Thermal Transport in Very Dilute Mixtures of ^{3}He in ^{4}He near the Superfluid Transition

M. Dingus, F. Zhong, and H. Meyer
Department of Physics, Duke University, Durham, North Carolina 27706
(Received 20 February 1985)

We report measurements of the temperature difference across layers of liquid ^{3}He-^{4}He mixtures in the superfluid phase as produced by a heat current at temperatures very close to $T_\lambda(X)$. Here X is the ^{3}He molar concentration that covers the range $2\times10^{-9} < X < 2\times10^{-2}$. The results are analyzed in terms of contributions from the boundary and the bulk fluid resistivities R_K and $\kappa_{\rm eff}^{-1}(X)$. Assuming $R_K(X)$ to be independent of X, we find that for $X < 10^{-3}$ there is a systematic departure from the prediction $\kappa_{\rm eff} \propto X^{-1}$. The thermal-relaxation times are consistent with this observation. An alternative analysis invoking an anomalous boundary resistance is presented.

PACS numbers: 67.60.−g

The singular behavior of the transport properties in dilute mixtures of ^{3}He in ^{4}He near the superfluid transition $T_\lambda(X)$ has recently received increased attention.[1-5] Here we are concerned with mixtures where the mole fraction X of ^{3}He is smaller than ≈ 0.05, and this paper describes measurements of the thermal conductivity and of the thermal-relaxation time down to $X \simeq 2.5\times10^{-9}$, extending previous conductivity experiments[1] by about four orders of magnitude in X. Unexpected results were obtained which we report here.

In the asymptotic region $X \to 0$, for the normal phase, the result of several predictions[2-4] for the conductivity κ at T_λ is that $\kappa_\lambda(X) \propto X^{-1}$. From the calculations of Dohm and Folk[3] and of Onuki,[4] this regime extends roughly up to $X \simeq 10^{-2}$. A closed-form expression is due to Onuki,[4] who showed that $\kappa_\lambda(X) = AS_\lambda^2(X)X^{-1}$, where $S_\lambda(X)$ is the entropy at $T_\lambda(X)$ and A is proportional to the mass-diffusion coefficient of an isolated ^{3}He particle in ^{4}He. With diminishing X, it becomes increasingly difficult to estimate the experimental value of κ_λ by extrapolation as $T_\lambda(X)$ is approached from the normal phase. From the predicted behavior[3] we conclude that κ_λ cannot be obtained by extrapolation for $X < 10^{-3}$, even for the highest temperature resolution achieved[6,7] so far, $\delta T \simeq 10^{-9}$ K.

By contrast, extrapolating the thermal conductivity at T_λ from the superfluid side appears to be simple, since there the conductivity is found[5] to be independent of $\epsilon = (T - T_\lambda)/T_\lambda$ for $3\times10^{-6} < -\epsilon < 10^{-2}$, where $-\epsilon = 3\times10^{-6}$ is our experimental range limit. One must also assume no singular behavior beyond this limit. In the superfluid phase one writes $\kappa_{\rm eff}$, the effective bulk conductivity, that includes both the heat transport by convection (κ_m) and by diffusion (κ), in contrast to the normal phase, where thermal diffusivity is the only transport mechanism. The theory of Khalatnikov[8] for dilute superfluid mixtures can be written in the form

$$\kappa_{\rm eff} = \kappa + \kappa_m = \kappa + \frac{\rho D k_{\rm B}}{cm_3}(k_T^* - k_T^2), \qquad (1)$$

where $k_T^* \approx (m_3\sigma_0/k_{\rm B}) + c$, $c \simeq m_3X/m_4$ is the mass concentration, D is the mass-diffusion coefficient of the ^{3}He impurities in ^{4}He, ρ is the mass density, σ_0 is the entropy per unit mass of pure ^{4}He, m_3 and m_4 are the isotopic masses, and k_T is the thermal-diffusion ratio. Khalatnikov[8] states that for sufficiently dilute mixtures, $\kappa_m > \kappa$. From measurements by Ptukha,[9] this appears to be the case near T_λ, and hence one would expect $\kappa_{\rm eff} \propto c^{-1} \propto X^{-1}$. Because k_T^* is only very weakly dependent on X and on ϵ for dilute mixtures near T_λ, this expectation assumes tacitly that D is only weakly dependent on c and that $k_T \ll k_T^*$, except possibly very close to T_λ. Should $\kappa_{\rm eff} = \kappa$ at T_λ, it would follow that $k_T^* = k_T$ at T_λ.

