

Liquid ^3He Thermomechanical Effect in Regular Cylinders

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We have measured the normal liquid ^3He thermomechanical effect with the liquid confined in the regular 210 nm diameter cylinders of Anopore. The pressure difference resulting from the temperature difference was $\Delta P/\Delta T = 25 \text{ Pa/mK}$, independent of temperature, from 0.7 to 12 mK. This value was $100\times$ the theoretical value and $3\times$ the result with a packed powder constriction. Two monolayers of ^4He did not change the value, but four monolayers reduced it by 2 orders of magnitude; the high value was therefore attributed to boundary scattering from magnetic ^3He surface layers.

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Transport processes in a system couple: examples include the Seebeck thermoelectric effect, where a temperature gradient causes a voltage gradient, and the Knudsen thermomolecular effect in a gas, where a temperature difference between two chambers connected by a small orifice causes a pressure difference. It was recently discovered that this coupling of transport processes also applies to the quasiparticle gas of normal liquid ^3He [1,2], where it has been termed the thermomechanical effect (TME). When two chambers of normal liquid ^3He are connected by a channel with diameter d much smaller than the quasiparticle mean free path in the bulk, λ_{bulk} , a steady state pressure difference ΔP arises from a temperature difference ΔT between the chambers. In the theory of irreversible thermodynamics, one considers generalized fluxes J_i and forces X_i with coupling coefficients L_{ij} , and writes equations

$$J_1 = L_{11}X_1 + L_{12}X_2, \quad (1)$$

$$J_2 = L_{21}X_1 + L_{22}X_2. \quad (2)$$

In metals, J_1 is the heat current and J_2 the electrical current; in liquid ^3He , J_1 is also the heat current but J_2 is the mass current. Traditionally one defines the generalized forces such that $\sum_i J_i X_i = dS/dt$, the rate of entropy production. Then in liquid ^3He the conjugate forces are $X_1 = -\Delta T/T^2$ and $X_2 = -\Delta\mu/T$, where μ is the chemical potential related to P by the Gibbs-Duhem equation $n\Delta\mu = \Delta P - s\Delta T$; n is the number density, and s the entropy per unit volume. In the case of no mass flow ($J_2 = 0$), one has from (2)

$$\frac{\Delta\mu}{\Delta T} = -\frac{L_{21}}{L_{22}T}. \quad (3)$$

Calculating $-L_{21}/L_{22}$ involves solving the appropriate Boltzmann transport equation for $J_2 = 0$ and simultaneous gradients of T and μ . For electrons in metals the result is well known [3]; for the Landau quasiparticles of normal liquid ^3He Edwards *et al.* [2] found, in the collisionless limit $d \ll \lambda_{\text{bulk}}$,

$$\frac{\Delta P}{\Delta T} = \frac{s}{3} = 0.2T \text{ Pa/mK}, \quad (4)$$

where $s \propto T$ is the ^3He entropy per unit volume and T is in mK. Outside the collisionless limit (Knudsen regime), bulk Poiseuille flow ensures that the steady state condition is $\Delta P = 0$, as in a Maxwell gas.

Our previous measurements of the TME [1,4] were made using a packed powder constriction, with pores of diameter 90 nm. The measured TME was 7 Pa/mK at 2 mK, $15\times$ the theoretical value, and only weakly temperature dependent. This enhancement was shown to be due to magnetic scattering from the monolayers (ML) of ^3He on the surface of the packed powder. The first two ML of ^3He on a surface are solid and have a Curie-Weiss paramagnetism, rather than the much weaker Pauli paramagnetism of the bulk liquid; scattering from these magnetic atoms alters transport properties. When the solid ^3He was replaced by preplating with 4 ML ^4He , which is nonmagnetic and by virtue of its larger mass replaces ^3He in the surface layers, the observed TME was reduced to 0.2 Pa/mK at 2 mK, a reduction of $30\times$ from the result with pure ^3He and half the theoretical result. This behavior was reminiscent of that in metals, where electron scattering from magnetic impurities can lead to the giant thermoelectric effect [5].

