Simultaneous Measurements of the Torsional Oscillator Anomaly and Thermal Conductivity in Solid $^4$He

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In these torsional oscillator experiments the samples of solid $^4$He were characterized by measuring their thermal conductivity. Polycrystalline samples of helium of either high isotopic purity or natural concentration of $^3$He were grown in an annular container by the blocked-capillary method and investigated before and after annealing. No correlation has been found between the magnitude of the low-temperature shift of the torsional oscillator frequency and the amount of crystalline defects as measured by the thermal conductivity. In samples with the natural $^3$He concentration a substantial excess thermal conductivity over the usual $T^3$ dependence was observed below 120 mK.

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Solid helium is the best example of a quantum crystal, in which substantial zero-point motion makes atoms and crystalline defects highly mobile even at very low temperatures. Kim and Chan observed anomalies in its response to rotational acceleration [1]: the resonant frequency shift [4, 5] but sometimes in its increase [5], while single samples often result in the reduction of the frequency shift. This frequency shift disappears when the TO is driven at high amplitudes. Such a shift would be expected to extend to our GHz frequencies in the temperature range of interest, 80–250 mK (at least within the models of glass-like dynamics of solid helium [8] and mobile dislocation segments [9], whose characteristic frequencies at $T \sim 100$ mK are $\sim 1$ kHz and $\sim 10$ MHz).

In our experiment, helium samples had a shape of cylindrical annulus of mean radius $r = 6.63$ mm, thickness $d = 0.30$ mm and height $h=14$ mm. Torsional oscillations were investigated about the cylinder axis while the thermal conductivity was measured along the axis. To estimate the phonon mean free path in the boundary-scattering limit, one can think of a long slab of rectangular cross-section of width $\sim 2(2rd)^{1/2}$ and hence aspect...
ratio \( n \sim 2(4\pi)^{1/2} = 13.3 \). For diffuse scattering off container walls, this leads to \( \ell \approx \frac{\pi}{3}(\ln 2 n + \frac{1}{2})d = 0.85\ \text{mm} \) [14], while exact numerical calculations for our annulus yield \( \ell = 0.97\ \text{mm} \) [13]. The cell was composed of two coaxial stainless steel tubes brazed together at the bottom (see inset in Fig. 1). Their roughness (asperity size) was on scale \( \sim 2\ \mu\text{m} \), and wall thickness of the inner (outer) tube was 0.5 mm (0.3 mm). The assumption of diffuse scattering off these walls would hold for phonons of wavelength 0.2 \( \mu\text{m} \) even for grazing angles \( \sim d/\ell = 0.3\ \text{rad} \) that mainly contribute in this geometry.

Thermal conductivity was measured by means of running a known current through the 1.5 kΩ gradient heater at the bottom of the cell and subsequently measuring the temperature difference established between two RuO₂ thick-film resistor thermometers glued with varnish to the side of the cell \( z = 7\ \text{mm} \) apart and 3.5 mm from either end. The end-effect correction to \( \kappa \) [14] is small for \( \ell \leq 1\ \text{mm} \). The heat leak through the thermometer wires was negligible. With homogeneous and isotropic samples of polycrystalline solid helium in the cell, as well as with the empty cell, this arrangement results in a uniform axial distribution of temperature gradient. The temperature of thermometer 1, \( T \), was stabilized by applying a computer-controlled power to the heater attached to the platform. At each value of \( T \) we used 4–5 different values of the current in the gradient heater (from zero to maximum compatible with the cooling power of the cryostat). Depending on temperature, the process of stabilization took up to 1 hour, after that it could take up to 1 hour to average the readings of the thermometers in the steady state regime. Their temperature difference \( \Delta T \) was proportional to the power dissipated in the gradient heater \( Q \) with an uncertainty of 15% (at the lowest reported temperature) or less. The value of \( \kappa = \frac{Q}{\Omega \Delta T} \) was thus calculated with the absolute accuracy of \( \sim 10\% \). The typical values of \( Q \) ranged from 10 nW to 1 µW, and \( \Delta T \) were between 0.1 and 1 mK, i.e. within \( 10^{-3}–10^{-2}T \).

