Interfacial Friction of Thin ^{3}He Slabs in the Knudsen Limit

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A high precision torsional oscillator has been used to study ^{3}He films of thickness from 100 to 350 nm, in the temperature range $5 < T < 200$ mK. This thickness is much smaller than the viscous penetration depth, and also much smaller than the low temperature inelastic mean-free path of ^{3}He quasiparticles. Such films are observed to decouple from the motion of the oscillator below 60 mK. We interpret the frequency shift and dissipation data in terms of an interfacial friction model.

The boundary condition on shear flow past a solid surface is a fundamental problem that has been under consideration since Maxwell [1]. The simplest assumption is the no-slip boundary condition, for which the tangential component of the relative velocity is zero at the surface. In practice slip does occur, and this leads to the introduction of a slip length that is of the order of the mean-free path of the fluid, scaled by a factor which depends on the nature of elastic scattering processes from the surface. Phenomenologically, this scattering is described by specifying the relative fraction of particles which scatter specularly or diffusely. More recently, there have been attempts to achieve a microscopic understanding of surface slip through theoretical simulations of the shear flow of classical fluids [2]. Elsewhere, striking experimental results on atomic scale friction have been obtained [3], including the nanotribology of atomic monolayers [4]. For example, it is found that at low temperatures submonolayer solid films can slip on a variety of substrates, the interfacial friction being thermally activated [5,6].

The flow of the quantum fluid, liquid ^{3}He, in both its normal and superfluid phases, and the effects of boundary slip, have also been studied extensively [7,8]. In contrast to classical fluids, the inelastic mean-free path in normal ^{3}He is strongly temperature dependent, as a result of Fermi degeneracy. In the degenerate Fermi liquid, the inelastic mean-free path for scattering between quasiparticles $\lambda \propto 1/T^2$ ($\lambda = v_F \tau_\eta$, where τ_η is a weighted angular average of the quasiparticle relaxation time and v_F is the Fermi velocity). At low temperatures, λ is much larger than the interatomic separation, and is ~65 μm at 1 mK. Thus, in a suitably chosen geometry, temperature can be used as a parameter to enter the Knudsen regime, for which $\lambda \sim d$, where d is the relevant sample dimension. First order slip corrections can successfully account for the observed behavior in many experiments that just access the Knudsen regime [7], although studies of liquid ^{3}He in contact with highly polished surfaces [9], or surfaces covered by a ^{4}He film [10], have produced a number of unexplained results. The extreme Knudsen regime has recently been explored in observations of the dc flow of ^{3}He at temperatures as low as 1 mK through an assembly of 10^8 cylinders of 210 nm diameter [11], which find the expected temperature independent flow in the Knudsen limit $\lambda \gg d$.

In this Letter, we explore the transition from viscous to Knudsen flow, by investigating the flow of a thin slab of liquid ^{3}He with one free surface, of thickness d in the range 100 to 350 nm. In this study, at all temperatures investigated, the slab thickness is much smaller than the viscous penetration depth $\delta = (2\eta/\omega\rho)^{1/2}$, where the viscosity $\eta = n p_F \lambda/5 \propto 1/T^2$, n is the number density and p_F is the Fermi momentum. At low temperatures, λ becomes much larger than the slab thickness, so the sample enters the Knudsen regime. We can explore high Knudsen numbers, to Kn $= \lambda/d \gtrsim 20$, where boundary effects are dominant. Given these length scales, it can be seen that the flow regime we investigate is intermediate between the two limiting cases of flow of bulk liquid past a planar surface and slip of an atomic monolayer on a surface. With the torsional oscillator technique, we remain in the hydrodynamic regime $\omega\tau_\eta \ll 1$, at all temperatures investigated. A preliminary report of measurements on this system has appeared previously [12].

Our experiments have required the development of a very sensitive torsional oscillator, details of which are reported elsewhere [13], to detect the mass loading of such ^{3}He slabs. The head of the oscillator consists of two pieces of coin silver, with highly polished inner surfaces on which the ^{3}He film resides, diffusion bonded together with a copper gasket of nominal thickness $h = 50$ μm. Careful mechanical polishing achieves a roughness of approximately ± 10 nm as measured by an atomic-force microscope and surface scans using a profilometer. The upper part of the head, a hollow torsion rod, and the cylindrical body are machined from a single piece of coin silver. The oscillator is operated in its antisymmetric torsion mode at 2841 Hz. At the lowest temperatures, the typical quality factor, Q, is close to 10^6. The choice of coin silver is governed by considerations of the background temperature dependence of the frequency and Q

of the empty cell, which are found to be significantly weaker than comparable oscillators fabricated from BeCu [13]. The oscillator frequency can be determined to better than one part in 10^9, corresponding to the inertial contribution of a 0.1 nm thick film on both the upper and lower surfaces of the oscillator.

