

## Unexpected observation in measurements of transport coefficients in $^3\text{He}$ - $^4\text{He}$ mixtures near $T_\lambda$

J. N. Shaumeyer\*

*Department of Physics, Wesleyan University, Middletown, Connecticut 06547*

R. P. Behringer

*Department of Physics, Duke University, Durham, North Carolina 27706*

(Received 23 December 1985)

Measurements of the shear viscosity  $\eta$  and thermal conductivity  $\kappa$  in four  $^3\text{He}$ - $^4\text{He}$  mixtures having  $^3\text{He}$  concentrations  $X \leq 0.09$  indicate different  $\lambda$ -transition temperatures for  $\eta$  and  $\kappa$  at the same  $X$ . Absent in additional data on pure  $^4\text{He}$ , this difference in transition temperatures increases with  $^3\text{He}$  concentration. We tentatively attribute the effect to an unexpectedly large enhancement of the  $^4\text{He}$  concentration near the walls of either the viscometer or the thermal conductivity cell.

During an experiment performing simultaneous measurements of the thermal conductivity  $\kappa$  and the shear viscosity  $\eta$  of dilute  $^3\text{He}$ - $^4\text{He}$  mixtures near the superfluid transition, we obtained an unexpected result: For a given concentration, the transition indicated by the viscosity measurements occurred at a higher temperature than that determined in thermal conductivity measurements. Five mixtures were studied, with  $^3\text{He}$  molar concentrations of  $X=0.0000$ ,  $X=0.0127$ ,  $X=0.0491$ ,  $X=0.0893$ , and  $X=0.0916$ . This article documents the effect, giving the relevant experimental details, and explores the possible causes of the discrepancy.<sup>1</sup>

A schematic of the experiment is given in the inset of Fig. 1. The thermal conductivity measurements used a parallel-plate geometry,<sup>2</sup> and the data were obtained by measuring the temperature difference across the 0.5-mm-high cell induced by a steady heat flux. Particularly near the superfluid transition,  $T_\lambda$ , very small temperature differences ( $< 50 \mu\text{K}$ ) were used in order to avoid finite heating effects.

The viscosity was deduced by measuring the damping on a cylindrical beryllium-copper torsional pendulum<sup>3</sup> which contained the helium. To increase the viscous damping, the interior of the pendulum was filled with a 1-cm-high stack of 14 aluminum disks, 2 cm in diameter. The gap between adjacent disks was  $250 \mu\text{m}$ . All interior surfaces of the pendulum were polished to an optically reflecting finish. The empty pendulum had a mechanical  $Q$  of  $1.5 \times 10^5$  at low temperatures; when filled with liquid, the  $Q$  was  $1.3 \times 10^4$  and its estimated power dissipation was 1 nW. Driving the pendulum electrostatically on resonance, we determined that it had a linear response by varying the amplitude over a broad range. This linearity was found both when the cell was empty and when it was full. The oscillation frequency  $f=273$  Hz was made low on the premise that the viscous penetration depth,  $L_p=(\eta/\pi\rho_n f)^{1/2}$ , should be as large as possible to avoid departures from hydrodynamic behavior near  $T_\lambda$ . Here  $\rho_n$  is either the density of the normal fluid below  $T_\lambda$  or simply the density above  $T_\lambda$ . In the present case, the viscous penetration depth was  $L_p=4.5 \mu\text{m}$ , small compared to usual experimental dimensions, but greater than the correlation length over all but a very narrow temperature range,  $|\epsilon|=|(T-T_\lambda)/T_\lambda| < 10^{-5}$ .

The sample spaces of the conductivity cell and viscometer

had a common midplane and were mounted on an isothermal sample chamber filled with the liquid mixture at saturated vapor pressure. After filling, the sample chamber was sealed by a cold valve. The conductivity cell and viscometer were connected by copper capillaries to this shared reservoir of fluid.

