Momentum Transfer between ^{3}He Quasiparticles and Surfaces: The Effective Viscosity of Dilute Solutions of ^{3}He in ^{4}He

D. A. Ritchie,[a] J. Saunders,[b] and D. F. Brewer
Physics Building, University of Sussex, Falmer, Brighton BN1 9QH, United Kingdom

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We present the first measurements at millikelvin temperatures of both the real and the imaginary parts of the transverse-acoustic impedance of an oscillating surface in contact with dilute solutions of ^{3}He in ^{4}He, as well as with pure ^{3}He with and without a surface boundary layer of ^{4}He. Except for pure ^{3}He, large departures from hydrodynamic behavior are found which are inconsistent with slip theories incorporating specular scattering at the boundary. The effects of the ^{4}He-rich surface boundary layer and of surface roughness are discussed.

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The interaction of solid surfaces with quantum fluids is a subject of great current interest. We report here measurements of the exchange of transverse momentum between an oscillating surface and liquid helium, using a torsional oscillator of the type that has been employed extensively to study the viscosity of normal and superfluid ^{3}He.[1] In general, departures from hydrodynamic behavior occur when the quasiparticle mean free path λ becomes comparable with the internal dimensions of the oscillator (size effect) or with the viscous penetration depth δ. These have been modeled by slip of the fluid at the surface,[1] where the slip length ξ is related to λ in a way which depends on the nature of the boundary scattering. With ^{3}He-^{4}He liquid solutions the boundary scattering is known to be profoundly affected by the preferential adsorption of ^{4}He at the wall.[2] The ^{4}He surface boundary layer thus formed is impenetrable to the ^{3}He quasiparticles and this was understood to result in an enhancement in specular scattering. In this paper we present the first measurement of the complex transverse acoustic impedance of these inhomogenous systems in the absence of substantial size effects. Although $\lambda \ll \delta$,[3] significant departures from hydrodynamic behavior are observed which cannot be explained by the conventional models of boundary scattering.

The torsional oscillator consists of a hollow cylinder of internal height $h=5.7$ mm and diameter $d=17.7$ mm machined from Stycast 1266. It is glued to a hollow beryllium-copper torsion rod of internal diameter 2 mm through which the oscillator is filled and it resonates at a frequency of approximately 1.8 kHz. The oscillator is mounted below the main body of the liquid cell which is also constructed of Stycast 1266 and contains a total of 44 m^2 of sintered silver powder. Thermal contact to the mixing chamber of a dilution refrigerator is made through silver wires. The temperature of the liquid is measured by a melting-curve thermometer attached via silver wires to a separate segment of silver sinter. The melting-curve thermometer is calibrated against a Paroscientific[4] pressure gauge whose calibration is normalized to the A transition of superfluid ^{3}He on another cryostat, and the melting-curve scale of Greywall[5] is used. The torsional oscillator was designed to guarantee that the quasiparticle mean free path was much less than the smallest internal dimensions at all achievable temperatures ($T > 4$ mK) at the pressure (0 bar) used throughout. Apart from small size effects the oscillator may be thought of as a single planar surface bounding a semi-infinite region of fluid. Although quantitative measurements of the viscosity are not as precise as those made with an oscillator of small internal height (comparable with the viscous penetration depth), the interaction of the liquid and the surface may be unambiguously studied in this way and in the absence of substantial size effects.

The complex transverse acoustic impedance of a surface in a fluid, $Z=X+iY$, is defined as $\pi_{xz}(z=0)/v_x$ where the z axis is normal to the surface, v_x is the transverse velocity of the surface, and π is the fluid stress tensor. The measured quantities are the resonant period P and quality factor Q of the oscillator, determined either with the oscillator self-resonating at constant amplitude or by frequency sweeps through resonance. We have $X=(Q^{-1}-Q_N^{-1})\alpha$ and $Y=2(P-P_0)\alpha/P_0$ where Q_N is the background Q of the oscillator and P_0 the background period; α is a factor determined by the cell geometry, its moment of inertia, and the operating frequency, and the small size effects have been neglected. The background quantities are measured as functions of temperature both with the cell empty and with it full of superfluid liquid ^{4}He. Significantly there is a uniform shift between the two period backgrounds of 10.3 ns which is attributed to entrainment of superfluid by the surface due to roughness of order 1 μm, a roughness confirmed by scanning electron microscope photographs.