In order to form the helium film, a shot of ^{3}He gas is admitted from the room temperature gas handling system into the fill line, with the oscillator held at 60 mK and the fill line heated to somewhat above 1 K. The oscillator temperature is then increased to near 500 mK and its temperature regulated. Growth of the film is monitored by the decrease in oscillator frequency, and typically occurs at an inferred rate of 10 nm/h. The total mass of the film is quoted here as a nominal film thickness, determined from the frequency shift of the oscillator at 60 mK, assuming full mass loading, using the calculated moment of inertia of the oscillator head. The precise film profile inside the oscillator head, in particular, the difference in thickness of the film on the top and bottom surfaces of the oscillator, d_u and d_l, is determined by the condition that the chemical potential of the film should be constant. Thus, $V(d_l) = V(d_u) + mgh$, where $V(d)$ is the van der Waals potential evaluated at the surface of the film, $V(d) = -\gamma(d)/d^3$. For thick films retardation effects are theoretically expected to be important. Thus, for $d \ll d_0$, $V(d) = -\gamma(0)/d^3$, while for $d \gg d_0$ it is found [14] that $V(d) = -\gamma(0)d_0/1.4d^4$, where d_0 is a characteristic length of ~ 20 nm. For our geometry, this predicts that, for $d_l \ll h$, d_u has an upper limit ~ 100 nm. For these thick films, the surface profile around the perimeter of the oscillator will be determined by minimizing the surface energy, leading to a radius of curvature of half the plate separation, reducing the effective film thickness on the upper and lower surfaces.

In the following, we characterize our films by the nominal average thickness $d_t = (d_u + d_l)/2$. Measurements have been made for nominal film thicknesses, d_t, of 100, 240, and 350 nm, and they show a number of remarkable features. The results for the frequency shift due to the film and film dissipation are shown in Fig. 1. Initially, the background dependence of the frequency and Q of the empty cell are determined. The resonance frequency f_I is first calculated from the measured in-phase and quadrature response and the drive frequency f_D; the measurement is then refined by shifting the drive frequency to f_I. The Q is determined from the amplitude of oscillation, calibrated by slow frequency sweeps through resonance. The frequency of the empty oscillator and the loaded oscillator are subtracted, as are the dissipations Q^{-1} with and without helium, in order to generate the data shown in Fig. 1. Data were always taken with the temperature regulated using a ^{3}He melting curve thermometer.

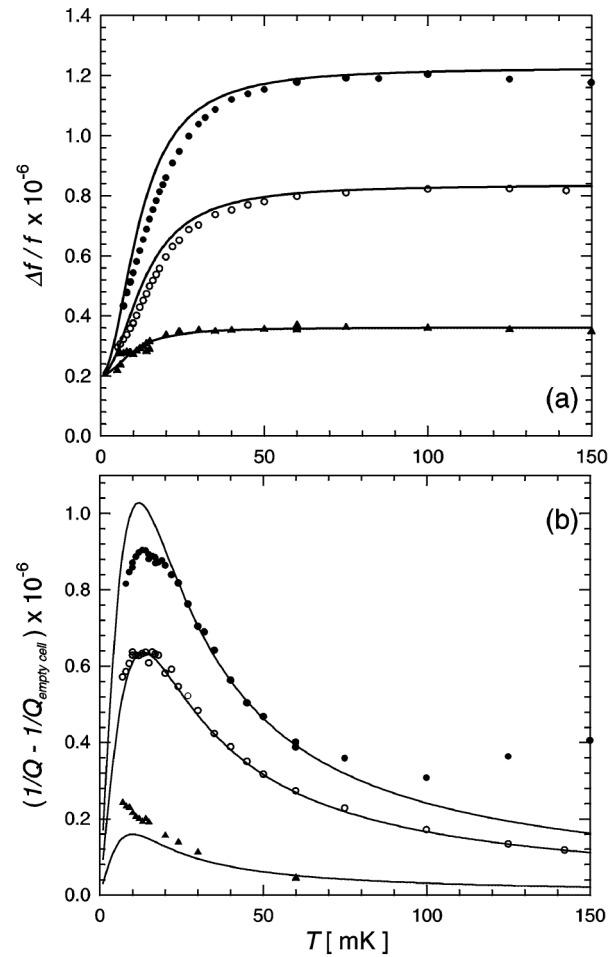At sufficiently high temperatures, it can be seen that the observed frequency shift due to the film is essentially

FIG. 1. (a) Fractional frequency shift and (b) dissipation due to a ^{3}He slab of nominal thickness 100 nm (▲), 240 nm (○), and 350 nm (●), corresponding to corrected film thickness 40, 180, and 290 nm, respectively. Fits relate to interfacial friction model (see text).

independent of temperature, consistent with full mass loading. At 100 mK, the viscosity of bulk liquid ^{3}He implies a viscous penetration depth $\delta \sim 5000$ nm, much larger than the film thickness. Thus, in the absence of surface slip, we indeed expect the film to move rigidly with the oscillating surface. Since in the degenerate Fermi liquid regime the viscosity increases as $1/T^2$, so $\delta \propto 1/T$, we might expect this to continue as the temperature is decreased. However, at low temperatures we see a reduction in frequency shift, indicating that the motion of the film tends to decouple from that of the surface. This is accompanied by an increase in the film dissipation. For the two thicker films, the dissipation shows a maximum with decreasing temperature.