The measured damping on the viscometer was proportional to  $(\rho_n \eta)^{1/2}$ . Accordingly, the results are presented in the form  $(\rho_n \eta)/(\rho_n \eta)_\lambda$ , where the denominator, unique to each mixture, normalizes the data to unity at the temperature of the superfluid transition determined by the thermal conductivity. Figure 1 shows the results for  $X=0.0916$

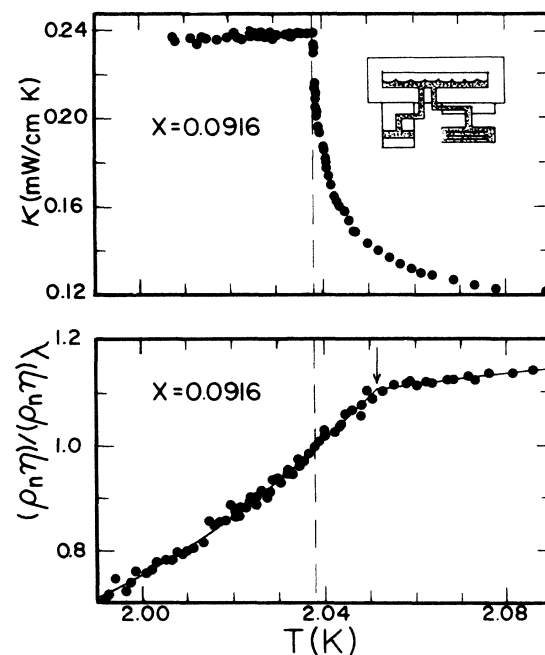


FIG. 1. Values of the thermal conductivity,  $\kappa$ , and (shear viscosity)  $\times$  (normal density),  $\rho_n \eta$ , for a mixture with  $X=0.0916$ . The dashed line is  $T_\lambda$  determined by the singularity in  $\kappa$ ; the arrow marks the  $T_\lambda$  singularity in  $\rho_n \eta$ . The inset shows a schematic of the thermal conductivity cell (left), viscometer (right), and mixture reservoir.

over a narrow temperature range near  $T_\lambda$ , demonstrating the precision with which  $T_\lambda$  can be determined for each type of measurement, and clearly displaying the difference in observed  $\lambda$  temperatures. The thermal conductivity yields a  $\lambda$  temperature determined to better than  $50 \mu\text{K}$ . The  $\rho_n\eta$  data, which are more weakly singular, have a  $\lambda$  temperature which is correspondingly less well determined; the uncertainty in  $T_\lambda$  is  $\sim 2 \text{ mK}$ , which is nevertheless small compared to the difference  $\Delta T_\lambda$  between the transition temperatures for  $\rho_n\eta$  and  $\kappa$ . Figure 2 concentrates on the region near the superfluid transition, showing  $\rho_n\eta$  for all five mixtures superposed so that their respective values for  $T_\lambda$ , given by the thermal conductivity, coincide. Striking is the displacement to higher temperatures of the  $\lambda$ -point singularity in  $(\rho_n\eta)/(\rho_n\eta)_\lambda$  indicated by the arrows, as the concentration increases. The solid lines in Fig. 2 are the results of least-squares fits of the form

$$\frac{(\rho_n\eta)}{(\rho_n\eta)_\lambda} = A + B^-|\epsilon|^x + C^-|\epsilon|^\zeta + D^-|\epsilon|^{x+\zeta} \quad (1)$$

for  $T < T_\lambda$

and

$$(\rho_n\eta)/(\rho_n\eta)_\lambda = A + B^+|\epsilon|^x \quad \text{for } T > T_\lambda \quad (2)$$

These are based on the expectation<sup>4-6</sup> that near  $T_\lambda$ ,  $\eta - \eta_\lambda \propto |\epsilon|^x$  and  $\rho_n - \rho_{n\lambda} \propto |\epsilon|^\zeta$ , where we fixed the viscosity