In light of the disagreement with theory, the question remained, what was the effect of the geometry? Did using a packed powder, with its complicated geometry, change the result from that of the theory, with its cylindrical geometry? We therefore replaced the packed powder constriction with the 210 nm diameter cylinders in Anopore filters [6]. The cylinder diameter was much smaller than λ_{bulk} (given by $\lambda_{\text{bulk}}T^2 = 50 \mu\text{m} \cdot \text{mK}^2$ [7]) at low mK temperatures. SEM photos showed that the cylinders were uniform, parallel and cylindrical. The parallelism of the cylinders ensured that despite a porosity of 38% there was little intersection. A stack of eleven 60 μm thick disks was used, with each pair separated by a 50 μm ($\geq \lambda_{\text{bulk}}$) thick Mylar washer with an inner diameter of 3 mm. To prevent the Anopore (aluminum oxide) from pulling

away from the walls of the chamber on cooling, the Anopore disks were epoxied with Stycast 1266 into a 1266 holder, which was then glued with Stycast 2850GT into the copper chamber. No large hole existed in parallel with the cylinders, as shown in two ways: Measurements of liquid ^3He mass flow [8] were inconsistent with a large hole, and with pure ^3He the superfluid transition was suppressed to below our minimum temperature, consistent with theoretical predictions [9]. (Below the bulk superfluid transition temperature $T_c^{\text{bulk}} = 0.93$ mK, there was a ^3He superleak in four out of the eleven disks).

Figure 1 shows our apparatus, which consisted of two chambers of ^3He separated by the stack of Anopore disks. The lower chamber held 10 cm^3 ^3He , mostly in the pores of a silver sinter heat exchanger connected to our PrNi_5 refrigerant. A melting curve thermometer attached to this chamber served as our primary thermometer. The upper chamber contained 3.4 cm^3 ^3He with a heater and two thermometers immersed in it. The heater was a CuNi meander line evaporated onto a Homosil glass disk. One thermometer was a cylinder of powdered lanthanum-diluted cerous magnesium nitrate (LCMN), calibrated against the melting curve thermometer and used below 6 mK; extrapolation of the calibration according to the Curie-Weiss law enabled us to use the LCMN down to 0.4 mK. The other thermometer, used above 3 mK, was a $4\text{ mm} \times 4\text{ mm} \times 12\text{ }\mu\text{m}$ square of aluminized Mylar [10]; we measured the capacitance at 1 kHz with a bridge excitation of $0.25\text{ V}_{\text{rms}}$. The two chambers were separated by a 20 mm diam, $8\text{ }\mu\text{m}$ thick Kapton membrane, pre-stretched at room temperature by 0.5% and coated with 85 nm of gold. (The high thermal boundary resistance between ^3He and solids ensured there was no heat transfer through the Kapton.) This flexible diaphragm was one plate of a parallel plate capacitive differential pressure transducer [11]; the other plate was a solid copper disk held rigidly in the lower chamber. The pressure-capacitance relation of the diaphragm was determined

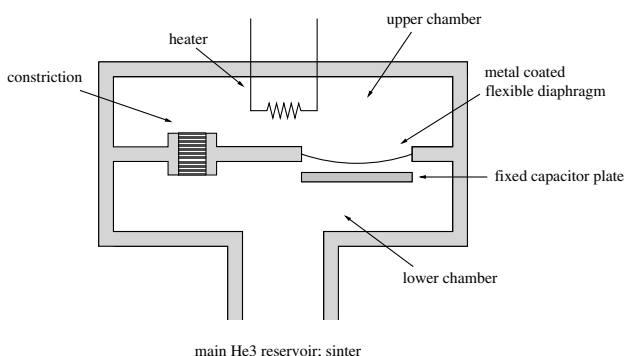


FIG. 1. The experimental apparatus, shown schematically, consisted of two chambers of ^3He separated by the stack of Anopore porous disks and a flexible diaphragm pressure transducer. The ^3He in the top chamber could be heated, and thermometers (not shown) were in or on both chambers.

electrostatically [11] at mK temperatures to be $\Delta P/\Delta C = 2.11 \times 10^{14}$ Pa/F, which agreed with the values determined from gas flow measurements at 4.2 K and the superfluid ^3He fountain effect. With a capacitance resolution of 10^{-6} pF, the pressure resolution was 2×10^{-4} Pa and the displacement resolution was <1 Å. The small volume change resulting from a pressure change, $\Delta V/\Delta P = 1.80 \times 10^{-11}$ m³/Pa, resulted in a mechanical time constant for ^3He flow through the Anopore of ~ 200 s, compared to 10 h in our previous experiments.

