

Thermal Conductivity of Hexagonal Close-Packed Solid Helium Four at High Densities*

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Thermal conductivity measurements between 1.1°K and 7°K have been made on solid helium samples grown under constant pressures from 185 atm to 1050 atm. At high temperatures the phonon mean free path \bar{l} for the highest conductivity samples at six different densities is in good agreement with the expression, $\bar{l} = A \exp(\theta_D/bT)$, where θ_D is the Debye temperature at $T = 0^\circ\text{K}$, $A = 2.5 \times 10^{-8}$ cm, $b = 2.13$. Several samples grown from gas repurified by an adsorption trap exhibited mean free paths in agreement with this expression over almost four orders of magnitude. Slightly impure samples showed a considerable attenuation in the peak thermal conductivity at pressures above 320 atm. There was some evidence for anisotropy of the thermal conductivity in the umklapp region and for Poiseuille flow in the low-temperature region, but both these effects were considerably smaller than reported by other investigators for specimens grown at lower pressures.

I. INTRODUCTION

Because of its large zero-point energy, helium 4 does not solidify at pressures less than 25 atm, even near 0°K. At this temperature and pressure, the molar volume of the solid is 21 cm³, and the phase is hcp. Other than a small region of stability of the bcc phase, the hcp phase exists at melting pressures up to 1100 atm (molar volume 12 cm³ and melting temperature 15°K). The large value of the compressibility is a consequence of the weak Van der Waals forces and the large zero-point motion of the helium atoms. A further consequence is that the harmonic approximation is not applicable to solid helium, and so more sophisticated theoretical methods are required. For example, Nosanow and others¹ have used a time-dependent Hartree approximation in which the actual interatomic potential is replaced by a pseudopotential which includes the effects of short-range correlations. The calculations bear out the experimental observation that the thermodynamic behavior of hcp solid helium 4 is rather like that of ordinary solids. In particular, the temperature dependence of the specific heat may be described by a density independent function of the Debye temperature at $T = 0^\circ\text{K}$. As for ordinary solids, then, the Debye temperature is a valid and convenient parameter with which to qualitatively characterize the thermal conductivity.

Large changes in the thermal conductivity of solid helium accompanying changes in density are predicted from the dependence of the thermal conductivity on the Debye temperature and therein the density. In the simplest form of the single-mode relaxation time approximation,² the thermal conductivity of a nonmetal is given by the equation

$$\kappa(T) = \frac{1}{3} \sum_{\bar{q}} C(T, \bar{q}) V_{\bar{q}} l(T, \bar{q}).$$

Here $V_{\bar{q}}$ is the group velocity of the \bar{q} th phonon mode, $l(T, \bar{q})$ is the combined phonon mean free path in the presence of all scattering processes

which do not conserve wave vector, and $C(T, \bar{q})$ is the contribution to the specific heat of the \bar{q} th phonon mode. A good approximate solution can be obtained if the sum is replaced by an integral over a Debye density of states, assuming a constant group velocity.³ Since the specific heat and sound velocities are well known in most cases, a calculation of the phonon-scattering rates, and hence of the mean free paths, is sufficient to determine the thermal conductivity. For the purposes of this paper, we shall ignore the detailed wave-vector dependence of the mean free path and write, in analogy with a particle gas,

$$\kappa(T) = \frac{1}{3} C(T) V \bar{l}(T).$$

In this expression $C(T)$ is the total specific heat, V is the sound velocity, and $\bar{l}(T)$ is an average phonon mean free path.

In a perfect but finite crystal, the phonon mean free path will be limited only by collisions with the boundaries and by phonons scattering from other phonons, i.e., by anharmonic interactions. Furthermore, the normal (N) processes, that class of anharmonic interactions in which crystal momentum is conserved, should not be included in the calculation of the average mean free path, since these interactions do not alter the total heat current.⁴ The umklapp (U) processes, on the other hand, do not conserve crystal momentum, and these must be included in the calculation of $\bar{l}(T)$.⁵ Peierls showed that for temperatures well below the Debye temperature, θ_D , the U -process mean free path, $\bar{l}_U(T)$, is proportional to $\exp(\theta_D/bT)$.⁶ Here b is a constant approximately equal to 2. The mean free path l_B determined by boundary collisions alone is a constant approximately equal to the crystal diameter.⁷ If the probabilities for boundary and umklapp scattering are added, the average mean free path is given by

$$\bar{l}(T) = [\bar{l}_U(T)^{-1} + l_B^{-1}]^{-1}.$$

As a rule of thumb, the two terms $\bar{l}_U(T)$ and l_B are equal at $T \approx \theta_D/50$. At low temperatures, $T < \theta_D/50$,

$$\kappa(T) \approx \frac{1}{3} C(T) V d \propto d T^3 \theta_D^{-2},$$

where d is the crystal diameter. For $T > \theta_D/50$, umklapp scattering dominates, and the thermal conductivity decreases exponentially as $\exp(\theta_D/bT)$. Near $T = \theta_D/50$, κ reaches its maximum value (see Fig. 1).

Although N processes do not contribute directly to the thermal resistance, they must be considered in certain instances.⁸ It was first suggested by Peierls,⁹ and later shown by several other workers,¹⁰ that if the N -process scattering is sufficiently strong, there will be an enhancement of the thermal conductivity caused by Poiseuille flow of the phonon gas.¹¹ The effect of Poiseuille flow on the thermal conductivity has been discussed in detail by Guyer and Krumhansl.¹⁰ They showed that if the mean free path for N -process scattering, \bar{l}_N , obeys the inequalities $\bar{l}_N \ll d$ and $\bar{l}_N \bar{l}_U \gg d^2$, the thermal conductivity has the form

$$\kappa(T) \propto C(T) V d^2 \bar{l}_N(T)^{-1}.$$

The term $\bar{l}_N(T)$ decreases with increasing temperature as T^{-n} , where n is between 3 and 5.¹² Thus, in the above limits, the thermal conductivity may vary as the sixth to eighth power of the temperature (see Fig. 1).