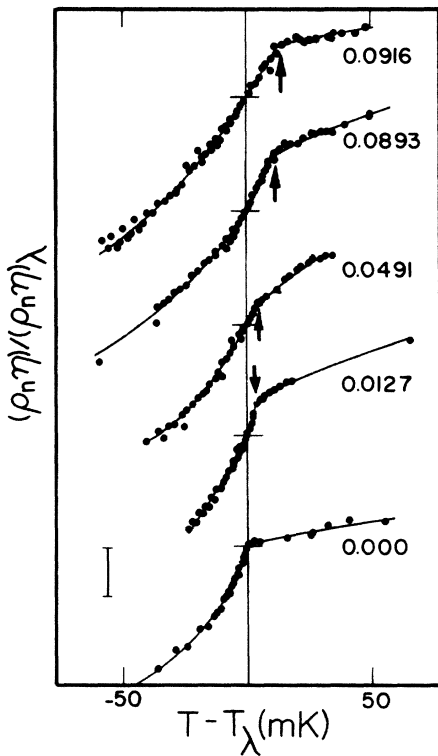


FIG. 2. Values of  $(\rho_n\eta)/(\rho_n\eta)_\lambda$  for all five mixtures as a function of temperature. The vertical line marks  $T_\lambda$  determined for each mixture by the thermal conductivity, where each set of data is normalized to unity; arrows mark the singularity associated with the superfluid transition in  $\rho_n\eta$ . Solid lines are least-squares fits. The vertical bar corresponds to a change of 0.1 in the vertical coordinate.

exponent at  $x=0.8$ , and the superfluid density exponent at  $\zeta=0.68$ . In Eqs. (1) and (2),  $T_\lambda$  in  $\epsilon$  refers to the superfluid transition temperature seen by the viscometer, and we have assumed that  $x$  is the same above and below  $T_\lambda$ . Here, the point of the fits is to determine the viscosity transition temperature. In principle,  $x$  may be different above and below  $T_\lambda$ . However, reasonable variations of  $x$  do not appreciably affect the determination of  $T_\lambda$  for  $\rho_n\eta$ . Details of the fits will be given elsewhere. Note, in reference to Fig. 1, that  $A \geq 1$ . As shown in Fig. 3,  $\Delta T_\lambda$  is a monotonically increasing function of  $^3\text{He}$  concentration, and vanishes for  $X=0$  (pure  $^4\text{He}$ ).

To our knowledge, the only previous measurements of viscosity near the superfluid transition in helium mixtures are those of Webeler and Allen.<sup>7</sup> Using a quartz rod torsionally excited at 11 kHz as a viscometer, they measured  $\rho_n\eta$  in mixtures with  $X=0.005, 0.047,$  and  $0.100$ . However, their experiment lacked a precise method to determine  $T_\lambda$  independently of the viscosity measurements, so they would have been unlikely to detect the effect we have described. We note that for  $X=0$  our results for  $\rho_n\eta$  agree within experimental resolution with recent results<sup>4</sup> obtained very near  $T_\lambda$ , i.e., for  $|\epsilon| \leq 10^{-2}$ ; further from  $T_\lambda$  our values differ by  $\sim 5\%$  from older results tabulated by Barenghi, Lucas, and Donnelly.<sup>8</sup>

Gravitational or heating effects in the experiment, which might account for our observations by creating concentration differences between the viscometer and thermal conductivity cell, can be dismissed either because of their magnitude or in consideration of the experimental design.

(1) *Gravitational effects.* The midplanes of the liquid in the thermal conductivity cell and viscometer coincided, ensuring the same average concentration in both, since they shared the sample reservoir. In principle, gravitationally induced concentration gradients existed and were larger in the viscometer because it was taller. But in this worst case, the gravity concentration gradient is calculated to be less than  $10^{-5}$  in  $X$  over the height of the viscometer, and therefore negligibly small. Centrifugal effects in the viscometer could also be considered, but these accelerations were much smaller than that of gravity.

(2) *Heating effects.* Since they involved thermal dissipation, both the thermal-conductivity cell and viscometer were very slightly warmer ( $\sim \mu\text{K}$ ) than the main germanium thermometer mounted on the sample reservoir, causing the superfluid onset in both the viscometer and thermal con-

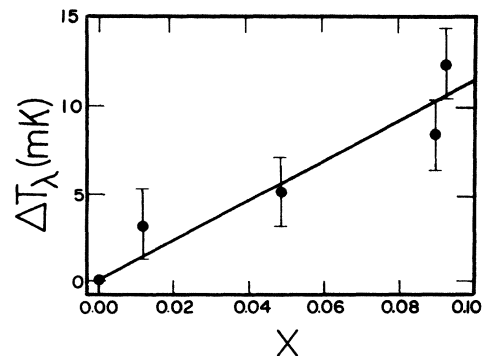


FIG. 3.  $\Delta T_\lambda$  as a function of concentration. The line has a slope of 0.13 K, and passes through the origin.

ductivity cell to appear at a slightly low ( $\sim \mu\text{K}$ ) temperature on the main thermometer. However, due to heat flush, the warmer sections suffered a slight reduction of  $^3\text{He}$  concentration. Since decreasing the  $^3\text{He}$  concentration increases  $T_\lambda$  (again by  $\sim 1 \mu\text{K}$ ), the consequences of thermal dissipation were in competition.