Recently Folk and Iro[10] have calculated $\kappa_{\rm eff}$ for dilute mixtures, and their results show that for sufficiently dilute mixtures, $\kappa_{\rm eff} \propto c^{-1}\sqrt{C_p}$. Because no discontinuity in the thermal conductivity is expected[10] at $T_\lambda(X)$, the predictions for $\kappa(T_\lambda)$ (Refs. 3 and 4) can be compared with experimental data extrapolated from the superfluid phase. However, we note that the predicted dependence[10] of $\kappa_{\rm eff}$ on ϵ (namely, $\kappa_{\rm eff} \propto \sqrt{C_p}$) is inconsistent with observations.[5]

We have carried out measurements of the thermal transport through a horizontal cylindrical layer of liquid helium of thickness $l = 0.15$ cm and diameter 2.68 cm. The heat current Q was directed upwards to minimize the convection currents caused by gravity and was in the range $30 < Q < 120$ ergs/cm$^2\cdot$s. The measurements of the temperature difference across this layer were made via two conventional germanium thermometers thermally attached to the top and the bottom flat copper pieces that are the boundaries of the fluid. Therefore, the measured temperature drop ΔT (with the small contribution from the stainless-steel spacer removed) included twice the contributions from R_K, the liquid-copper boundary resistivity, plus that of the copper between the thermometers and the boundary, and from $\kappa_{\rm eff}^{-1}$, namely

$$r \equiv \frac{\Delta T}{Q} = 2R_k + \kappa_{\rm eff}^{-1} l. \qquad (2)$$

Our experiments were carried out under conditions

of saturated vapor pressure, and the procedures were essentially the same as in Ref. 5. The superfluid transition was located before each conductivity measurement to within 0.3 μK. Because in the absence of a perturbation there were no observable drifts of the thermometer with time, it was possible to measure the temperature shifts in T_λ after dilution for δX as small as 10^{-6}. Four series of thermal transport measurements with progressive dilutions were carried out. The first three started with ordinary helium from the Amarillo fields ($X \simeq 2\times 10^{-7}$) and the last with specially purified ^{4}He with $X = 2.4\times 10^{-9}$. After the measurements for a given X were completed, a small, well-determined fraction (of the order of 3%) of the sample was rapidly evaporated from the cell into a Toepler pump, and in its place the same volume of an appropriate standard mixture with a much higher concentration was added to the sample, leading to a mixture with known composition greater than before. Corrections were made to take into account the ^{3}He evaporated from the cell. It was estimated that the uncertainty of the mixture composition was less than 3%.

In the inset of Fig. 1, we show the time dependence of the temperature change ΔT for a representative sample with $X = 5.3\times 10^{-7}$. After the heat flux is switched on, the temperature change at the cell bottom with respect to the cell top rises in a matter of less than ~ 30 s, followed by a second, much slower rise, $\Delta T_B(t)$, as a function of time t. We have tentatively attributed ΔT_K to the boundary resistance and ΔT_B to the bulk-fluid resistance. This interpretation is made plausible because long relaxation times are expected for the bulk fluid (see below). The approach to the steady-state conditions is via a diffusive process, so that $\Delta T_B(t)$ is to be described by an infinite sum of terms from modes with different relaxation times.

