Boundary-limited thermal conductivity of hcp ^{4}He

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We have been able to grow constant-volume hcp ^{4}He polycrystalline samples that exhibited Poiseuille flow of phonons as determined by thermal-conductivity measurements. These measurements were also extended well into the boundary scattering region to an order of magnitude lower temperature than previously reported. We find the conductivity is $k \propto T^3$ throughout this region. We were also interested in searching for any significant departures from this T^3 behavior that may have indicated the onset of specular reflection of phonons, or a supersolid transition. Such phenomena have been predicted for sufficiently low temperatures, and in view of our results, 30 mK would now appear to be an upper bound for any occurrence of these phenomena.

The theory of heat transport in a solid has played a central role in the development of the quantum physics of condensed matter. Both as a test of these theories, and as an indicator of which direction is appropriate to more elaborate models, thermal-conductivity measurements on hcp ^{4}He have been very fruitful. The interesting experimental attributes of this matter include the wide range of molar volumes readily available, extremely low natural isotopic impurity levels (<1 ppm ^{3}He), a thermal conductivity that is highly anisotropic, and the relative ease with which defect-free high quality crystals can be grown.

It was this last attribute that attracted several researchers to study single crystals of hcp ^{4}He and test predictions for the observation of Poiseuille flow of phonons.[1,2] There are rather stringent conditions on the quality of the crystal for Poiseuille flow to be present.[3] The first condition can be expressed in terms of the resistive process mean free path λ_R; one must have $\lambda_R >> d^2/\lambda_N$, where d is the distance a phonon travels until scattered at a boundary, typically the crystal dimension, and λ_N is the wave-vector–conserving normal process mean free path. The second necessary condition is that there are a large number of normal-type collisions compared to boundary scattering collisions: $\lambda_N << d$.

Among the many resistive processes that can limit λ_R is phonon scattering by crystal defects or strains. A polycrystalline sample will often times have a large number of imperfections which makes for a short resistive mean free path. In the case of ^{4}He, Poiseuille flow has only been observed in nearly defect free single crystals grown at constant pressures.

We have been able to grow polycrystalline ^{4}He samples, using the blocked capillary technique, that exhibit Poiseuille flow of phonons. In Fig. 1, we show two samples that were grown at the same molar volume $v_M = 21.0$ cm^3/mole. The sample represented by open and closed circles and open triangles was formed by filling the sample chamber with liquid ^{4}He at 83.4 bar, and then starting to cool the sample with a ^{4}He pot. The melting curve was first intersected by the liquid in the fill capillary, and this solid plug resulted in a constant volume growth. The sample was then cooled at a rate of about 2 mK/min until an all solid sample was formed.

The sample chamber and the resistor thermometry have already been described.[4] However, because of the small magnitude of the thermal conductivity of ^{4}He at very low temperatures, $T < 50$ mK, it was necessary to carefully consider several additional experimental corrections. These corrections did not play a significant role in the case of solid ^{3}He in Paper I. For this reason, the resistor calibration was rechecked and the thermal conductivity of the empty Stycast sample chamber was remeasured after all the conductivity data was collected. In the case of ^{4}He, the thermal conductance of the empty chamber amounts to nearly a 10% correction to the ^{4}He conductivity at our lowest temperatures.

What amounted to a comparable correction to the Stycast conductance was what we believe to be a vibrational heat leak into our sample chamber. We estimated the amount of this correction by the fall off of the Stycast data points below a $k \propto T^2$ dependence at very low temperatures. This heat leak, which was approximately 3×10^{-9} watts, was also consistent, at our lowest temperatures, with a small deviation of our resistance thermometers from an $\exp(AT^{-1/4})$ behavior.[5] For the samples with the lowest conductivities (Fig. 2), we show the uncorrected data points, as well as the corrected data points which are indicated by a circle with a cross.

The data points shown as circles in Fig. 1 were taken during warming after the sample was cooled to $T \lesssim 25$ mK. These points show a thermal hysteresis