To phenomenologically model these results, since $\delta \gg d$, we treat the fluid slab as a rigid object coupled by an interfacial friction to the oscillating substrate [15]. The frictional force per unit area is given by the velocity of the film relative to the surface multiplied by a friction

coefficient η_f. For a linearly oscillating substrate, the solutions of the coupled equations of motion for slab and substrate, of mass m, M, respectively, have relative amplitude $x_0/X_0 = 1/(1 + i\omega\tau)$, where the characteristic time $\tau = \rho_2/\eta_f$ is the momentum relaxation time between the film and substrate, and ρ_2 is the film mass per unit area. Hence, the film modifies the frequency and Q of the resonator according to

$$\frac{\Delta f}{f_0} = -\frac{m}{2M}\frac{1}{1 + (\omega\tau)^2}; \qquad \Delta\left(\frac{1}{Q}\right) = \frac{m}{M}\frac{\omega\tau}{1 + (\omega\tau)^2}.$$

Thus, within this model, we can analyze both measured quantities (frequency shift and dissipation due to the film) in terms of the two parameters m/M (temperature independent and scaling with the thickness of the film) and τ (temperature dependent). Clearly, simply dividing the fractional frequency shift by the dissipation determines $\omega\tau$. The decoupling of the slab from the substrate arises from an increase in τ with decreasing temperature.

The measured frequency shift extrapolates to a finite value at zero temperature of approximately the same value for all three films, and this is attributable to the fixed amount of liquid trapped at the perimeter corners and locked to the oscillator [16]. We proceed with the analysis by subtracting the same $T = 0$ frequency shift, attributed to a fixed amount of locked perimeter liquid, from all the frequency shift data, shown in Fig. 1. The corresponding corrected average film thicknesses for the three films studied are 40, 180, and 290 nm, respectively. Figure 2 shows the inferred relaxation time for the three films studied. The data show that, below 100 mK, τ appears to vary approximately as $1/T$, and is only a
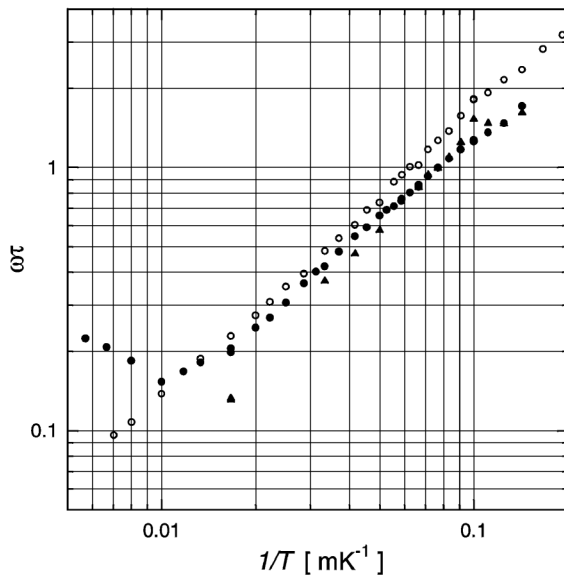

FIG. 2.   Temperature dependence of ^{3}He slab-substrate relaxation time, $\omega\tau$, for nominal thickness 100 nm (▲), 240 nm (○), and 350 nm (●).

weak function of film thickness. We fit the total frequency shift data, assuming $\omega\tau = A/T$, with A and m/M as fitting parameters, again taking the $T = 0$ frequency shift to be thickness independent. Fixing these parameters, the curves in Fig. 1(b) show the calculated dissipation, which is in semiquantitative agreement with the data. The maximum around 10 mK, for the two thicker films, corresponds to the condition $\omega\tau \sim 1$. The inferred relaxation times depend only weakly on the film thickness. Since $\eta_f = \rho_2/\tau$, this implies that the friction η_f scales approximately as film thickness. A pronounced increase in friction with film thickness up to 3 nm has previously been reported in nitrogen films on a gold substrate, investigated using quartz microbalance techniques [17].