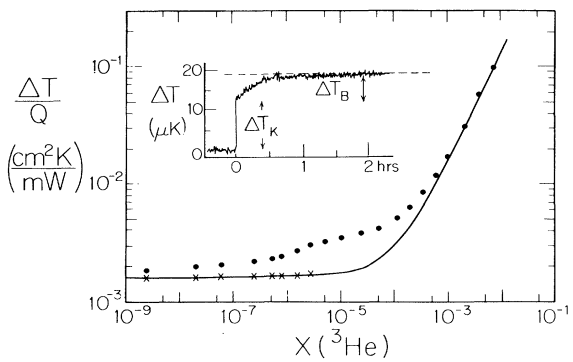

FIG. 1. The ratio $\Delta T/Q$ for $-\epsilon < 10^{-2}$ vs X for one dilution series. Symbols are the experimental data: Solid circles, $\Delta T/Q$; crosses, $\Delta T_K/Q$. Solid line: calculation via Eq. (2) as discussed in text. Inset: representative temperature change ΔT vs time, as heat is switched on, for $X = 5.3\times 10^{-7}$. The top dashed line expresses the steady state, reached after about 2 h.

The slowest mode with relaxation time τ is measured to a good approximation from the time dependence of $\Delta T_B(t)$ when $[\Delta T_B(\infty) - \Delta T_B(t)]/\Delta T_B(\infty) \lesssim \frac{1}{3}$, which decays exponentially with a single time constant. The steady-state value $\Delta T_B(\infty)$ for the particular example in Fig. 1 is reached after about 2 h and is indicated by a dashed line. At least for $X < 10^{-5}$, $\Delta T_K/Q$ and hence R_K were found to be independent of X, as shown by the crosses in Fig. 1. At higher X, the separation into a fast and a slow change in T became progressively less sharp possibly because the amplitudes of the fast modes become larger, but our subsequent analysis assumes that R_K is independent of X for all dilute mixtures.

In the normal phase for $X < 10^{-6}$, the data for κ were found to be consistent within experimental error with previous results obtained with a cell of $l = 0.122$ cm.[5] In the superfluid phase we found that, within experimental scatter, the ratio r had no temperature dependence for $t < 10^{-2}$, and also for $X > 10^{-6}$ it did not depend on the heat flux used. However, for $X < 10^{-6}$, there appeared to be nonlinear effects beyond a certain heat flux Q_c, and we only present here data for $Q < Q_c$. Under our experimental conditions, the superfluid velocity V_s was calculated to be always much smaller than the critical velocity $V_{s,c}$. In Fig. 1 we show by solid circles the ratio r for the one of four dilution series which extended over the largest number of decades in X. There were some slight differences in the value of R_K between the various dilution series, no doubt caused by the cryostat warmup between these series. The solid line in Fig. 1 is the ratio r calculated via Eq. (2) under the assumption of a dependence $\kappa_{\rm eff} \propto X^{-1}$ (obtained by extrapolation of the data below $X = 10^{-3}$) and $R_K = 8.3\times 10^{-4}$ cm^2 K/mW that is consistent with the crosses. The difference from the measurements is quite striking.

The data analysis was made on the assumption that R_K is independent of X and determined from the results for the mixtures below $X < 10^{-5}$. From the data for $r(T_\lambda, X)$ we calculate $\kappa_{\rm eff}(T_\lambda, X)$ via Eq. (2), and the result is shown in Fig. 2. We also show the results of Tanaka, Ikushima, and Kawaski,[1] presumably obtained with the same assumptions, and their data are consistent with ours, showing the departure from a X^{-1} dependence for $X < 10^{-3}$. This differs from the point at $X = 1.4\times 10^{-4}$ obtained by extrapolation of Ptukha's data.[9] Also shown are the data of Refs. 5 and 12 obtained in this laboratory. Because no discontinuity is predicted[10] at T_λ, we compare the data with predictions by Onuki[4] for $\kappa_\lambda(X)$ fitted to the data for $X > 10^{-3}$ by a single parameter.