In the hydrodynamic regime the impedance is $Z=(1-i)\eta/\delta$ where η is the liquid viscosity and $\delta=(2\eta/\rho\omega)^{1/2}$ is the viscous penetration depth; thus $Y/X=1$. Observed values of Y/X as a function of inverse temperature are shown in Fig. 1. The results for pure ^{3}He are in good agreement with the predictions for hydrodynamic response, the systematic upward deviation at low tem-

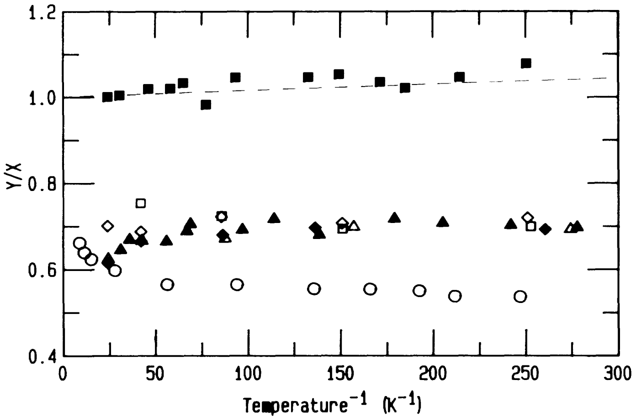

FIG. 1. Measured values of the ratio Y/X vs inverse temperatures. Filled squares, pure ^{3}He; open squares, $x_4=0.005$; open lozenges, $x_4=0.01$; filled lozenges, $x_4=0.02$; open triangles, $x_4=0.03$; filled triangles, $x_4=0.04$; circles, $x_3=0.0514$. The dashed line indicates the theoretical result for pure ^{3}He (see text).

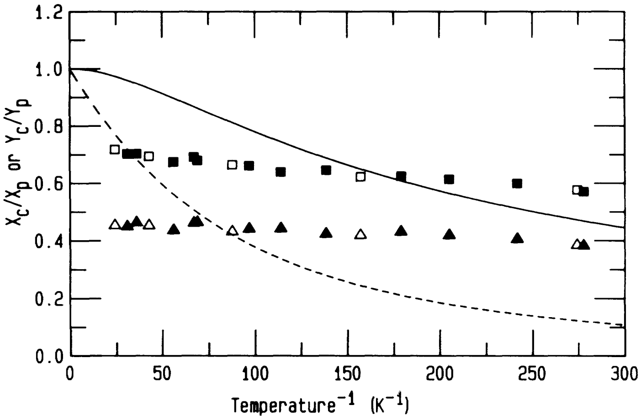

FIG. 2. The ratios of X_c, Y_c ($x_4=0.03$ and 0.04) to the value X_p, Y_p for pure ^{3}He are plotted vs inverse temperature. X_c/X_p: open squares, $x_4=0.03$, and filled squares, $x_4=0.04$. Y_c/Y_p: open triangles, $x_4=0.03$, and filled triangles, $x_4=0.04$. The curves show the results of slip theory with surface specularity $s=0.975$ for X_c/X_p (solid line) and Y_c/Y_p (dashed line).

peratures being accounted for by a small size effect (corrections in δ/h and δ/d); the dashed line indicates the exact result for our finite cylinder. However, the results for a solution with the ^{3}He concentration $x_3=n_3/(n_3+n_4)=0.0514$ show Y/X significantly less than unity and only weakly temperature dependent.