The first extensive study of the thermal conductivity of solid helium 4 was done by Webb, Wilkinson, and Wilks.¹³ Their results on samples grown at pressures between 50 and 150 atm showed that the U -process mean free path was indeed exponentially dependent of θ_D/T , as predicted by Peierls. In the same pressure range, Bertman *et al.*¹⁴ found the same exponential dependence over almost three orders of magnitude in the conductivity and, furthermore, showed that the conductivity below the peak had the characteristic T^3 dependence of boundary scattering. Supporting results in this pressure range are found in the He³-He⁴ mixture studies of Walker and Fairbank¹⁵ and Berman *et al.*¹⁶ In each of these cases, measurements were made on samples grown with the blocked fill-capillary technique, that is, grown under constant volume. Mezhev-Deglin,¹⁷ applying the constant-pressure growth technique developed by Shal'nikov,¹⁸ obtained thermal conductivity results indicating that the concentration of lattice defects in his crystals was far less than in those of the previous workers. Such a low concentration of defects was essential for his observation of the previously unobserved Poiseuille flow and, furthermore, greatly simplified the determination of N - and U -process scattering rates.¹⁹ In Mezhev-Deglin's work, crystals were grown at 60, 85, 153, and 185 atm, corresponding to Debye temperatures between 30°K and

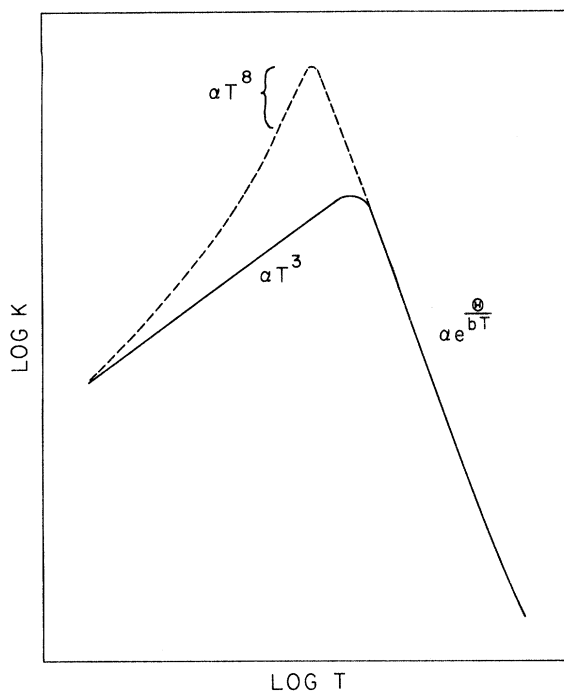


FIG. 1. Plot of thermal conductivity κ versus temperature, T , for an isotopically and chemically pure dielectric solid such as solid helium. The dashed line shows possible effects of Poiseuille flow.

45°K. Recent work by Hogan *et al.*²⁰ has been done on the thermal conductivity of crystals grown under constant pressures of 53, 85, and 126 atm. Solid helium 4, however, exists in the hcp phase at melting pressures up to 1100 atm ($\theta_D = 98^\circ\text{K}$). The only efforts to study the higher pressure range have been those of Webb and Wilks²¹ and of Berman *et al.*²² who did not grow their specimens at constant pressure. Webb and Wilks found rather low values for the maximum in the thermal conductivities. The present study was undertaken to obtain high-quality crystals using the constant-pressure growth technique at higher densities than the work of Mezhev-Deglin. The change in umklapp thermal conductivity over a wide range of density was studied.

II. EXPERIMENTAL PROCEDURE

A. Apparatus

Figure 2 shows the experimental chamber which was immersed in liquid helium. The heat sink, consisting of the high-pressure coupling and the helium pot, was thermally coupled to the bath through an exchange gas thermal switch.²³ With the pot empty, it could be warmed above the bath temperature with an attached heater. To reach the lowest temperatures the pot (volume 200 ml) was filled with liquid helium from the bath through

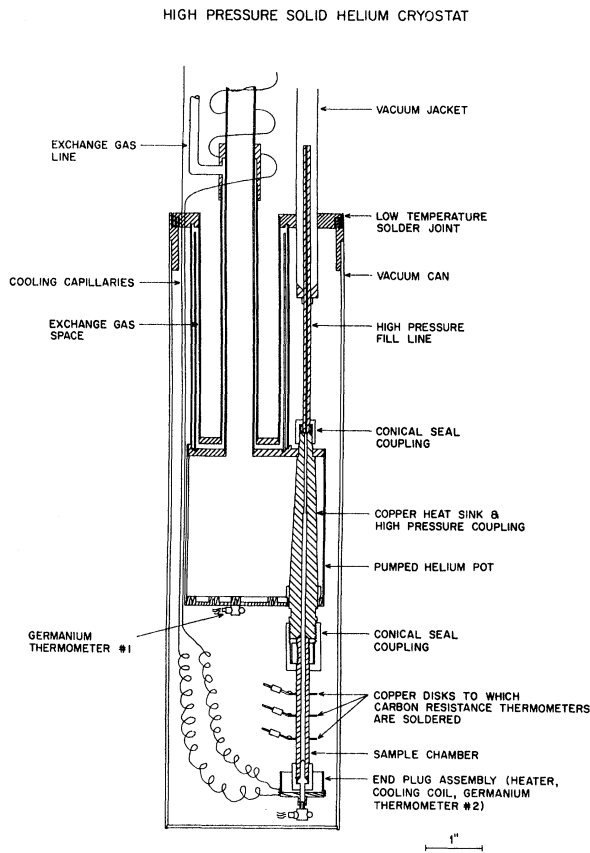


FIG. 2. Experimental chamber. Not shown are the tubes for evacuating the chamber and for filling the helium pot and the terminals for thermally grounding the electrical leads to the helium pot.

a needle valve. The practical temperature range was 1.1°K to about 40°K.

The sample chamber, filled through a vacuum-jacketed stainless-steel tube (0.125 in. o. d. × 0.025 in. i. d.), was either a thick-wall stainless-steel tube (2.1 mm i. d. × 2.1 mm wall) or a thin-wall inconel tube (2.2 mm i. d. × 0.1 mm wall). The inside surfaces of the sample chambers were smooth in the tubing, and were not treated further. The thick-wall tube, shown in Fig. 2, was sealed at both ends with conical assembly seals.²⁴ The thin-walled tube assembly is shown in Fig. 3. The copper end plugs ended in a sharp point to aid nucleation. Soft-soldered to the sample tubes at 1.0 cm intervals were three 0.005 in. thick copper discs to which the gradient thermometers, 80 Ω, 1/10 W Allen Bradley resistors, were soldered with Wood's Metal.

