exact cross section is 25 percent higher than the Born approximation value at a gamma-ray energy of $5 mc^2$ and about a factor of two higher at $3 mc^2$. They also find⁴ that at $3 mc^2$ the Z -dependence of the cross section is best represented by an equation of the form $aZ^2 + bZ^4$. The object of the present experiment is to check quantitatively these predicted deviations from the Born approximation theory, using gamma-rays of 2.62 Mev and 1.33 Mev.

While this experiment was in progress, indications of the predicted deviations from the Born approximation theory were obtained by Cleland, Townsend, and Hughes⁶ using the 2.76-Mev gamma-ray from Mg^{24} . Hahn, Baldinger, and Huber⁷ have recently reported an experiment similar to this one. Their results are in general agreement with ours and with the theoretical predictions.

General Considerations

At the gamma-ray energies used in this experiment, the pair spectrometer technique, which has been used at 17.6, 88, and 280 Mev⁸⁻¹⁰ to study pair production,

³ J. C. Jaeger and H. R. Hulme, Proc. Roy. Soc. (London) **A153**, 443 (1936).

⁴ J. C. Jaeger, Nature **137**, 781 (1936).

⁵ J. C. Jaeger, Nature **148**, 86 (1941).

⁶ Cleland, Townsend, and Hughes, Phys. Rev. **84**, 298 (1951).

⁷ Hahn, Baldinger, and Huber, Helv. Phys. Acta **25**, 505 (1952).

⁸ R. L. Walker, Phys. Rev. **76**, 1440 (1949).

⁹ J. L. Lawson, Phys. Rev. **75**, 433 (1949).

¹⁰ DeWire, Ashkin, and Beach, Phys. Rev. **83**, 505 (1951).

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¹ For references to earlier work see: W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, London, 1944), second edition, p. 201; K. H. Spring, *Photons and Electrons* (John Wiley and Sons, New York, 1950), Chap. V.

² See Heitler, reference 1, Chap. 4.

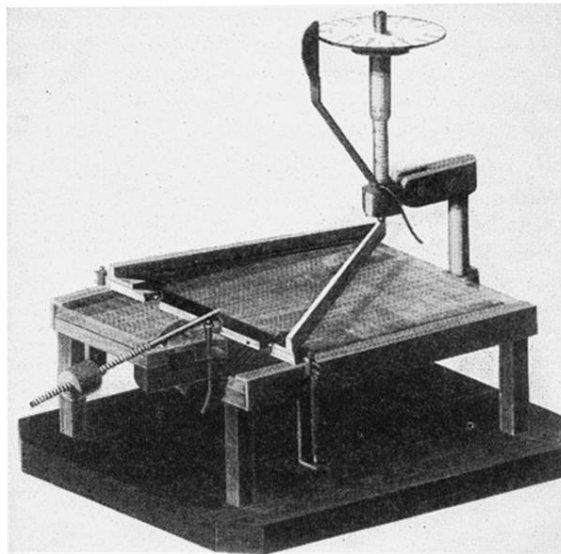


FIG. 3. The pressure gauge used to measure P_2 . See the text for a description and details.