Dilute solutions of ^{3}He in ^{4}He are known to have a region of enhanced ^{4}He density near the wall[2] of thickness of order 20 Å due to the preferential adsorption of the ^{4}He atoms to the wall. In an attempt to investigate the role of surface ^{4}He, the pure ^{3}He sample was progressively contaminated with ^{4}He to produce average ^{4}He concentrations of $x_4=n_4/(n_3+n_4)=0.005$, 0.01, 0.02, 0.03, and 0.04. This operation was performed with the cell at 1.5 K after some ^{3}He had been removed from the cell fill line and from a small region at the top of the cell heat exchanger. With the assumption that the ^{4}He coats all surfaces uniformly, $x_4=0.005$ would correspond to 10 μmol m^{-2} or 1.7 layers of pure ^{4}He. The effect of the additional ^{4}He on Y/X is indicated in Fig. 1. The value of Y/X fell on addition of the first shot, indicating that ^{4}He moved to the walls of the torsional oscillator. Below 10 mK the effect of additional ^{4}He on Y/X is not significant within experimental error, but a progressive reduction in both X and Y does result. The effect appears to saturate between $x_4=0.03$ and 0.04 (10–14 layers). The ratios of the real and imaginary parts of the surface impedance for these data (X_c,Y_c) to the values for a pure ^{3}He sample (X_p,Y_p) are shown in Fig. 2 and display a weakly temperature-dependent term linear in T^{-1}. The theoretical curves are discussed later.

These results strongly suggest that the anomalous values of Y/X in both dilute solutions and pure ^{3}He contaminated with ^{4}He are due to sample inhomogeneities at the wall, namely the ^{4}He-rich surface boundary layer. In order to explore the systematics of the data further it is instructive to extract from the measured Q the effective viscosity $\eta_{\text{eff}}(T)$ assuming hydrodynamics. The data for pure ^{3}He, for ^{3}He with surface ^{4}He ($x_4=0.03$ and 0.04), and for two dilute solutions of ^{3}He in ^{4}He ($x_3=0.0514$ and 0.0107) are shown in Fig. 3. For pure

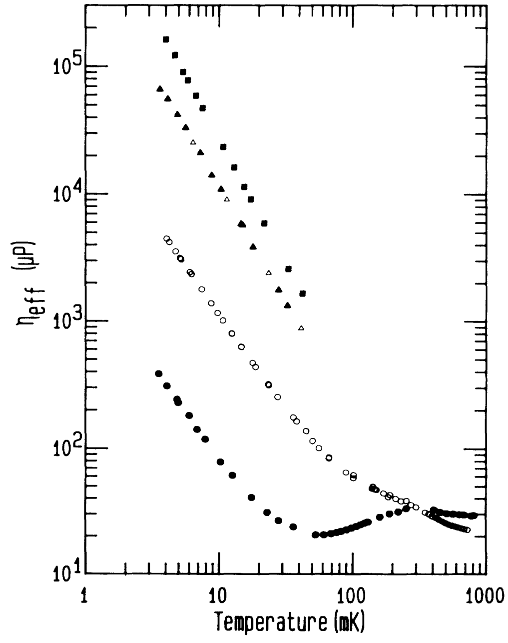

FIG. 3. The measured effective viscosity, η_{eff}, as a function of inverse temperature. Filled squares, pure ^{3}He; open triangles $x_4=0.03$; filled triangles, $x_4=0.04$; open circles, $x_3=0.0514$; filled circles, $x_3=0.0107$.

^{3}He, $\eta_{\text{eff}} \propto T^2$ as expected, but for both dilute solutions the asymptotic low-temperature dependence is $\eta_{\text{eff}} \propto T^{-(1.50 \pm 0.05)}$, and for ^{3}He with surface ^{4}He ($x_4 = 0.03$ and 0.04), $\eta_{\text{eff}} \propto T^{-1.8}$. If the measured internal dimensions of the oscillator and calculated moment of inertia are used to determine the apparatus constant α our result for pure ^{3}He is $\eta T^2 = 2.62$ P mK$^2 \pm 10\%$ where a correction has been made for the finite size of the cylinder. This should be compared with the result of Parpia and Rhodes,[6] scaled to the temperature scale adopted here, of $\eta T^2 = 2.54$ P mK$^2 \pm 5\%$.

For dilute solutions the viscosity is given by[7] $\eta = p(T)\tau_\eta$, where $p(T)$ is the ideal Fermi-gas pressure and τ_η is the viscous relaxation time. $p(T)$ is temperature independent for $T \ll T_F$ and τ_η has the form $\tau_\eta = (A/T^2)[1 + B(T/T_F)^2]$. Our results are the first measurements of the viscosity of dilute solutions to go to sufficiently low temperatures for the correction terms in $(T/T_F)^2$ to τ_η and $p(T)$ to be negligible. The theoretical expectation is thus $\eta T^2 = \text{const}$ at low temperatures. We note that the data obtained at higher temperatures with different techniques by Fisk and Hall[8] and Kuenhold, Crum, and Sarwinski[9] are also consistent with $\eta T^{1.5} = \text{const}$ as a low-temperature limit. Furthermore an increase in the temperature exponent of the effective viscosity is seen in our experiments for pure ^{3}He with a surface boundary layer of ^{4}He, as had previously been observed.[10] This increase in exponent is thus a second indication that the boundary conditions on the flow need to be modified.