During crystal growth, cooling of the lower end-plug assembly below the pot temperature was provided by helium gas cooled to bath temperature and then passed through a 1/64 in. capillary tubing coil wound around the end-plug assembly. By adjusting the flow of gas, the amount of cooling could

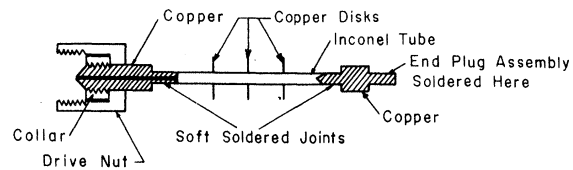


FIG. 3. Thin-wall sample tube showing conical seal coupling. (Figure 3 is not to scale.)

be varied between 0 and 25 mw. The capillaries leading to the coil were each 2 ft long and thermally anchored to the helium pot to minimize unwanted heat flow in the thermal conductivity measurements. The temperatures of the upper and the lower ends of the sample tube were monitored with calibrated-germanium resistance thermometers and controlled with heaters wound on the helium pot and the end-plug assembly, respectively. The resistance bridge and temperature control systems (Fig. 4) used were capable of maintaining the temperatures constant to better than 1 part in 10⁴.

Several purities of sample gas were used. Matheson "ultra high purity" helium with a chemical impurity < 15 ppm²⁵ was used with and without additional purification by an adsorption trap²⁶ cooled

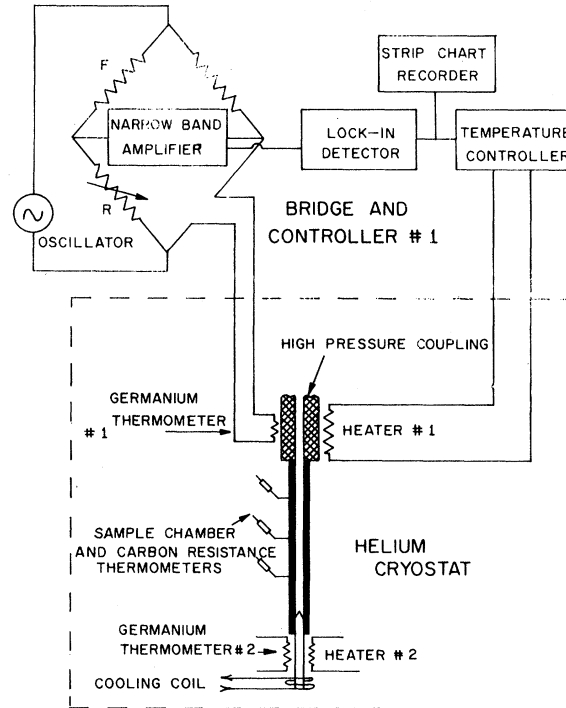


FIG. 4. Temperature control system. *F*-fixed arms of the Wheatstone bridge (5×10^5 ohms). *R*-motor-driven resistor for programming temperature during growth or a decade resistor for thermal conductivity measurements. Heater No. 2 and thermometer No. 2 are connected to an identical controller.

to 63°K. Matheson "research grade" helium with a chemical impurity < 4 ppm²⁷ was used without the adsorption trap for three samples. The sample gas was compressed to the desired pressure with helium gas of ordinary purity contained in a 1 liter vessel (see Fig. 5). Separation of the two grades of gas was accomplished with a cylinder and floating piston sealed with rubber O rings. The one-liter vessel, pressurized with an auxiliary gas system, served as a buffer volume insuring that the pressure in the sample tube remained constant during crystal growth. The pressure was measured with a 10,000 psi (100,000 psi for sample 1050-1) Heise gauge and remained constant during growth to within $\frac{1}{2}\%$.

B. Sample Growth

Prior to cooling the cryostat down to liquid-nitrogen temperature, the pressure system and sample tube were evacuated for at least four hours with a diffusion pump. The fill line of length 11 ft and i. d. 0.025 in. had sufficient transmission to insure that the sample-tube pressure was much less than 10μ . With the two cold traps in the pressure system cooled with liquid nitrogen, the high-purity helium gas was admitted, and the sample tube and pressure system were held at 2000 psi as the cryostat was cooled to liquid-nitrogen temperature. Just prior to transferring liquid helium, the sample gas was pressurized to the desired growing pressure by opening the valve between the ballast volume and the gas separator. With the vacuum jacket of the fill line evacuated, liquid helium was transferred and the sample chamber was allowed to cool to a temperature just above the freezing point.²⁸ In one case (601-8) the high-purity gas, after passing through the adsorption trap, was slowly admitted to the sample chamber *after* the evacuated sample chamber had been cooled to below 12°K.

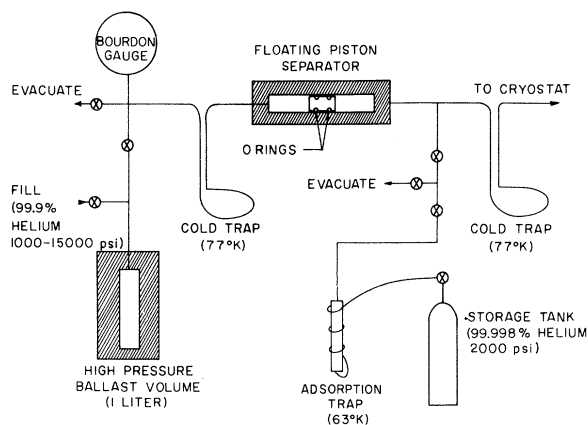


FIG. 5. High-pressure gas system. The adsorption trap was used for about a third of the samples grown.

The cryostat design with cooling at both ends of the sample tube allowed several variations in growth method. For Method I, an initial temperature difference of from 0.2 to 1.0°K was established between the ends of the tube with the lower end (the colder) being just above the freezing point. The temperatures of both ends of the tube were then programmed down by motor-driven resistors. A warming spike in the temperature of the lower end signaled the onset of solidification which usually occurred when the helium gas was supercooled by a few hundredths of a degree. As the temperature was further lowered, the solid-gas interface progressed up the sample tube until the gas in the copper heat sink froze. A double-pen recorder across the output of the lock-in detectors provided a continuous record of the deviations of the upper and lower temperatures from the programmed temperatures, and hence, showed clearly when nucleation occurred at the lower end and when the gas froze in the copper heat sink. The growth time shown for Method I in Table I for various samples is the time between these two events.