The characteristic times τ of the slowest relaxation mode in the superfluid phase mentioned before are presented in Fig. 3, and they can be understood in terms of the hydrodynamic theory by Khalatnikov.[8] It

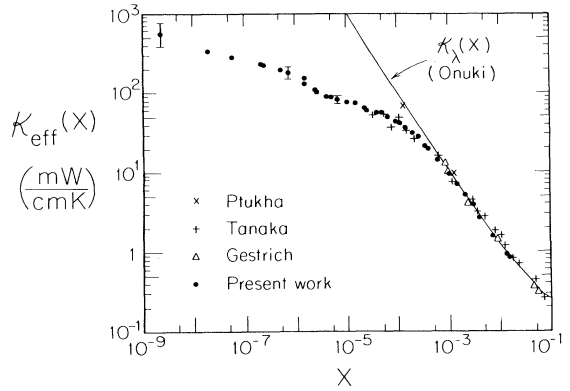

FIG. 2. The limiting conductivity $\kappa_{\rm eff}(X)$ for $-\epsilon < 10^{-2}$ vs X, including the results of four dilution series. The data by Tanaka and Ikushima (Ref. 1), Ptukha (Ref. 9), and Gestrich and co-workers (Refs. 5 and 12) are shown.

can be shown that the expression for τ derived by Behringer and Meyer reduces in the situation for very dilute mixtures ($X < 10^{-2}$) to the equation

$$\tau = \left(\frac{l}{\pi}\right)^2 \frac{k_B \rho (k_T^*)^2}{m_3 \kappa_{\rm eff} c}. \quad (3)$$

Because $\kappa_{\rm eff}$ and k_T^* are practically independent of ϵ for $-\epsilon < 10^{-2}$, we also expect τ to be so, which is in agreement with experiment. Using the value $k_T^* = 0.57$ expected from theory[8] for vanishing X, and also the measured values for the apparent $\kappa_{\rm eff}$, we obtain the solid curve in Fig. 3, and there is semiquantitative agreement with the data down to concentrations of $X \simeq 10^{-5}$. At lower concentrations, a faster relaxation mechanism than predicted by the solid curve appears to dominate. A constant relaxation time (dashed curve in Fig. 3) would have been observed as a function of X, according to Eq. (3), had the relation $\kappa_{\rm eff} \propto X^{-1}$ been valid at concentrations $X < 10^{-3}$.

An alternative approach for the analysis of the $\Delta T/Q$ data in Fig. 1 consists in ignoring the evidence of Fig. 1 (inset), assuming $\kappa_{\rm eff} \propto X^{-1}$ for $-\epsilon < 10^{-2}$, and then calculating R_K as a function of X. Here we use an extrapolation of the conductivity data from the region $10^{-3} < X < 10^{-2}$, where $2R_K \ll l\kappa_{\rm eff}^{-1}$. The results are shown in Fig. 4. Because the boundary resistance is usually attributed to the acoustic mismatch of the media (here copper surface and the fluid), it seems very surprising that R_K should be so dependent on X at such low ^{3}He concentrations. Hence we tentatively discard this analysis approach in favor of the first one, but bear in mind that additional unexpected effects within the helium (such as enrichment of ^{3}He near the Cu surface) might cause the apparent R_K to increase with X.

In conclusion, our thermal-transport experiments have extended the range in X by about four orders of

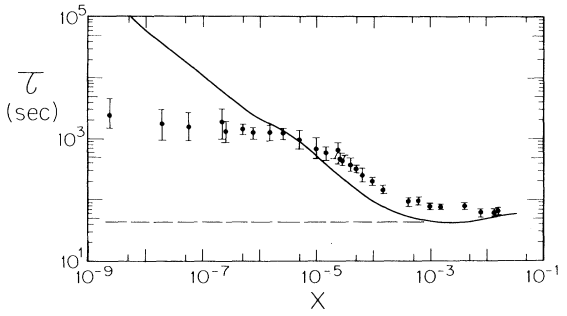

FIG. 3. The relaxation times τX for $-\epsilon < 10^{-2}$ in the superfluid phase. Solid line: calculated from Eq. (3) using the observed $\kappa_{\rm eff}$. Dashed line: calculated assuming $\kappa_{\rm eff} \propto X^{-1}$.

magnitude to $X \sim 2\times 10^{-9}$ and have shown that (a) the apparent bulk thermal conductivity $\kappa_{\rm eff}$ departs from the predicted X^{-1} dependence near T_λ for $X < 10^{-3}$; (b) there is no dependence of $\kappa_{\rm eff}$ on the reduced temperature ϵ for $-\epsilon < 10^{-2}$; and (c) the dependence on X of the measured relaxation times τ is different from expectations based on the relation $\kappa_{\rm eff} \propto X^{-1}$ for $X < 10^{-3}$. The times τ are consistent with the observed $\kappa_{\rm eff}$ down to $X \simeq 10^{-5}$.