In order to gain some insight into possible microscopic models for this behavior, we describe the system by a generalized Langevin equation in which the substrate is subject to random forces $f(t)$ arising from collisions with quasiparticles in the film. It follows from equipartition that the mean square force is given by $\langle f^2 \rangle = m\omega^2 k_B T$, independent of the nature of the film-substrate coupling. We find this leads to the result

$$\tau = \left(\frac{1}{m\omega^2 k_B T}\right)\int_0^\infty \langle f(0)f(t)\rangle\, dt.$$

Thus, within this picture the observation that $\tau \propto 1/T$ implies that the time integral of the autocorrelation function is independent of temperature.

The surprising feature of the present results is that mass decoupling occurs at higher temperature as the film thickness is increased. This is not consistent with applying the simple criterion $\lambda \sim d$ for entering the Knudsen regime. This should be contrasted with the result for dc flow through cylindrical channels of the commercial filter Anopore [11], which indicate a Knudsen minimum close to the expected condition for diffuse scattering $\lambda = 3.3d$. In that work, the increase in specularity on plating the surfaces with ^{4}He is inferred from flow in the Knudsen regime, which scales as $(1 + s)/(1 - s)$. The Knudsen minimum shifts to higher temperatures with increased specularity, but no full theory exists for this at present. We note that in our film geometry the free surface should scatter specularly, while the highly polished silver surface *may* have a high degree of specularity.

The theory of boundary effects on the flow of quantum fluids based on solutions of the generalized Boltzmann equation [18] determines a first order slip correction giving an effective viscosity (damping) of $\eta_{\text{eff}}^{-1} = \eta^{-1}(1 + 6\xi/d)$, for oscillatory flow of liquid confined between two planar surfaces of separation d. When $\lambda > d$, it is found that the damping scales as $\ln(1.526\lambda/d)$. This corresponds to a damping that initially decreases with temperature as T^2, with an extrapolated finite dissipation in the $T = 0$ limit, which passes through a "Knudsen

minimum," followed by a logarithmic increase at the lowest temperatures. The temperature dependence of the damping in this limit is very different from our observations, where as discussed one liquid surface is free.

Unfortunately, while the expressions for ballistic dc flow are established there is no available calculation of the transverse surface impedance in the extreme Knudsen limit. The results that do exist are for the transverse oscillations of a boundary in bulk liquid (at arbitrary $\omega\tau$) [19] and in contact with an inhomogeneous fluid [20] (such as a film of finite thickness) in the hydrodynamic limit with Kn $\ll$ 1. According to the interfacial friction model, the surface impedance in the presence of slip is given by $Z_s^{-1} = \eta_f^{-1} + Z^{-1}$, where the impedance in the absence of slip $Z = -i\omega\rho_2$. The real and imaginary parts of Z_s determine the change in Q^{-1} and f, quoted earlier. The interfacial friction model implies a slip length given by $\xi = \eta/\eta_f$. Using the relaxation time inferred from the fits of Fig. 1, with $\eta_f = \rho_2/\tau$, yields a slip length $\xi \gg \lambda$. These results imply that surface slip of planar thin films with a free surface of confined liquid and of bulk liquid may be very different.

We now consider the potential influence of surface roughness. While it is expected that the atomic scale roughness of the surface can be parametrized by s, it is important to consider the consequences of larger scale surface imperfections. Theories of the influence of mesoscopic surface roughness [21,22] have considered the surface impedance of a rough surface in contact with *bulk* liquid. In the simplest case, a surface profile of the form $h(x) = h_0 \cos kx$ is assumed. At sufficiently low temperatures, when $k\xi \gg 1$, the effective slip length is independent of the inelastic mean-free path λ, with a value determined by $\kappa = kh_0$. In our case $h_0 \sim 10$ nm, with characteristic $k = 4$ μm^{-1} estimated from the surface scans; thus, $\kappa = 0.4$. The model gives a low temperature maximum slip length of 1.2 μm, somewhat greater than the maximum film thickness, but much smaller than that obtained from the momentum relaxation time inferred from the data through the interfacial friction model.

Another feature of the model is the prediction of a stick-slip transition with decreasing temperature and, hence, increasing mean-free path, from a region where the slip length is negative (and the film is locked to the substrate) to one in which the slip length becomes positive. For $\kappa = 0.4$, this transition occurs at $k\xi = 0.16$, which for $\xi = 0.6\lambda$ (assuming diffuse quasiparticle surface scattering) corresponds to 31 mK. However, it appears from the data of Fig. 1 that significant slip remains at higher temperatures, and, moreover, no clear stick-slip transition is observed. Recently, a model to account for the influence of random surface roughness has also been proposed [23]. However, all these results refer to the slip of bulk liquid, and need to be extended to Kn $\gg$ 1.

In conclusion, studies of normal liquid ^{3}He into the extreme Knudsen limit ($\lambda/d \gtrsim 20$), exploiting the strong temperature dependence of the inelastic mean-free path, offer to provide new insights into the problem of boundary slip, interfacial friction, and nanotribology. There is a need for a theory of the surface impedance in this limit.

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