The other growth methods were used only with the thin-wall sample tube. For all of these methods, the progress of the solid-gas interface up the sample tube was monitored by observing the change in temperature of the carbon resistors along the sample tube. The growth time given for these methods in Tables I and II is the time required for the interface to transverse the 2 cm path used for thermal-conductivity measurements. Before growth was started, both ends of the sample tube were held at the same temperature – just above the freezing point. The upper end of the sample tube was held at this temperature while the lower-end temperature was lowered below the freezing point. In Method II the lower-end temperature was programmed down by a motor-driven resistor. This provided an approximately linear growth rate. In Method III, used only for the 601 atm growths, the lower-end temperature was quickly stepped from just above the freezing temperature, 10.15°K, to 8.35°K. The initial growth rate was quite rapid, but the growth slowed down as the interface progressed upward. The growth time for the lower portion was about half that for the upper portion. In Method IV the heater on the lower end was turned off and the cooling flow increased to provide a very rapid but poorly controlled growth. After the solid-gas interface had gone well past the upper copper disc in Methods II, III, and IV, the temperature of the lower end was fixed, and the temperature of the heat sink was lowered slowly through the freezing temperature. As in Method I, a warming spike indicated when the gas in the copper heat sink froze.

After growth was completed, the temperature of the upper end of the sample was reduced to that of the lower end in about an hour, and the helium-gas flow through the cooling capillaries was cut

TABLE I. Eighteen samples grown from gas not repurified. Samples 601-10, 11, 12 were grown in a thin-wall tube from gas with nominal chemical impurity less than 4 ppm. Samples 185-2, 3 and 601-2 were grown in a thin-wall tube from gas with nominal chemical impurity less than 15 ppm. The other samples were grown in the thick-wall tube from the 15 ppm gas.

Sample number	Our run number	Growth pressure (atm.)	Shown in figures	Growth method ^a	Growth time (h)	Umklapp "split" ^b	Max. value κ_{up} (W/cm deg)	Max. value κ_{10}	ppm gas
185-1	10	186	6, 12	I	9.5	No	4.4	5.6	15
185-2	23	188	6	I	4.5	Yes	2.8	3.8	15
185-3	24	188	6, 11, 12	I	8.0	Yes	4.1	2.7	15
290-1	8	290	7, 12	I	5.7	Yes	6.8	6.6	15
290-3	6	290	•••	I	10.2	No	3.4	3.5	15
290-4	15	290	•••	I	9.2	No	3.8	4.8	15
320-1	18	320	•••	I	10.0	No	2.6	2.6	15
343-1	19	343	8	I	10.1	Yes	1.1	1.4	15
343-3	17	343	•••	I ^c	8.5	No	1.0	1.3	15
400-1	16	400	•••	I ^c	13.2	No	0.6	0.7	15
486-1	14	486	9	I	10.0	Yes	0.7	0.7	15
601-1	12	602	10, 12	I ^c	8.5	No	0.9	0.8	15
601-13	13	600	•••	I	10.5	No	0.9	0.9	15
601-2	39	603	10	III	0.6	No	0.9	0.9	15
601-10	36	600	•••	III	0.6	Yes	3.2	2.1	4
601-11	37	603	•••	II	0.8	Yes	2.7	2.3	4
601-12	38	601	•••	II	0.4	No	1.9	2.3	4
1050-1	20	1050	11, 12	I ^c	8.0	Yes	1.1	0.8	15

^aGrowth methods are described in detail in the text.

^bThe umklapp conductivity is said to be "split" if the conductivities of the two portions of the sample differ more than 10% at the right of the conductivity maximum (see Ref. 20).

^cGrowth stopped for a half hour to two hours immediately after nucleation and then resumed.

off. The temperature of the sample was further lowered by approximately 25% in several hours to the point at which thermal-conductivity measurements were begun and helium exchange gas (10 μ) was put in the jacket of the fill line to freeze it off.

C. Thermal Conductivity Measurements

The technique used for measuring the thermal conductivity is standard except that, since three gradient thermometers were used, the thermal conductivity of two adjacent 1 cm-long portions of the sample could be measured. At each temperature two steps were followed. With the sample under isothermal conditions, the carbon resistors were calibrated against the germanium thermometers; then a known power (from 10⁻⁵ to 10⁻² W) was put into the heater on the lower end, establishing a thermal gradient.²⁹ From the changes in resistance accompanying the power input, the thermal gradient was calculated from the T versus R curve for the resistors.³⁰ Temperature differ-

ences of about 0.2% of the ambient temperature were used. The conduction of the thin-walled sample tube was negligible at most temperatures. However, the conduction of the thick-walled sample tube had to be subtracted from the measured values. The thermal conductivity of the thick-walled sample tube (304 stainless steel) was found to be given by the expression

$$\kappa = (54.7T + 0.18T^3) \times 10^{-5} \text{ W cm}^{-1} \text{ K}^{-1}$$

to within 2% between 1°K and 6°K. The total error in the thermal conductivity measurements was estimated to be at worst $\pm 5\%$.

III. RESULTS

Tables I and II give the growth pressure, method, and time for each sample, as well as the maximum thermal conductivity observed for the upper and lower portions of the sample. Guyer and Hogan²⁰ used the difference in the thermal conductivities of the two halves of their crystals as an indicator of crystallinity. If the two conductivities at the

TABLE II. Twelve samples grown from gas with nominal chemical impurity less than 15 ppm that had been repurified by passing through an adsorption trap at nitrogen temperature. All samples were grown in a thin-wall tube.