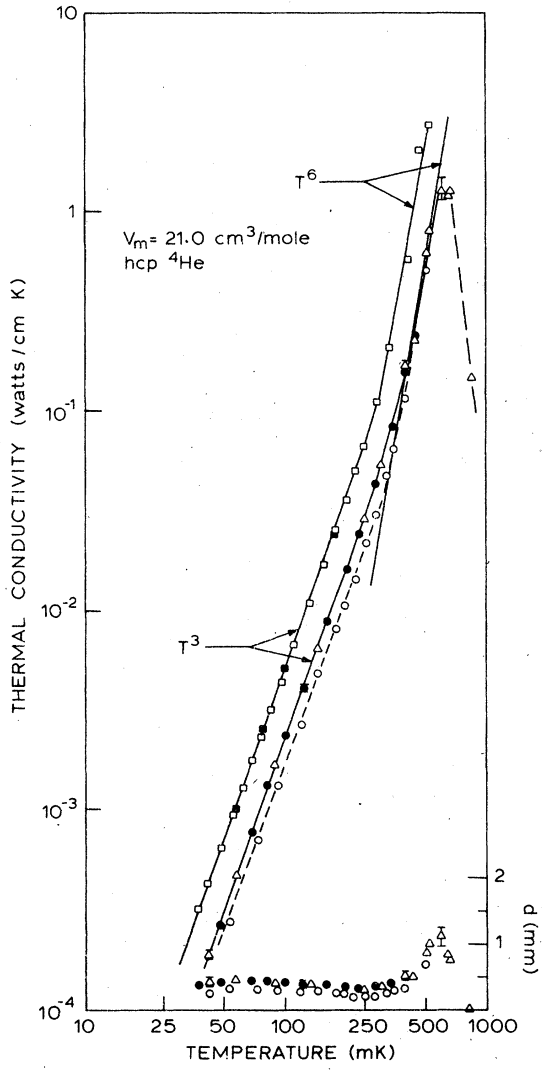

FIG. 1. Data points shown as open circles are taken during the first warming cycle after the sample had been cooled to 25 mK. The closed circles are data taken on a subsequent cooling, and the open triangles on the subsequent warming. The sample was then annealed and allowed to cool. Data taken during warming are shown as open squares, and the subsequent cooling are the closed squares.

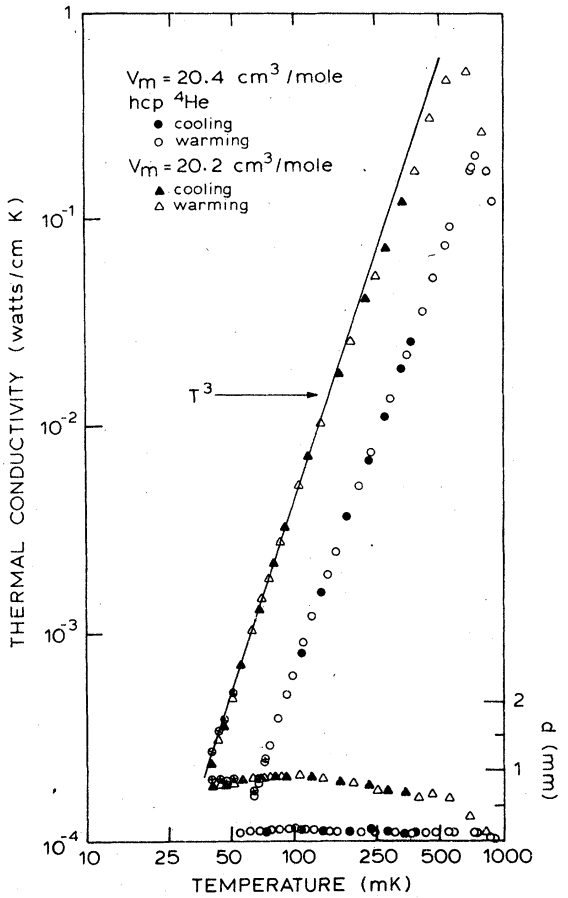

FIG. 2. Small depression in the thermal conductivity below a T^3 dependence near k_{max} in the sample at 20.2 cm^3/mole is believed to be due to scattering from edge dislocations. The ⊕ data points are corrected values when a heat leak is taken into account as described in the text.

when compared to the data taken during a subsequent cooling and warming. We believe the shift in the conductivity in the first warming cycle is due to a healing of strains, and it was very similar to several constant volume bcc ^{3}He samples which we described in Paper I. The conductivity, within the limits of uncertainty as indicated by error bars on representative data points, did not change under continuing thermal cycling.

We annealed this sample by holding it close to melting temperature for 24 hours. These results are indicated by the squares in Fig. 1. As expected, the anneal resulted in an increase in the mean free path; it increased by 50% for this case. Except for an overall enlarging of crystal size, both samples show a similar thermal conductivity. In what follows, we confine our comments to the data points represented by closed circles and open triangles since this data set also includes conductivity points on both sides of the thermal-conductivity maximum, k_{max}.

The conductivity measurements of the samples grown at constant volume show Poiseuille flow in the temperature range 300 to 600 mK. As expected for such flow below k_{max}, we find $k \propto T^6$. In terms of the mean free path d, we observe an enhancement of d by the factor $\frac{5}{32}(d/\lambda_N)$ in the region between 400 and 600 mK. We analyzed the data for λ_N using the expression for the thermal conductivity in the Poiseuille region[3]

$$k_p = \frac{1}{3} C_V \frac{5}{32} \frac{d^2 v}{\lambda_N}, \tag{1}$$

where v is the Debye velocity,

$$v = \frac{k_B \Theta}{\hbar (6\pi^2 N)^{1/3}} \tag{2}$$

and d was taken to be 0.5 mm. The latter value was determined from the thermal conductivity in the boundary scattering region by inverting the kinetic equation for the boundary-limited thermal conductivity, k_b,

$$k_b = \tfrac{1}{3} C_V v d \ , \tag{3}$$

to arrive at an expression for the mean free path in terms of molar volume, v_m, and Debye temperature, Θ:

$$d = \frac{3.9 \times 10^{-5} v_m^{2/3} \Theta^2}{T^3} k_b (\text{mm}) \ . \tag{4}$$

The appropriate sound velocity in Eq. (3) actually depends on crystal orientation because of the anisotropic nature of hcp ^{4}He, but in the boundary scattering region, this angular dependence is weak[2] and setting this velocity equal to the Debye velocity is a reasonable approximation.