For the thermal-conductivity measurements, the mean temperature of the layer of sample was known; thus, only the reduction in  $X$  from heat flush is relevant, an effect which, taken alone, would reverse the sign of  $\Delta T_\lambda$ . For the viscosity measurements, the temperature of the sample was known only through the main thermometer, but an increase by  $\delta T$  of the temperature of the viscometer over that of the reservoir would again lead to negative values of  $\Delta T_\lambda$  by an amount

$$\Delta T_\lambda = \delta T [(dT_\lambda/dX)(\delta X/\delta T) - 1] \approx -0.5\delta T.$$

That is, the elevation of  $T_\lambda$  by heat flush cannot compensate for the low determination of  $T_\lambda$  caused by  $\delta T$ . Here  $\delta X$  is the (negative) change in concentration in the viscometer associated with a (positive) change in  $\delta T$ , and  $dT_\lambda/dX < 0$  is the slope of the superfluid transition line. In the normal phase,  $\delta X = -k_T(\partial X/\partial c)\delta T$ , where  $k_T$  is the thermal diffusion ratio and  $c$  is the  $^3\text{He}$  mass concentration; in the superfluid phase,  $k_T$  is replaced by the effective thermal diffusion ratio  $k_T^*$ . Values of  $k_T$  and  $k_T^*$  from recent experimental data<sup>9</sup> are positive and less than 0.6 in our experimental range. The quantities  $k_T$  or  $k_T^*$  would have to be larger than 1.1 to give a positive  $\Delta T_\lambda$ . Thus, heating effects in either part of the instrument, which are expected to be quite small, would produce an effect opposite in sign to the observed value of  $\Delta T_\lambda$ . As a final point, we emphasize that the oscillation amplitude of the liquid-filled viscometer was linear over three orders of magnitude in the driving amplitude, both above and below  $T_\lambda$ . Since viscous heating effects depend on the square of the oscillation amplitude, this observation directly demonstrates that heating effects in the viscometer are not relevant.

After disregarding the above possibilities, we tentatively conclude that the difference in transition temperatures is related to a surface effect, although the physical mechanism is not entirely clear. If the effect occurred in the viscometer, the observed  $\Delta T_\lambda$  would imply a depletion of the  $^3\text{He}$  for each concentration by approximately  $0.1X$  near the walls of

the viscometer, persisting over a sizable portion of a viscous penetration depth. It is well known that  $^4\text{He}$  is preferentially attracted to solid surfaces,<sup>10-15</sup> particularly through recent experiments on thin films of  $^3\text{He}$ - $^3\text{He}$  mixtures. In thin films, however, the  $^4\text{He}$  enhancement is limited to about  $0.005 \mu\text{m}$  near the wall. Nevertheless, there exist other surface and/or finite-size effects which may be related to the present case. Several experiments<sup>11,12</sup> have demonstrated the formation of a superfluid film on surfaces adjacent to bulk  $^3\text{He}$ - $^4\text{He}$  mixtures at temperatures higher than the  $\lambda$  transition temperature, although only for  $X > 0.55$ . The present measurements are unique in indicating surface phenomena for  $X < 0.55$ . The size of the effect is also striking. If a  $^4\text{He}$ -rich fluid layer of thickness  $\delta = 0.005 \mu\text{m}$  formed near the walls one would anticipate only a fractional contribution of order  $\delta/L_p = 0.001$  to the damping from this layer which would be insufficient to explain our observations. Moreover, previous observations of film formation adjacent to bulk fluid were generally static measurements; the role of the frequency at which transport measurements are made and the effect of shearing have yet to be determined. We note that in the presence of heat flow, superfluid hydrodynamics allows a boundary layer near a solid surface,<sup>16</sup> but the nature of this layer is not well defined, nor is it clear that it should influence either of the present measurements.