Sample number	Our run number	Growth pressure (atm)	Shown in figures	Growth method ^a	Growth time (h)	Umklapp "split" ^b	Max. value κ_{up} (W/cm deg)	Max. value κ_{10}	Comments
290-2	25	290	7, 11, 12	II	2.2	Yes	4.2	10.2	
343-2	26	343	8, 11, 12	II	3.0	Yes	5.4	13.0	
486-2	27	486	9, 11, 12	II	2.2	Yes	10.2	3.6	
486-3	28	487	9, 12	II ^c	2.3	No	6.2	6.2	
486-4	29	491	•••	II	1.0	Yes	2.1	5.2	
601-3	30	601	10	II	2.8	Yes	2.7	2.8	
601-4	31	602	10	II	2.8	Yes	8.4	4.2	Growth same as 601-3
601-5	32	601	10, 11, 12	III	0.6	No	11.4	9.2	Excellent crystal
601-6	33	601	10	IV	0.2	No	2.3	3.0	0.1 mm mean free path
601-7	34	600	•••	III	0.6	Yes	9.0	6.3	Growth same as 601-5
601-8	40	603	•••	III	0.6	No	12.0	5.7	Growth same as 601-5
601-9	35	600	•••	III	0.6	Yes	1.9	4.7	

^aGrowth methods are described in detail in the text.

^bThe umklapp conductivity is said to be "split" if the conductivities of the two portions of the sample differ more than 10% at the right of the conductivity maximum (see Ref. 20).

^cGrowth stopped for a half hour immediately after nucleation and then resumed.

same temperature above the conductivity maximum differed by more than ten percent, they called this a "split" in the umklapp region. We have used the same terminology in our tables. Guyer and Hogan considered "unsplit" samples as single crystals. Table III shows the observed melting temperatures, T_M , for the various pressures, P , as well as the molar volumes, V_M , and Debye temperatures at $T=0^\circ\text{K}$, θ_D , used in this discussion. The melting temperature at a given pressure was accurate to about $\pm\frac{1}{2}\%$; this uncertainty was largely caused by supercooling of the sample gas before nucleation occurred. Molar volumes were determined from the melting temperatures using the data of Dugdale and Simon³¹

TABLE III. Growth data and thermodynamic parameters used in the analysis. Molar volumes are used to identify samples shown on the graphs (for comparison with other experiments on samples grown at constant volume).

P (atm)	T_m (deg K)	V_m (cm ³ /mole)	θ_D (deg K)
185 ± 1	4.95	16.4	48.5
290 ± 1	6.51	15.2	58.0
320 ± 1	6.87	•••	•••
343 ± 1	7.16	14.8	62.0
400 ± 1	7.88	•••	•••
486 ± 1	8.91	14.0	70.0
601 ± 3	10.15	13.5	76.0
1050 ± 10	14.50	12.2	95.0

and are accurate to 0.1 cm³/mole. Debye temperatures at $T=0^\circ\text{K}$ were determined from the molar volumes using an average of the specific heat results of Franck³² and Ahlers³³ and are estimated to be accurate to several percent.

Thermal conductivity as a function of temperature for a number of the samples is shown in Figs. 6-10, arranged by molar volume. Thermal-conductivity curves for the highest-conductivity samples at six different molar volumes are shown on a common graph, Fig. 11. Mean free paths were calculated for high-conductivity samples at six molar volumes and are shown in Fig. 12 as a function of reduced temperature, T/θ_D . The Debye form of the specific heat near $T=0^\circ\text{K}$ and the Debye velocity of sound (the longitudinal and transverse velocities assumed equal) were used in the calculations. The results of such calculations are inexact, since the actual specific heat does not exhibit a precise T^3 behavior for the reduced temperature range of this experiment, and since the appropriate sound velocity is not known; however, the Debye approximation is sufficiently accurate for purposes of the experiment.

IV. DISCUSSION

A. Effects of Gas Purity

One feature of these data is evident from the tables and graphs. The purity of the sample gas significantly affects the shape and size of the

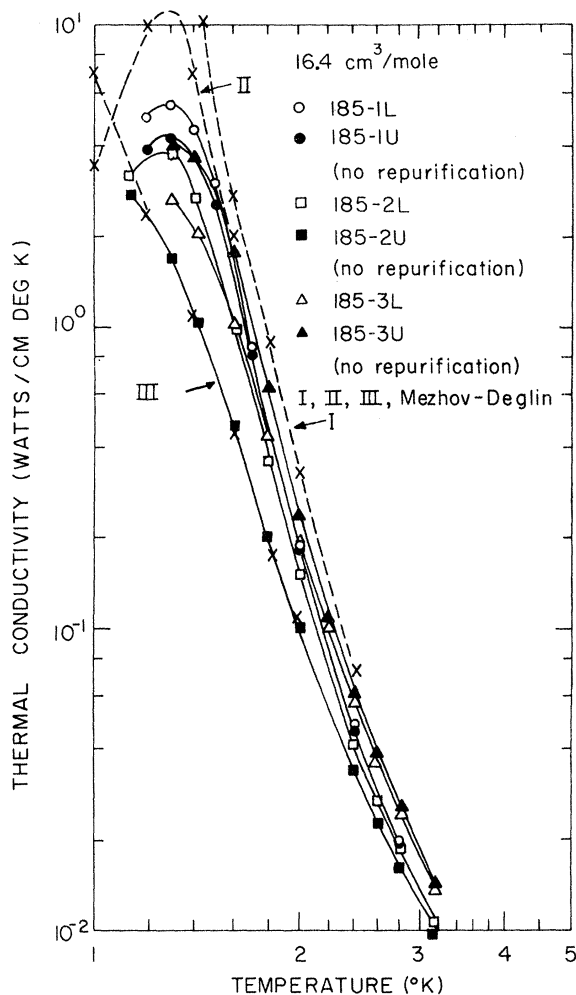


FIG. 6. Thermal conductivity of samples at 16.4 cm^3 molar volume (grown under constant pressure of 185 atoms). *L* refers to lower portion; *U* to upper. I, II, and III are characteristic conductivities observed at the same molar volume by Mezhov-Deglin.¹⁷

peak of the conductivity, especially at the higher densities. While there is little difference for the samples grown at 290 atm between unrepurified and repurified gas, all samples grown at 343 atm and above from the unrepurified gas show peaks of less than $1.5 \text{ W/cm}^\circ\text{K}$ (except the three samples grown at 601 atm from the unrepurified 4 ppm gas which showed higher peaks, but still less than $3.4 \text{ W/cm}^\circ\text{K}$). This suggests that at high densities, impurities result in the formation of scattering centers, which appreciably decrease the maximum phonon mean free paths. The impurities that cause the effect are apparently greatly reduced in concentration by passing the gas through an adsorption trap at 63°K , as seen (Fig. 10) by the difference in conductivities for samples 601-2 and 601-5 grown in the same sample tube with the same growth method and