The molar volume for the samples in Fig. 1 is 21.0 cm^3/mole, which is close to melting density. If we use $\Theta = 24.0$ K, appropriate to this molar volume, we find in the Poiseuille region from Eq. (1)

$$\lambda_N(t) = 7.8 \times 10^{-3} T^{-3} \ (\text{mm}) \ . \tag{5}$$

This value, when adjusted for the differences in molar volumes, is in excellent agreement with the values reported by Mezhov-Deglin[1] and Hogan *et al.*[2] on hcp crystals grown under constant pressure.

To the best of our knowledge, all previous thermal-conductivity measurements on hcp ^{4}He were limited to temperatures $T \gtrsim 0.3$ K, since refrigeration was accomplished by pumping on a ^{3}He pot. As a result, conductivity measurements did not extend much beyond the transition between the Poiseuille flow and boundary-limited region.[6] By making use of a helium dilution refrigerator, we were able to cool our samples to $T \approx 30$ mK and more extensively explore the boundary-limited region of thermal conductivity.

We show, at the bottom in Fig. 1, the mean free path of the first sample computed using Eq. (4). Below 400 mK, the mean free path is temperature independent and is $d \approx 0.5$ mm. This value of the mean free path is interpreted as phonons diffusively scattering from the crystal boundaries.[7] Since the sample chamber diameter is 3.07 mm, we clearly have a polycrystalline sample. Similarly, the annealed sample has a temperature-independent mean free path ≈ 1.0 mm.

In Fig. 2, we show data for two samples formed at molar volumes 20.4 and 20.2 cm^3/mole. Neither sample displays Poiseuille flow. In fact, the sample at 20.2 cm^3/mole has a depression in the conductivity for temperatures below $k_{\max}$. We have observed similar depressions in bcc ^{3}He crystals, and we attribute the reduced conductivity to phonon scattering by edge dislocations. Such scattering limits $\lambda_R \lesssim d^2/\lambda_N$ and prevents development of Poiseuille flow. The lower molar volume of this sample compared to the samples in Fig. 1 resulted in a larger locked-in density of edge dislocations.

A resistivity analysis for the 20.2 cm^3/mole sample, similar to that used in Paper I for ^{3}He, was consistent with an additional additive resistivity $W \propto T^{-2}$. However, there were only a limited number of data points that were depressed below $k \propto T^3$, and consequently, the fit to the resistivity was not as certain as in the cases of the bcc ^{3}He. The analysis was also complicated by a slight enhancement of k close to $k_{\max}$ suggesting a Poiseuille flow which was not quite completely suppressed.

The sample at $v_m = 20.4$ cm^3/mole also does not display Poiseuille flow. However, in this case, the resistive mean free path is $\lambda_R \lesssim d = 0.1$ mm and is due to the phonons scattering at crystal boundaries. As can be seen at the bottom of Fig. 2, this mean free path is temperature independent from $k_{\max}$ to the lowest measured temperatures.

In exploring the boundary-limited region of hcp ^{4}He, we were especially interested to see if we would observe an increase in the mean free path at very low temperatures due to specular reflection. Such an increase is expected when the phonon wavelength, $\lambda_{ph} \approx h v / k_B T$, where v is defined in Eq. (2), becomes large compared to some rms measure of the surface roughness, f. An estimate for the probability for specular reflection[8] is

$$p(\lambda_{ph}) \sim \exp(-16\pi^3 f^2 / \lambda_{ph}^2) \ . \tag{6}$$

This probability is small unless $f < \lambda_{ph}/4\pi$. For the lowest temperatures in our experiments, we estimate $f \simeq \lambda_{ph}/4\pi \simeq 500$ Å. Although we expect a distribution of rms surfaces in our samples, we would have also expected that, since the phonons are scattering primarily from crystal boundaries, such surfaces would be sufficiently smooth at the lowest temperatures, so that we would have observed some indication of specular reflection. One interpretation of this result is that the estimate of surface roughness for specular reflection to occur, provided by Eq. (6), must be too generous.

There have been several theoretical investigations into the possibility that a supersolid state may exist in certain crystals at very low temperature.[9] Although the exact nature of the transition is still uncertain, it has been predicted that some form of superfluidity is a distinct possibility and that solid ^{4}He would be a likely candidate for experimental confirmation of the theoretical predictions.

We were originally enticed by the possibility of ob-

serving a change in the thermal conductivity that could be interpreted as a sign of enhanced heat flow. Such a heat flow may be similar to the fountain effect in superfluid He II. We have no estimate for the size of such an effect in hcp ^{4}He, but as can be seen in Figs. 1 and 2, we see no significant change in the conductivity which may be indicative of such a transition. In view of our results, future searches for a supersolid transition would appear to be more successful for $T < 30$ mK.

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