A number of questions are raised by these measurements. For instance: Is the measured $\kappa_{\rm eff}$ representative of the bulk fluid in the limit $Q \to 0$? Here measurements with still smaller Q and higher temperature resolution[6,7] will be very useful. Are there resistive effects within the helium near the surface—in addition to the anticipated Cu-^{4}He Kapitza boundary resistance—that cause the measured $\kappa_{\rm eff}$ to be lower than predicted? If these surface effects are responsible for the added resistivity, are they also the source for the long relaxation times observed? (Experiments with a different spacing l in the conductivity cell will give an answer to these questions.) Are there assumptions made in Khalatnikov's hydrodynamic equations that need to be reexamined for very dilute mixtures? What is the vertical concentration profile in the fluid layer for a given Q at these extreme dilutions and what

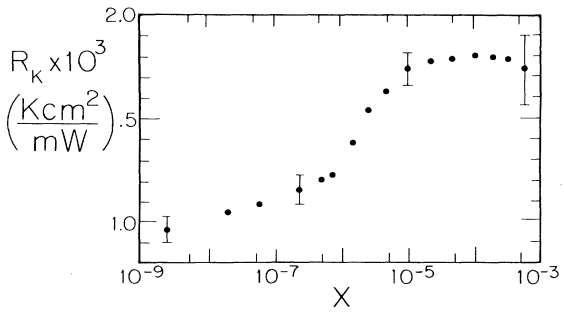

FIG. 4. Calculated boundary resistance R_K for one dilution series assuming $\kappa_{\rm eff} \propto X^{-1}$.

nonlinear effects are expected? It is clear that investigations of very dilute ^{3}He-^{4}He mixtures might reveal new and interesting aspects on transport phenomena.

The authors are grateful to D. Greywall for the gift of some purified ^{4}He ($X = 2 \times 10^{-9}$). They are appreciative for a number of discussions and correspondence with G. Ahlers, R. P. Behringer, R. Folk, A. Ikushima, and M. Tanaka, and acknowledge R. P. Behringer's comments on the manuscript. This research was supported by Grant No. DM 8024056 from the National Science Foundation.

[1]M. Tanaka, A. Ikushima, and K. Kawasaki, Phys. Lett. **61A**, 119 (1977).

[2]E. D. Siggia, Phys. Rev. B **15**, 2830 (1977).

[3]V. Dohm and R. Folk, Phys. Rev. B **28**, 1332 (1983).

[4]A. Onuki, J. Low Temp. Phys. **53**, 189 (1983).

[5]D. Gestrich, M. Dingus, and H. Meyer, Phys. Lett. **99A**, 331 (1983).

[6]V. Steinberg and G. Ahlers, J. Low Temp. Phys. **53**, 255 (1983).

[7]J. Lipa and T. C. P. Chui, Phys. Rev. Lett. **51**, 2091 (1983).

[8]I. M. Khalatnikov, *Introduction to the Theory of Superfluidity* (Benjamin, New York, 1965), Sec. 24.

[9]T. P. Ptukha, Zh. Eksp. Teor. Fiz. **40**, 1583 (1961) [Sov. Phys. JETP **13**, 1112 (1961)].

[10]R. Folk and H. Iro, to be published; R. Folk, private communication.

[11]R. P. Behringer and H. Meyer, J. Low Temp. Phys. **46**, 435 (1982).

[12]D. Gestrich, R. Walsworth, and H. Meyer, J. Low Temp. Phys. **54**, 37 (1984).