For the measurements of the TO response the temperature has been typically swept from 300 mK to the lowest achievable temperature, 22 mK, over 15 hours. On sweeping the temperature twice slower we have not observed any difference; hence, the presented temperature dependences are equilibrium. The TO amplitude was kept sufficiently low, so that 50% increase of the amplitude of the driving voltage did not result in any decrease of the magnitude of the frequency shift at low temperatures.

The thermal conductivity of the empty cell was found to be proportional to temperature and consistent with that for stainless steel; it did not exceed 20% of the total thermal conductivity at the lowest temperatures and was subtracted. We have also measured the temperature dependence of the resonant frequency and linewidth of the empty TO. These were subsequently subtracted from the data obtained with the filled cell. The resonant frequency of the empty cell increased by 0.3 mHz upon filling the cell and solidification. To melt and regrow the sample we used either the gradient heater or the platform heater. By varying the heater current and heating time we were able to vary the sample growth time between \( \sim 40\) s and several hours.

The thermal conductivity of the sample of purified \( ^4\text{He} \) was subtracted. We have also measured the temperature dependence of the resonant frequency and linewidth of the sample. These were subsequently subtracted from the data obtained with the filled cell. The resonant frequency of the sample increased by 0.3 mHz upon filling the cell and solidification. To melt and regrow the sample we used either the gradient heater or the platform heater. By varying the heater current and heating time we were able to vary the sample growth time between \( \sim 40\) s and several hours.

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where we could measure it, 80–250 mK (see example in Fig. 1 △). The value of \( \ell \), calculated using Eq. (1–3), was typically 0.23–0.27 mm for rapidly grown samples and increased to 0.34–0.40 mm after annealing. As all these values are smaller than the theoretical upper limit of \( \ell \approx 1 \text{ mm} \), phonon scattering off crystalline defects was dominant. Thus the quality of each sample can be characterized by the parameter \( \ell \).

Surprisingly, the thermal conductivity in all 9 samples of non-purified \(^4\)He was higher than that of purified samples. Moreover, all of them had low-temperature flat parts below \( T = 120 \text{ mK} \) (Fig. 1 • and ◦). The phonon m.f.p., as inferred from \( \kappa \propto T^3 \) parts, was 0.60–0.73 mm for all as-grown samples. Annealing resulted in further increase of \( \kappa \) – by a factor of 1.6–2.0 at \( T < 150 \text{ mK} \) and by a progressively larger factor at \( T > 150 \text{ mK} \). The resulting values of \( \ell \) (as extracted from \( \kappa \propto T^3 \) parts at \( T \sim 140 \text{ mK} \)) became 1.1–1.4 mm – in fair agreement with the upper limit expected for boundary-scattering regime, \( \ell \approx 1 \text{ mm} \) (perhaps enhanced slightly by specular scattering). It thus appears that only in annealed non-purified \(^4\)He phonon scattering was boundary-dominated.

Fig. 2 shows examples of the TO resonant frequency \( f \) and linewidth \( \Delta f \) for samples of purified and non-purified \(^4\)He. In the latter, the characteristic temperature is \( T_0 \approx 80 \text{ mK} \). In purified \(^4\)He the cross-over temperature is \( T_0 \approx 30 \text{ mK} \) [4], hence we could not reach the saturation of the frequency shift at our lowest attainable temperature \( T = 22 \text{ mK} \). Nevertheless, the resonant frequency shift between 80 mK and 22 mK, \( f_{22} - f_{80} \), can be used as a measure of the size of the effect under discussion. Using different rates of growing and annealing we were able to obtain samples of purified \(^4\)He with \( \ell \) ranging from 0.23 to 0.40 mm and measured the temperature dependence of resonant frequency for all of them. To our surprise, we have not observed any correlation between the magnitude of the frequency shift and \( \ell \) in these samples (Fig. 3). Similarily, in samples of non-purified \(^4\)He, no correlation was found between the TO frequency shift \( f_{22} - f_{300} \) and thermal conductivity (characterized by the value of \( \ell \) extracted from the \( \kappa \propto T^3 \) part at \( T > 120 \text{ mK} \)).