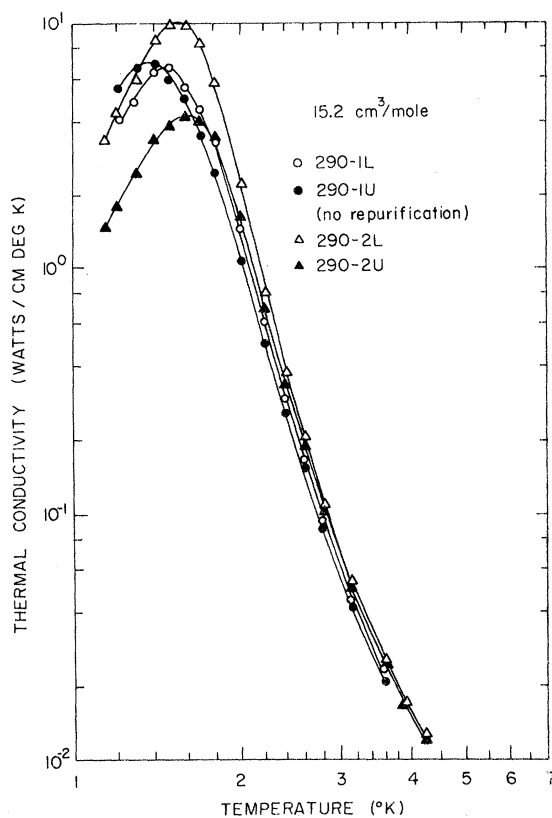


FIG. 7. Thermal conductivity of samples at 15.2 cm^3 molar volume (grown under constant pressure of 290 atoms). *L* refers to lower portion; *U* to upper.

time. Even after repurification by the adsorption trap, the purity may have varied from sample to sample. Three of the high-conductivity samples 601-4, 5, 6 were grown from the same fill of repurified gas kept at constant pressure and below 12°K between sample growths. The growth methods were different in each of these cases so the conductivities cannot be compared directly. This same fill of gas was also used for samples 601-3 and 601-4, but kept under constant pressure and below 80°K . These two samples were grown by the same method and had considerably different conductivities. The one sample, 601-8, grown from repurified gas admitted to the evacuated sample tube at temperatures below 12°K , did not have a peak conductivity much higher than 601-5. All these effects were small compared to the considerable difference in the conductivities between the unrepurified and the repurified 15 ppm gas. We note that Mezhov-Deglin¹⁷ obtained his best results when using gas obtained from a superfluid leak, indicating that extremely high purity was essential for observation of the large conductivities in his experiments.

B. Behavior in Umklapp Region

In spite of effects of different growth parameters

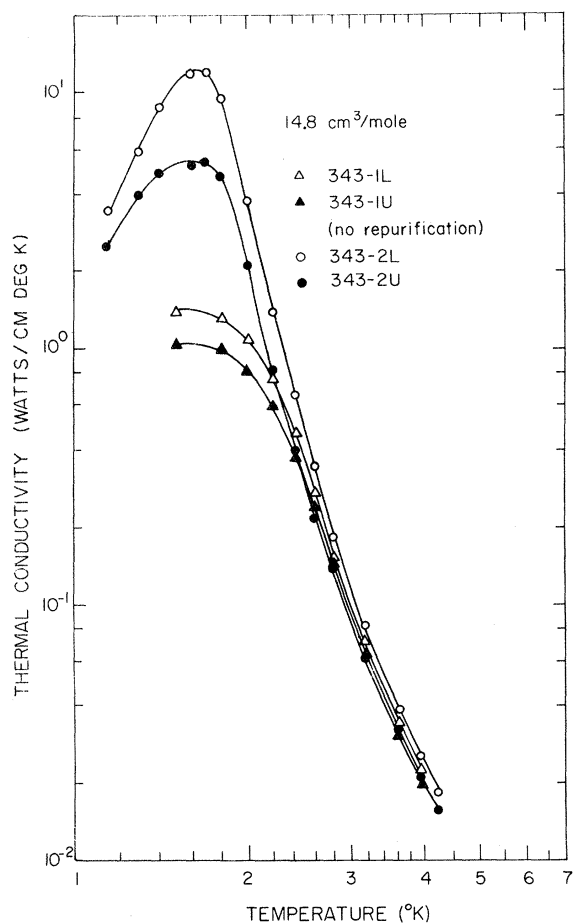


FIG. 8. Thermal conductivity of samples at 14.8 cm^3 molar volume (grown under constant pressure of 343 atoms). *L* refers to lower portion; *U* to upper.

and of different gas purities (the later effect being much larger), the high-temperature behavior of the samples exhibits a common feature at all the densities studied. The conductivity consistently varies as $\exp(\theta_D/bT)$ at the higher temperatures. The value of b does vary from sample to sample, consistent with some anisotropy resulting from differing crystal orientations. However, the range seen at $13.5 \text{ cm}^3/\text{mole}$ (Fig. 10) is significantly smaller than that found by Guyer and Hogan²⁰ at a larger molar volume ($18.3 \text{ cm}^3/\text{mole}$). Not enough samples were grown to justify an orientation analysis such as that of Guyer and Hogan, but other apparatus is currently being assembled to make thermal-conductivity measurements on crystals whose orientation is determined directly by x-ray analysis. In spite of effects of impurities and orientation, the high-temperature mean free paths for the best of the samples, when plotted as a function of T/θ_D , fall quite close to a common curve, as shown in Fig. 12. Several samples grown from reperfired gas follow this

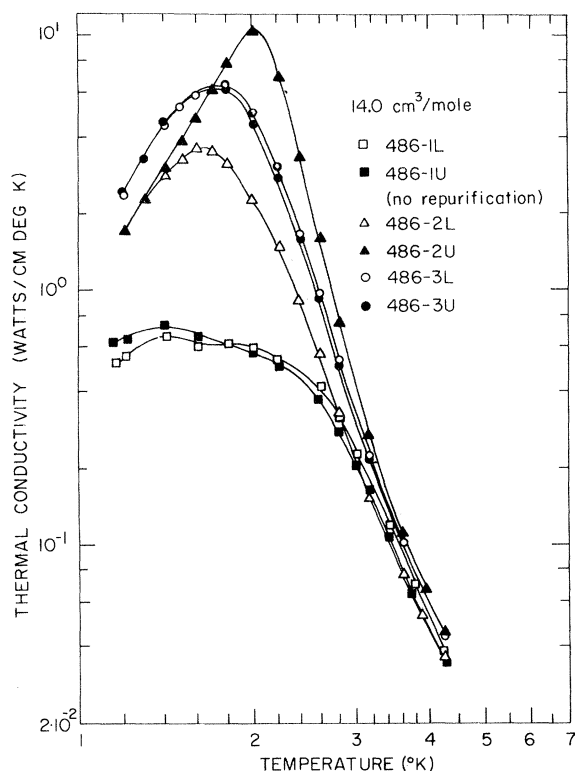


FIG. 9. Thermal conductivity of samples at 14.0 cm^3 molar volume (grown under constant pressure of 486 atoms). *L* refers to lower portion; *U* to upper.