The general theoretical approach to the study of the interaction of quantum fluids with torsional oscillators has been to solve the Landau-Boltzmann kinetic equation with appropriate boundary conditions. In normal ^{3}He the surface scattering is usually described in terms of the relative probabilities of specular or diffuse scattering. In superfluid ^{3}He Andréev reflection is an additional channel. The transverse acoustic impedance has been calculated by Jensen *et al.*[11] for purely diffuse scattering and this has been extended by Einzel *et al.*[12] to the general case of diffuse, specular, and back scattering. A simple phenomenological hydrodynamic model with slip correction gives $Z = (1-i)(\eta/\delta)[1 + (\xi/\delta)(1-i)]^{-1}$ which agrees with the exact calculation to first order in ξ/δ. The slip length ξ (proportional to the mean free path λ) is calculated exactly in the microscopic theory and is found to be enhanced by specularity according to the factor $(1+s)/(1-s)$, where s is the specular reflection coefficient. It is apparent that, to first order in ξ/δ, ReZ has no slip correction factor, while Im$Z = Y$ has a correction factor $1 - 2\xi/\delta$. The acoustic impedance Z includes a complex phase factor $e^{i\phi}$ with $\tan\phi = (\xi/\delta)/[1 + (\xi/\delta)]$ which reflects the relative motions of the surface and the boundary fluid. The experimental results imply, however, a phase factor that is largely temperature independent. Since the observed values of Y/X do not tend to unity at high temperatures the model would require s to be temperature dependent with $s \rightarrow 1$ at high temperatures to force agreement. This seems unphysical and also disagrees with other high-temperature measurements.[13] A comparison of the theory to the data is shown in Fig. 2 for the specular reflection coefficient fixed at the rather high value of $s = 0.975$. Another experiment which serves to highlight the importance of understanding in detail the scattering of quasiparticles by surfaces is that of Ref. 6. In this case the Knudsen minimum is observed in normal liquid ^{3}He by use of an oscillator for which the quasiparticle mean free path becomes equal to the size of the oscillator at some temperatures. The position of the minimum is inconsistent with the theory of Ref. 11, incorporating both diffuse and specular scattering.

We have examined models which attempt to take into account the variation in ^{3}He concentration with distance from the boundary. The simplified model inhomogeneous system consists of a surface boundary ^{4}He film with a sharp interface to bulk liquid. The surface impedance may be calculated from acoustic transmission line theory,[14] which incorporates continuity of transverse velocity and shear stress as interfacial boundary conditions. Alternatively a phenomenological frictional coupling between the surface and the surface boundary ^{4}He film may be introduced. It has not been possible, however, to account for the temperature independence of the phase angle ϕ. The experimental results are consistent with the bulk ^{3}He quasiparticles achieving momentum accommodation with a surface film whose velocity has a temperature-independent relative phase to that of the surface. This suggests the possibility that the ^{3}He quasiparticles may be strongly coupled to excitations at the surface, perhaps vortices, which move out of phase with it. The importance of vortices in superfluid ^{4}He-^{3}He mixture films has been discussed by McQueeney, Agnolet, and Reppy,[15] Finotello, Yu, and Gasparini,[16] and Wang and Yu.[17]

Any theory of the liquid-surface coupling must take into account the surface roughness. Our understanding prior to this experiment had been as follows. Roughness on a length scale larger than the de Broglie wavelength of the quasiparticles promotes exchange of transverse momentum through elastic collisions, inelastic scattering by the wall being extremely unlikely at low temperatures. Introduction of a surface boundary layer of ^{4}He will thus tend to reduce the diffusivity of the surface. In our case the surface roughness is estimated to be on the scale of 1 μm normal to the surface. This is large in comparison with the thickness of the ^{4}He surface boundary layer but small in comparison with the viscous penetration depth below 100 mK. The expectation then was that efficient transfer of transverse momentum would occur through the entrained fluid which moves in phase with the wall. However, according to this picture, it is difficult to see how the plating of a rough surface with a thin ^{4}He layer should have an effect.