In different as-grown samples, \( f_{22} - f_{80} \) for purified \(^4\)He were in the range 2–8 mHz, and \( f_{22} - f_{300} \) for non-purified \(^4\)He were within 10–15 mHz. Subsequent annealing was not found to change the values of these parameters significantly. For example, annealing at 1.77 K for 10–13 hours only resulted in changes of the frequency shift and linewidth within ±10% in both purified and non-purified samples; at the same time \( \kappa \) typically increased by 50%–100% (Fig. 3). This is in contrast to a substantial reduction of the frequency shift observed in highly-disordered samples [4, 5] but in agreement with little effect of annealing on the frequency shift in either already well-annealed polycrystals or single crystals [5](albeit containing dislocation walls mobile above \( \sim \) 0.8 \( T_m \) [10]). We can thus speculate that our samples, even if polycrystalline, might begin with large grains of a size on the order of the gap width, and these grains do not coarsen any further; on the other hand, dislocations that could be efficient scatterers of phonons (especially those freely vibrating in purified \(^4\)He) can still relax readily. The observed absence of correlation between the TO frequency shift and phonon m.f.p. does indicate that these two parameters are sensitive to different types of disorder. For instance, the former could be more sensitive to grain boundaries while the latter – to uncorrelated dislocations.

![Fig. 2](image2.png)

FIG. 2. TO resonant frequency \( f \) (solid lines) and linewidth \( \Delta f \) (dashed lines) for samples with different \( x_3 \). The value of the resonant frequency at \( T = 300 \text{ mK}, f_{300}, \) was substracted. The frequency shift of 1 mHz corresponds to the effective de-coupling of 0.064% of the mass of solid \(^4\)He. The change of 1 mHz in the linewidth corresponds to 1.2 \times 10^{-6} change in the damping, \( Q^{-1} \), of the TO.

![Fig. 3](image3.png)

FIG. 3. The shift in the TO resonant frequency (between \( T = 80 \text{ mK} \) and \( T = 22 \text{ mK} \) for purified \(^4\)He, •; and between \( T = 300 \text{ mK} \) and \( T = 22 \text{ mK} \) for non-purified \(^4\)He, ◦) vs. the phonon m.f.p. Arrows show changes upon annealing at \( T = 1.77 \text{ K} \) for 10–13 hours.

The upward deviations from \( \kappa \propto T^3 \) at \( T < 120 \text{ mK} \), observed in non-purified \(^4\)He, cannot be accounted for by the enhancement of specular scattering as the wavelength of thermal phonons increases – because the weak temperature dependence \( \ell(T) \) observed in this regime (even for
phonon m.f.p. much greater than the asperity size and for slabs of a higher aspect ratio \( n \sim 20 \) \(^{20}\) would only lead to a slight reduction in the slope of \( \kappa(T) \), and thus can hardly account for the observed plateau-like \( \kappa(T) \). Neither can this anomaly be attributed to end effect caused by increased \( \ell \) – as this would decrease the apparent \( \kappa \) (and by different extent for different \( \ell \)). Last but not least, the presence of crystalline defects limiting m.f.p. to \( \ell < 1 \) mm (e.g. \( \ell = 0.65 \) mm in Fig. 1, •) would prevent grazing-angle phonons from propagating far even if boundary scattering becomes specular. We should hence conclude that there exists an additional channel of heat transport in parallel with Debye phonons. Indeed, an excessive contribution to the specific heat \(^{12}\) at similar temperatures have been observed recently, although never yet in the samples of solid helium that can be simultaneously characterized by the TO effect. If both effects are caused by the same underlying phenomenon, it seems unlikely that it is superfluidity (that in liquid \(^{4}\)He boosts heat transport by means of the counterflow of the normal and superfluid components). This is because we found that \( \kappa \) is independent of the TO amplitude: with a sample of non-purified \(^{4}\)He at \( T = 90 \) mK with the gradient heater turned on we were increasing the TO drive amplitude by a factor of 50 – until both the TO frequency shift and excess damping disappeared (the superfluid component would have vanished at this point), but the temperature difference did not change within the experimental accuracy of \( \sim 5\% \). It would be tempting to attribute the observed plateau in thermal conductivity to the gas of \(^{3}\)He impurities at concentration \( x_3 \) and m.f.p. of \( \sim 1 \) mm. However, even with a generous assumption of the impurity band of 0.1 K, one can only account for \( 10^{-4} \) of the observed value.