The experimental method was to adiabatically demagnetize the PrNi_5 , cooling to 0.4 mK, then letting the apparatus slowly warm. Heat pulses, typically 0.2–2 nW, were applied to the upper chamber for ~ 1 h, while monitoring the temperatures of both chambers and the pressure difference between them. The directly measured quantities were the pressure and temperature differences between the chambers resulting from a heat input, $\Delta P/\dot{Q}$ and $\Delta T/\dot{Q} \equiv R$, respectively; the TME, apart from a correction to R discussed below, was given by the ratio of these two quantities. The experimental time constant, a combination of the mass flow and thermal time constants, was sufficiently short (250 s at 1 mK) that a steady state could be reached before, during, and after each heat pulse. TME data were acquired over the temperature range 0.5–12 mK, with pure ^3He and also with 2.3 and 4.5 nominal ML of ^4He preplated to the surfaces. All measurements were done at saturated vapour pressure and in the Earth's field.

First we present the thermal resistance R between the two chambers. These data will be discussed more fully in a separate publication [12]; here we use them only as input to the calculation of the TME. R was composed of both the thermal resistance of the ^3He in the cylinders and that of the bulk ^3He above and below the constriction; for

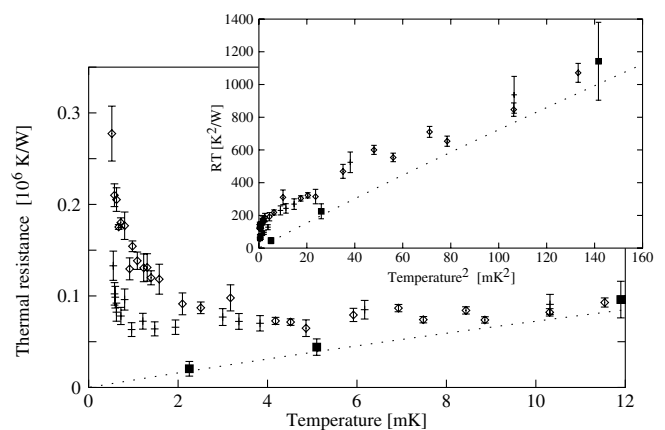


FIG. 2. Thermal resistance R between the two chambers vs temperature for the three different ^4He coverages. (\diamond): 0 ML; ($+$): 2 ML; (\blacksquare): 4 ML ^4He . Inset: Thermal resistance plotted as RT vs T^2 . The intercept gives the boundary-limited thermal resistance and the slope the bulk resistance. The line is the bulk component.

determination of the TME, it was necessary to isolate the thermal resistance due to boundary scattering in the cylinders, R_{bd} . The thermal conductivity in the cylinders was given by the gas-kinetic expression $\kappa = cv_F\Lambda/3$, where $c \propto T$ is the ^3He heat capacity per unit volume and v_F the Fermi velocity; $\Lambda = (1/\lambda_{\text{bulk}} + 1/d)^{-1}$ is the effective mean free path [13]. Because $\lambda_{\text{bulk}} \propto T^{-2}$, the thermal resistance in the cylinders was expected to be given by $a'T + b/T$, with a' and b constants and R_{bd} equal to b/T . This was added in series with the bulk thermal resistance above and below the cylinders, nearly proportional [14] to T , to obtain a total thermal resistance $R = aT + b/T$, or $RT = aT^2 + b$. Figure 2 (inset) is a plot of the measured RT vs T^2 for pure ^3He and with 2 and 4 ML ^4He . The main figure shows the same data plotted as R vs T . With pure ^3He and above 1 mK, the plot of RT vs T^2 is linear as expected, the slope (giving the bulk resistance) agreeing with the known geometry of the cell. Extrapolating the line above 1 mK to $T = 0$ gave $R_{bd}^0 T = 156 \pm 6 \text{ K}^2/\text{W}$, twice the value expected using the Anopore open area deduced from mass flow measurements [8]. Below 1 mK, the data deviated from the extrapolated line, with the measured values 30% lower than the line at 0.5 mK. With 2 ML ^4He , the data were on the same line, but the low temperature deviation was larger (R fell to 40% the extrapolated value) and began at 2.5 mK. To extract R_{bd} , the data were fit to the empirical relation