To summarize, we have documented a previously unobserved difference between the superfluid transition temperatures obtained by static measurements of the thermal conductivity versus dynamic measurements of the shear viscosity in dilute  $^3\text{He}$ - $^4\text{He}$  mixtures. Data for the latter indicate a higher transition temperature than the former, an effect which apparently cannot be explained by gravitational or heating effects. We tentatively attribute the difference to an unexpectedly large surface effect. Elucidating its physical cause remains an interesting challenge.

We gratefully acknowledge detailed comments by Professor G. Ahlers, Professor F. Gasparini, and Professor H. Meyer, and Dr. B. Law. This work was done while J.N.S. was at Duke University and was supported by the National Science Foundation under Low Temperature Physics Grant No. DMR-8314673. One of us (R.P.B.) acknowledges support from the Alfred P. Sloan Foundation.

\*Present address: Institute for Physical Science and Technology, University of Maryland, College Park, MD 20742.

<sup>1</sup>A brief account of these results has been given previously: J. N. Shaumeyer and R. P. Behringer, *Bull. Am. Phys. Soc.* **29**, 697 (1984). Further details can be found in J. N. Shaumeyer, Ph.D. dissertation, Wesleyan University (published by University Microfilm, 300 N. Zeeb Rd., Ann Arbor, MI 48106); J. N. Shaumeyer and R. P. Behringer (unpublished).

<sup>2</sup>The design is the same as that used in this laboratory for measurements on convecting liquid helium: see H. Gao, G. Metcalfe, T. Jung, and R. P. Behringer (unpublished).

<sup>3</sup>D. J. Bishop and J. D. Reppy, *Phys. Rev. B* **22**, 5171 (1978).

<sup>4</sup>L. Bruschi, G. Mazzi, M. Santini, and G. Torzo, *J. Low Temp. Phys.* **29**, 63 (1977).

<sup>5</sup>R. Biskeborn and R. W. Guernsey, Jr., *Phys. Rev. Lett.* **34**, 455 (1975).

<sup>6</sup>G. Ahlers, *Phys. Rev. A* **10**, 1670 (1974).

<sup>7</sup>R. W. Webeler and G. Allen, *Phys. Rev. A* **5**, 1820 (1972).

<sup>8</sup>C., F. Barenghi, P. G. J. Lucas, and R. J. Donnelly, *J. Low Temp. Phys.* **44**, 491 (1981).

<sup>9</sup>D. Gestrich, R. Walsworth, and H. Meyer, *J. Low Temp. Phys.* **46**, 407 (1982).

<sup>10</sup>D. R. Brewer and J. R. G. Keyson, *Phys. Lett.* **1**, 5 (1962).

<sup>11</sup>G. Ruppeiner, M. Ryschkewitsch, and H. Meyer, *J. Low Temp. Phys.* **41**, 179 (1980).

<sup>12</sup>J. P. Romagnan, J. P. Laheurte, J. C. Noiray, and W. F. Samm, *J. Low Temp. Phys.* **30**, 425 (1978); J. C. Noiray, D. Sornett, J. P. Romagnan, and J. P. Laheurte, *Phys. Rev. Lett.* **53**, 2422 (1984).

<sup>13</sup>G. Agnolet, S. L. Teitel, and J. D. Reppy, *Phys. Rev. Lett.* **47**, 1537 (1981).

<sup>14</sup>F. M. Ellis, R. B. Hallock, M. D. Miller, and R. A. Guyer, *Phys. Rev. Lett.* **46**, 1461 (1981); F. M. Ellis and R. B. Hallock, *Phys. Rev. B* **29**, 498 (1984).

<sup>15</sup>M. J. Dipirro and F. M. Gasparini, *Phys. Rev. Lett.* **44**, 269 (1980); B. Bhattacharja and F. M. Gasparini, *ibid.* **49**, 919 (1982).

<sup>16</sup>R. P. Behringer, *J. Low Temp. Phys.* **62**, 15 (1986).