We note that Armstrong et al. \(^{10}\), who used \(^{4}\)He of apparently natural purity (i.e. \( x_3 \sim 3 \times 10^{-7} \)), did not observe any deviations of thermal conductivity from \( T^3 \) dependence down to \( T = 30 \) mK. It is likely that the reason in the difference from our observations lies in the geometry of the experimental volumes that can affect the types and density of crystal defects: a long cylinder of 3 mm diameter in \(^{10}\) versus essentially a 0.3 mm-thick slab in our case. It is plausible that confinement between parallel walls not only allows rapid quench from liquid into the solid phase but also helps to arrest relaxation and annealing of dislocations and grain boundaries.

The upward deviation from \( \kappa \propto T^3 \) at high temperatures 160–250 mK (Fig. 1, •), was only observed in non-purified samples after annealing and will require further investigation. Even though it looks similar to \( \kappa \propto T^8 \) dependence \(^{10}^{21}\) that is often observed in perfect samples at higher temperatures \( T \sim 500 \) mK at which frequent normal phonon-phonon scatterings can result in an effectively enhanced \( \ell \) (Poiseuille flow), at \( T < 250 \) mK the phonon gas is too rarified for hydrodynamic behaviour.

To summarize, we found that in different samples of polycrystalline \(^{4}\)He the magnitude of the TO frequency shift and the crystalline disorder as probed by thermal phonons are not correlated. The phonon m.f.p. readily increases upon annealing at \( T = 0.8 T_m \) while the value of the frequency shift hardly changes. Driving TO at high amplitude eliminates the TO frequency shift but leaves thermal conductivity unchanged. All these observations allow to conclude that the defects responsible for the TO anomaly and for the phonon mean free path in \(^{4}\)He have different properties. The discovered enhancement of the thermal conductivity at low temperatures is likely to be related to the recently observed excess contribution to the specific heat \(^{12}\) – the origins of both are yet unknown and require further investigation.

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[1] E. Kim and M. H. W. Chan, Science 305, 1941 (2004).

[2] A. F. Andreev and I. M. Lifshitz, Sov. Phys. JETP 29, 1107 (1969).

[3] A. J. Leggett, Phys. Rev. Lett. 25, 1543 (1970).

[4] A. S. C. Rittner and J. D. Reppy, Phys. Rev. Lett. 97, 165301 (2006).

[5] A. C. Clark, J. T. West, M. H. W. Chan, Phys. Rev. Lett. 99, 135302 (2007).

[6] E. Kim et al., Phys. Rev. Lett. 100, 065301 (2008).

[7] L. Pollet et al., Phys. Rev. Lett. 101, 097202 (2008).

[8] J.-J. Su, M. J. Graf, and A. V. Balatsky, Phys. Rev. Lett. 105, 045302 (2010).

[9] I. Iwasa, Phys. Rev. B 81, 104527 (2010).

[10] G. A. Armstrong, A. A. Helmy, and A. S. Greenberg, Phys. Rev. B 20, 1061 (1979).

[11] J. M. Ziman, Electrons and Phonons (Oxford U. P., 1960).

[12] X. Lin, A. C. Clark, Z. G. Cheng, and M. H. W. Chan, Phys. Rev. Lett. 102, 125302 (2009).

[13] O. Syschenko, J. Day, and J. Beamish, Phys. Rev. Lett. 104, 195301 (2010).

[14] A. K. McCurdy, H. J. Maris, and C. Elbaum, Phys. Rev. B 2, 4077 (1970).

[15] H. J. Maris, private communication.

[16] P. C. Hendry and P. V. E. McClintock, Cryogenics 27, 131 (1987).

[17] A. Diessens, E. van der Poll, and I. F. Silvera, Phys. Rev. B 33, 3269 (1986).

[18] D. S. Greynall, Phys. Rev. B 16, 5127 (1977).

[19] C. A. Burns et al., Phys. Rev. B 78, 224305 (2008).

[20] C. G. Eddison and M. N. Wybourne, J. Phys. C 18, 5225 (1985).

[21] L. P. Mezhov-Deglin, Zh. Eksp. Theor. Fiz. 49, 66 (1965).