$$\begin{aligned} R(T)T &= aT^2 + R_{bd}^0 T[1 - \beta \exp(-T^2/T_0^2)] \\ &\equiv aT^2 + R_{bd}T. \end{aligned} \quad (5)$$

Physically, this corresponds to a coefficient $R_{bd}T$ that decreases at low temperatures. For pure ^3He we found $\beta = 0.3$ and $T_0^2 = 1.0 \text{ mK}^2$, and with 2 ML ^4He $\beta = 0.7$ and $T_0^2 = 4.6 \text{ mK}^2$. We note that at 1 mK, R_{bd} was 95% of the total resistance, so the effect of the correction for the bulk thermal resistance on the TME was small; however, at higher temperatures R became much larger than R_{bd} .

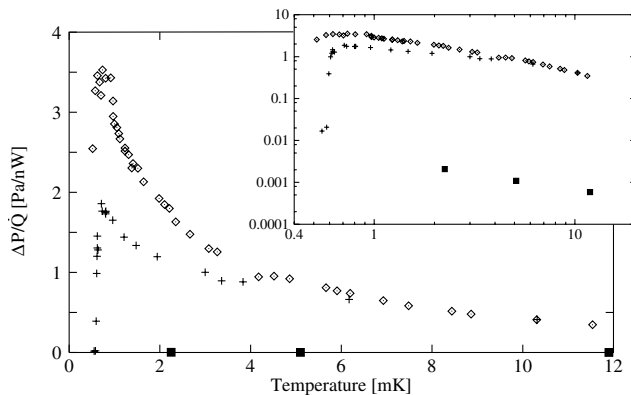


FIG. 3. Pressure difference ΔP across the constriction resulting from a heat input \dot{Q} , for 0 (\diamond), 2 (+), and 4 ML ^4He (\blacksquare). The inset shows the same data on a log-log plot.

With 4 ML ^4He the bulk resistance was unchanged from the 0 and 2 ML value but, because of the high degree of specular scattering (90%, determined from mass flow measurements [8]), R_{bd} was indistinguishable from zero. An independent confirmation of the thermal resistance was provided by the thermal time constant, approximately equal to RC_{top} ; because the heat capacity of the upper chamber C_{top} was known we could extract a thermal resistance from the time constant, in good agreement with the measured R .

Next we plot $\Delta P/\dot{Q}$, the pressure difference ΔP resulting from a heat input \dot{Q} , in Fig. 3. With pure ^3He , the data were on a smooth curve varying from 3.5 Pa/nW below 1 mK to below 0.5 Pa/nW at 12 mK. With 2 ML ^4He and above 4 mK, $\Delta P/\dot{Q}$ agreed with the pure ^3He result; below 4 mK $\Delta P/\dot{Q}$ was less than the result with pure ^3He , reduced by a factor of 2 at 1 mK. Below 0.6 mK with 2 ML of ^4He , the liquid was a superfluid in each disk and $\Delta P/\dot{Q}$, corresponding to the superfluid fountain effect, was much lower. With 4 ML ^4He , $\Delta P/\dot{Q}$ was reduced nearly 1000 \times , as seen in the inset. Because of a superleak in four out of the eleven Anopore disks, a correction has been applied to the normal fluid TME data below $T_c^{\text{bulk}} = 0.93 \text{ mK}$. Only the small superfluid fountain effect was present across those four disks; $\Delta P/\dot{Q}$ fell to 7/11 of its value above T_c^{bulk} . This drop was also observed in the mass flow time constant [8], but R was continuous across T_c^{bulk} as expected. We have therefore multiplied the $\Delta P/\dot{Q}$ results below 0.93 mK, except for below 0.6 mK with 2 ML ^4He where all the liquid was a superfluid, by 11/7.