curve for almost four orders of magnitude in mean free path, even though they have quite different densities. The simplest representation of the common curve is

$$\bar{l} = A \exp(\theta_D/bT),$$

where \bar{l} is the mean free path, $A = 2.5 \times 10^{-8} \text{ cm}$, and $b = 2.13$. (This value of b is not quite the same as one would obtain from the thermal conductivity curve at a given density because of the T^3 dependence of the specific heat). Any temperature dependence in A is masked by the scatter from sample to sample and the dominance of the exponential. For comparison with Mezhev-Deglin¹⁷ these data were also fit to the equation

$$\bar{l} = A'(T/\theta_D)^{5/2} \exp(\theta_D/b'T).$$

A fit of all data equally as good as that of Fig. 12 was obtained with $b' = 1.68$. This value compares quite well with the value of 1.76 ± 0.06 which Mezhev-Deglin obtained over the lower-density region.¹⁷ Thus, a purely classical model, in which the Debye temperature is presumed to be a function only of the density, appears adequate to fit the entire range of observed umklapp-limited thermal conductivities.

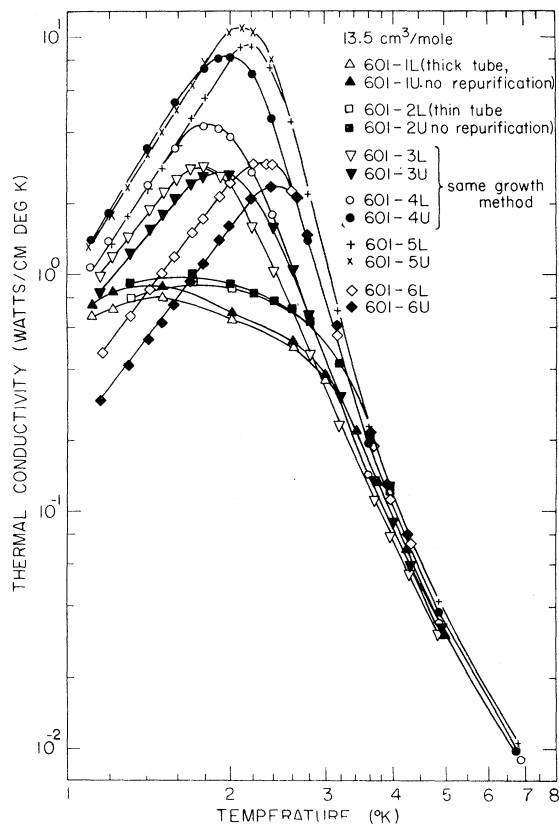


FIG. 10. Thermal conductivity of samples at 13.5 cm^3 molar volume (grown under constant pressure of 601 atoms). *L* refers to lower portion; *U* to upper. The dramatic effect of gas purity is clearly exhibited, as well as a large spread in the umklapp thermal conductivities. The largest number of samples was grown from repurified gas at this molar volume.

C. Low-Temperature Region

The mean free path at lower temperatures is expected to be a constant value equal to the sample diameter d shown on Fig. 12 at 0.22 cm. To demonstrate that the mean free path was temperature independent, it would have been necessary to go to temperatures below those possible in this experiment. The trend is demonstrated in Fig. 12, however, for samples grown from repurified gas. The constant values approached are not all the same, presumably because of structural defects or impurity scattering; the former is the most likely since this would lead to a constant mean free path. A sample grown very rapidly, 601-6, shown in Fig. 10, had a constant mean free path of about 0.1 mm for temperatures below 2° , even though the umklapp region was unsplit and showed a high conductivity. In this case, the growth rate was probably so fast that small grains were formed which were of average diameter 0.1 mm. The fact that the mean free path goes through a maximum in the other samples grown from repurified

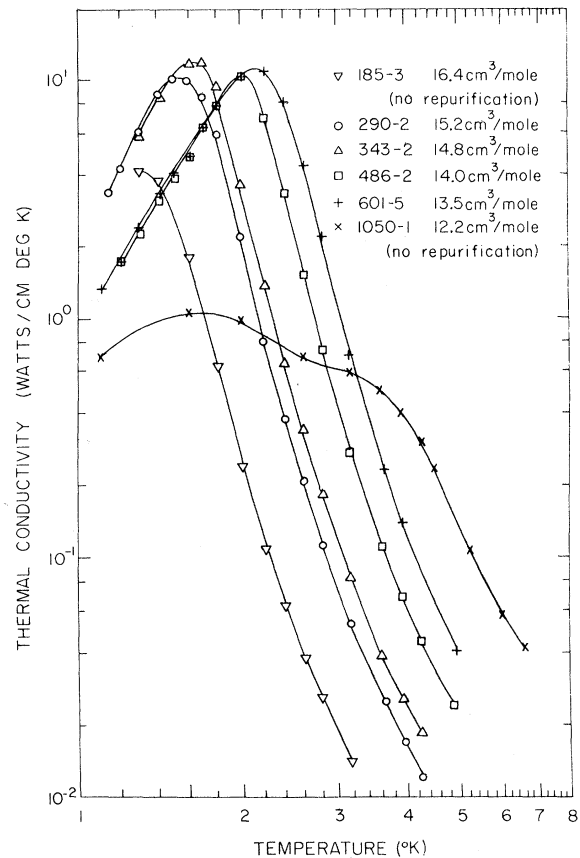


FIG. 11. Thermal conductivity of the highest-conductivity samples at six different molar volumes. *L* refers to lower portion; *U* to upper. Notice the large shift in the umklapp portion of the curves with density. Sample 185-3 and 1050-1 were grown with unrepurified gas and exhibit a much smaller range of umklapp-process limited thermal conductivity than those grown with repurified gas. The coincidence of the low-temperature conductivities of 290-2 and 343-2 and 486-2 and 601-5 is presumed to be accidental.

gas can only be attributed to the contribution of Poiseuille flow to the thermal conduction. The extent of the Poiseuille flow is not great enough to permit calculation of the normal-process mean free path.