Understanding of the momentum exchange between quantum fluids and moving walls is intrinsically of interest and vital to the measurement of bulk viscosities. This experiment has shown that the scattering of quasiparticles from walls in mixture solutions cannot be described simply in terms of the relative probabilities of specular and diffuse reflections and this is directly attributable to the presence of a region of enhanced ^{4}He concentration at the boundary. The experiment suggests that our knowledge of the boundary conditions is insufficient to allow reliable measurement of the bulk viscosity of dilute solutions. This is particularly important because of the current interest in transport properties of spin-polarized liquid ^{3}He-^{4}He solutions.

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[a] Present address: Department of Physics, Cavendish Laboratory, Madingley Road, University of Cambridge, Cambridge, CB3 0HE, United Kingdom.

[b] Present address: Department of Physics, Royal Holloway and Bedford New College, University of London, Egham, Surrey, TW20 0EX, United Kingdom.

[1] For a recent review see H. Smith, Physica (Amsterdam) **126B**, 267 (1984), and references therein; H. Smith, in *Progress in Low Temperature Physics,* edited by D. F. Brewer (North-Holland, Amsterdam, 1987), Vol. 11.

[2] For a review of this work see H. E. Hall, in *Liquid and Solid Helium,* edited by C. G. Kuper, S. G. Lipson, and M. Revzen (Wiley, New York, 1974), p. 375.

[3] We estimate, at a temperature of 4 mK, $\lambda = 5\ \mu$m and $\delta = 190\ \mu$m for pure ^{3}He and $\lambda = 21\ \mu$m and $\delta = 160\ \mu$m for a dilute solution of 5% ^{3}He in ^{4}He, on the basis of the data of J. M. Parpia and T. L. Rhodes, Phys. Rev. Lett. **51**, 805 (1983), and I. Fujii *et al.,* J. Low Temp. Phys. **57**, 163 (1984).

[4] Paroscientific Inc., Redmond, Washington 98052.

[5] D. S. Greywall, Phys. Rev. B **31**, 2675 (1985).

[6] Parpia and Rhodes, Ref. 3.

[7] G. Baym and C. Ebner, Phys. Rev. **164**, 235 (1967).

[8] D. J. Fisk and H. E. Hall, in *Proceedings of the Thirteenth International Conference on Low Temperature Physics, Boulder, CO, 1972,* edited by K. D. Timmerhaus, W. J. O'Sullivan, and E. F. Hammel (Plenum, New York, 1974), p. 568; D. J. Fisk, thesis, University of Manchester, 1972 (unpublished).

[9] K. A. Kuenhold, D. B. Crum, and R. E. Sarwinski, Phys. Lett. **41A**, 13 (1972), and in *Proceedings of the Thirteenth International Conference on Low Temperature Physics,* Ref. 8, p. 563; K. A. Kuenhold, thesis, Ohio State University, 1973 (unpublished).

[10] D. S. Betts, D. F. Brewer, and R. Lucking, in *Proceedings of the Thirteenth International Conference on Low Temperature Physics,* Ref. 8, p. 557.

[11] H. H. Jensen, H. Smith, P. Wöfle, and T. M. Bisgaard, J. Low Temp. Phys. **41**, 473 (1980).

[12] D. Einzel, H. H. Jensen, H. Smith, and P. Wölfle, J. Low Temp. Phys. **53**, 695 (1983); D. Einzel, private communication.

[13] M. J. Lea, P. W. Retz, and P. Fozooni, Phys. Lett. A **117**, 477 (1986).

[14] M. J. Lea and P. Fozooni, Ultransonics **23**, 137 (1985).

[15] D. McQueeney, G. Agnolet, and J. D. Reppy, Phys. Rev. Lett. **52**, 1325 (1984).

[16] D. Finotello, Y. Y. Yu, and F. Gasparini, Phys. Rev. Lett. **57**, 843 (1986).

[17] Cheng-tai Wang and Lu Yu, Phys. Rev. B **34**, 448 (1986).