The TME, given by $\Delta P/\Delta T_{bd} = (\Delta P/\dot{Q})/R_{bd}$, is plotted in Fig. 4. The present results with 0 and 2 ML ^4He are shown as (\diamond) and (+), respectively; the data sets are the same, nearly constant at 25 Pa/mK from 0.7 to 12 mK. Even though $\Delta P/\dot{Q}$ was reduced by a factor of 2 at 1 mK by adding 2 ML ^4He , this change was exactly canceled by

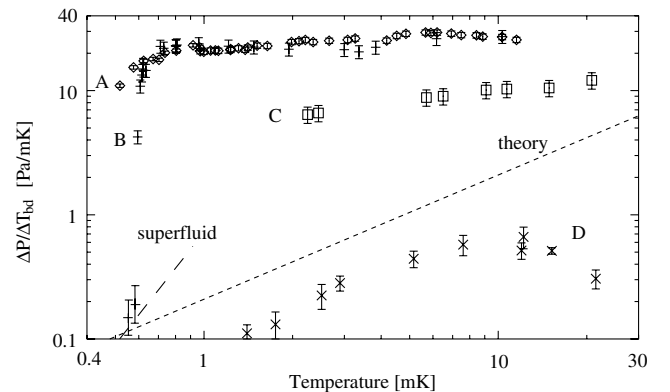


FIG. 4. Thermomechanical effect $\Delta P/\Delta T_{bd}$. A and B are the present results with Anopore cylinders with 0 and 2 ML ^4He , respectively; C and D are the results with a packed powder constriction with 0 and 4 ML ^4He . The theory is due to Edwards *et al.* [2]; the line labeled “superfluid” is the expected superfluid fountain effect.

the change in R_{bd} . The results with the Anopore constriction are greater than both the results with the packed powder [by $3\times$; shown as (\square) for pure ^3He] and the theoretical result ($100\times$ at 1 mK). The geometries of the Anopore cylinders and packed powder pores are very similar and dissimilar, respectively, to the theoretical geometry of a circular cylinder; because both data sets are much greater than the theory, the measurements rule out geometry as the cause of the increase from the theoretical value. Contrary to our previous expectations, the high value of the TME was not altered by the 2 ML ^4He . The actual amount of ^4He on the surface of the cylinders was unknown, however; the reduction by up to a factor of 2 in R , $\Delta P/\dot{Q}$, and the mass flow rate [8] showed that *some* ^4He was present in the cylinders. Conversely, the further reductions in the same coefficients on the addition of two more ML suggested that some solidlike ^3He remained with 2 ML ^4He . We note the similarity of our mass flow rate results [8] with the cylinders to those between parallel planes [15] suggests that the ML coverage was correct. With 4 ML ^4He , we could not extract R_{bd} from the thermal resistance measurements, and therefore could not directly determine the TME. If we speculate that R_{bd} was reduced by the same factor of 15 as the mass flow time constant on going from 0 to 4 ML [8], we may use the measured $\Delta P/\dot{Q}$ to find a TME close to the theoretical result. Therefore, only a small amount of magnetic scattering is necessary for the large TME, but further magnetic scattering does not increase the TME further. If we assume that the reduction in TME due to backscattering in the packed powder is the same with 4 ML ^4He as with pure ^3He , the measured 4 ML ^4He packed powder TME [shown as (\times) in Fig. 4] is the correct amount below the theoretical value for the theory to be correct. (We were able to measure a TME with 4 ML ^4He in the packed powder only because geometric backscattering created an R_{bd} ; in the Anopore cylinders, this backscattering was not present and R_{bd} was too small to be measured.)

An important check on our results was provided by the superfluid ^3He fountain effect, where $\Delta P/\Delta T$ equals the

superfluid entropy per unit volume. Below 0.6 mK with 2 ML ^4He we observed the correct value [16], showing that the pressure and thermometry calibrations were correct. The suppression of the superfluid transition with pure ^3He to below our minimum temperature was consistent with theory [9], as was the increase in T_c with the increase in specularity with 2 ML ^4He .

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