D. Effects of Annealing, Thermal Shock, and Growth Methods

Several samples were annealed at constant volume after one set of measurements, but the conductivity usually decreased as a result. Sample 601-8 was annealed for an hour with the heat sink a few millidegrees above the melting temperature so that the fill line was open all the way to the sample tube and the sample was actually slowly melting under constant pressure. The heat sink was carefully refrozen and the sample slowly

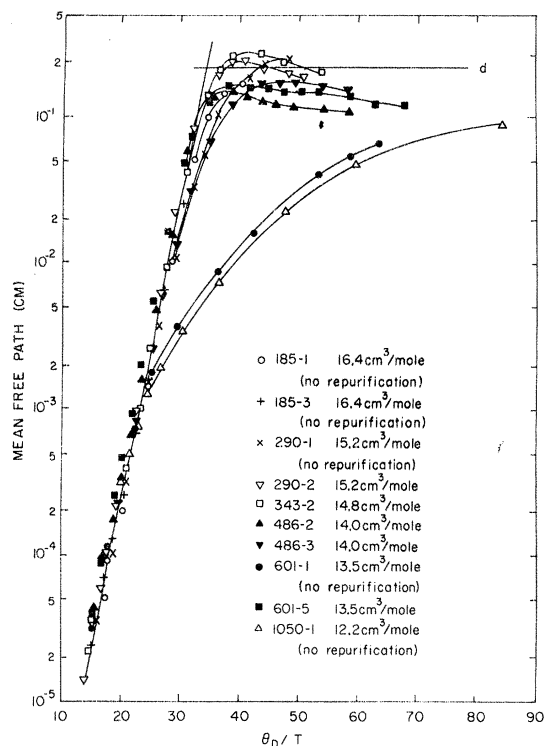


FIG. 12. Mean free path of the highest-conductivity samples at six different molar volumes. The diameter of the sample chambers, $d=2$ mm, is shown. Mean free paths, \bar{l} , were computed from $\bar{l} = 3\kappa/CV$, where κ is the thermal conductivity, C is the low-temperature Debye specific heat, and V is the Debye sound velocity (calculated with transverse and longitudinal velocities presumed identical). The Debye temperatures used are given in Table III. Note that even samples grown with unrepurified gas fall on the common curve at higher temperatures.

cooled. The conductivity was slightly decreased at all temperatures as a result.

Another sample, 601-5, was not annealed, but it did receive a severe thermal shock when the pot ran out of liquid while measurements were being made at 1.8°K. The temperature rose rapidly to more than 6°K but less than 8°K. Subsequent measurements indicated *no* change in the peak or umklapp conductivities. Mezhev-Deglin¹⁹ reports that the construction of the platinum thermometer connections in his glass sample tube is very critical and can cause thermal strains. This may be part of the reason he saw pronounced effects of thermal shock.¹⁷

None of the several growth methods tried yielded consistently high peaks, partly because the direction of growth is somewhat random. For several of the samples grown from unrepurified gas and for sample 486-3 grown from repurified gas, an alteration was made from the growth methods mentioned previously. Just after nucleation at

the lower end of the sample tube, the motor-driven resistors were stopped for a half hour to two hours. During this time the interface advanced very slowly, and it was hoped that the seed crystals would anneal. Then growth was resumed. Such a method has yielded consistently good crystals of neon, argon, and krypton for Batchelder *et al.*³⁴ and was helpful in obtaining high-quality solid helium crystals at a lower density for samples grown under constant pressure by Hogan *et al.*²⁰ We were not able to obtain high maxima in thermal conductivity in the cases tried, but typically the crystal grown after such a stoppage was uniform, as can be seen in Fig. 9 for sample 486-3.

The thick-wall sample tube was not used for the samples grown from repurified gas because the thin wall was expected to provide more favorable growth conditions by forcing most of the latent heat produced during freezing to be conducted through the solid as in Mezhev-Deglin's experiments in a glass tube. The thin-wall tube was not considered safe enough to hold 1050 atm so that density was not repeated with the repurified gas. In fact, no effects were definitely attributed to the difference in sample-tube thicknesses.

V. CONCLUSIONS

In this work we have obtained high-quality samples (possibly single crystals in some cases) of high-density hcp solid helium 4. Quality of the samples was inferred from the effective phonon mean free path. Variations in quality due to sample-tube wall thickness and sample growth method were small compared with the large impurity effect. The impurity effect was most significant at the higher densities. The effect was greatly reduced by purification with an adsorption trap at liquid nitrogen temperatures. We do not know if the impurities scatter phonons strongly or if they initiate structural defects which scatter phonons. The pressure dependence of the effect may be a clue to what impurity and what scattering mechanism is most important.

If Poiseuille flow occurs in the density range studied for our sample diameter, the quality of samples was not good enough to permit it to be significant. We suspect that better purification would improve sample quality and permit the normal-process mean free path to be obtained in the density range studied. One of the authors (WDS) will be investigating the thermal conductivity of higher-quality samples at lower temperatures than those attainable in this experiment.

Two aspects of our work are comparable with the results and interpretations of Guyer and Hogan at lower densities. We did observe a variation in umklapp conductivity from sample to sample at a given density that could be due to strong anisot-

ropy of the umklapp scattering processes. We also observed in some cases a correlation between lower umklapp conductivity and higher low-temperature conductivity; Guyer and Hogan suggest that this correlation may be due to anisotropy of the sound velocity. Neither of these effects was as large as they found at the lower densities.

The principal result of this work is that the effective umklapp mean free path for the purer samples is approximately the same function of the reduced temperature $T/\theta_D(0)$ for all densities used in this work. The impure samples are also described by this function at higher temperatures. Moreover, this quantum solid exhibits a mean free path that is in good agreement with Peierls's

classical theory of umklapp-process limited thermal conductivity over four orders of magnitude for the best samples.

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