PHYSICAL REVIEW A VOLUME 9, NUMBER 3 MARCH 1974

Superfluid helium film below 1 K: Thickness, flow rate, and influence of He^3 †

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(Received 9 August 1973)

The superfluid helium film flow rate and the film profile (thickness versus height) on polished stainless steel have been measured in the same series of experiments down to 0.03 K. The concentration of He^3 was varied from that of nominally "pure" He^4 (~ 0.2 ppm) to 0.44% He^3. The film profile was measured below 0.7 K by observing isothermal flow oscillations. Apart from a small hysteresis effect, it agrees with the simple Frenkel-Schiff theory using the van der Waals potential calculated from the Lifshitz equation. In agreement with theory the profile is not changed by small concentrations of He^3 (up to a few hundred ppm) which, however, significantly decrease the superfluid flow rate below 1 K. The effect of the He^3 on the flow rate is most striking below 0.5 K, where surface-tension measurements have shown that the He^3 is partially adsorbed on the surface of the He^4, lowering the surface tension. In this region the flow rate is observed to be approximately proportional to the square root of the surface tension, as predicted by Arkhipov. With a larger amount of He^3 (0.44%) the flow rate below 0.1 K increases quite rapidly with decreasing temperature. The increase is apparently connected with the beginning of Fermi degeneracy in the He^3 dissolved in the film. Some observations of the flow rate for very small level differences are also reported.

Recent work[1-3] has shown that small amounts of He^3 are adsorbed onto the free surface of liquid He^4 at very low temperatures (less than 0.5 K). The same phenomenon occurs in the saturated helium film and, in a conventional film-flow experiment at low temperatures, one may observe the transfer of the adsorbed He^3 across the lip of the beaker. In the course of an experiment to study this phenomenon (which will be described in another paper) we have measured the effect of He^3 on the superfluid film-flow rate and on the thickness and profile of the film. The addition of He^3 allows one to vary the condition of the film's free surface as well as the transfer of momentum and heat below the surface. Although this was not the main objective of our experiments, we hope that we have sufficient new results to cast some light on the theoretical problem of the superfluid critical velocity in the film.

MEASUREMENTS OF THE SUPERFLUID TRANSFER RATE

The superfluid flow rate Q was measured down to 0.03 K by a conventional method using capacitors to detect the change in level inside and outside a beaker following a change in the outside level. The beaker was made of a 19-mm-diameter 0.25-mm-wall 321 stainless-steel tube with one end sealed with Epibond 100 A epoxy.[4] The tube was polished to a mirror finish on a lathe, washed in a series of solvents, and rubbed with a clean cotton cloth before mounting in the cell. Strips of sintered copper sealed through the bottom of the beaker provided thermal contact between the helium inside and outside the beaker. A concentric epoxy cylinder inside the beaker made the cross-sectional area of the inner liquid level equal to 0.76 cm^2, about the same as that outside, 0.79 cm^2. (This was desirable for the He^3 transport experiment.) The cell was flushed with helium gas several times and pumped out with a mechanical pump before cooling.

A stainless-steel bellows formed the bottom of the cell. By varying the pressure on the other side of the bellows, the level outside the beaker could be raised or lowered. Both the inner and outer helium levels were monitored by a parallel plate capacitor in each chamber. Capacitances were measured to 10^{-4} pF using two General Radio 1615A capacitance bridges. The off-balance bridge signals could be displayed on a two-channel chart recorder to obtain continuous level readings. Temperatures were measured with a cerium magnesium nitrate magnetic thermometer.

The film-transfer rate was measured by first raising the levels to 4.8 ± 0.3 mm from the beaker rim. After equilibrium, the outer level was lowered by several mm over a period of about a minute. The subsequent outflow from the beaker was observed to rise quickly to a constant value which it maintained for at least 200 sec. (corresponding to a drop in level of one or two mm). After this time, sudden changes to a lower flow rate were observed in many runs. These changes are in agreement with observations of "preferred rates" made by others.[5,6] Small but significant differences between the transfer rates for inflow and outflow were also observed sometimes. These effects, which are similar to those reported by many previous workers, are discussed in Appendix A. All of the data presented in this section of the paper were obtained during the first 200 sec. of outflow and they are characteristic of flow with the levels close to each other and distance from the rim, $h = 5$ mm.

The measurements of the flow rate were taken in two separate series between which the cell was accidentally warmed to room temperature and the beaker was exposed to the atmosphere. Data points taken in the second series have been plotted as open symbols in Fig. 1. The first series of data are systematically higher and they have been multiplied by a factor of 0.89 before plotting as filled symbols on the figure. After this normalization, the two curves for nominally "pure" He^4 (which actually contains a few parts in 10^7 of He^3) are nearly identical. The concentration was changed in each series of measurements by adding He^3 through a capillary without warming above 4 K.

Several groups of workers[7-10] have measured flow rates for "pure" He^4 at temperatures below 1 K and the rates they obtain all show a significant increase as the temperature is lowered below about 1 K. The data of Herbert, Chopra, and Brown,[10] for a Pyrex beaker with h equal to a few cm, have been plotted on Fig. 1 for comparison with ours. Their results are qualitatively similar to those obtained by others, but lower in absolute magnitude presumably because of the large value of h.

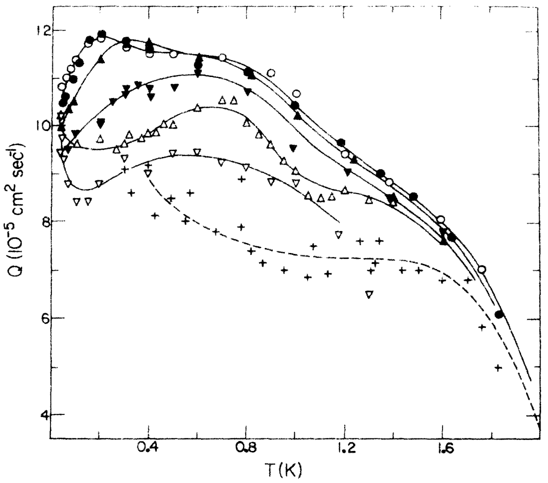

FIG. 1. Film flow rate Q vs temperature for some representative He^3 concentrations: ● ○, "pure" He^4 (~ 0.2 ppm); ▲, 55 ppm; ▼, 540 ppm; △, 1510 ppm; ▽, 4430 ppm; the closed symbols refer to the first series of measurements, the open symbols to the second. The crosses (+) are the data of Ref. 10.

Above 1 K, the magnitude our transfer rates is in good agreement with previous work using clean polished stainless-steel substrates.[11]

Perhaps the most important remark that we can make about Fig. 1 is that none of the complicated dependence of flow rate on temperature and concentration below 1 K is due to changes in either the superfluid fraction ρ_s/ρ, or in the equilibrium thickness or shape of the film. This is demonstrated by the measurements in the next section of the paper, where we find that, to a good approximation, the profile of the film below 1 K in subcritical oscillatory motion is independent of temperature and concentration and in agreement with a simple theory. All the observed changes in the flow rate must presumably be due to changes in the superfluid critical velocity.

For low concentrations, i.e., less than a few hundred ppm, and $T < 0.5$ K, there is close correlation between the temperature at which the flow rate decreases as the temperature is lowered (Fig. 1) and the temperature at which substantial amounts of He^3 are adsorbed on the surface. In Fig. 2 we have plotted the flow rate in this region against the fractional change in surface tension $(\sigma - \sigma_4)/\sigma_4$. (The surface tension was obtained from observations of the capillary rise in the two capacitors, normalized to a value of $\sigma_4 = 0.378$ dyn/cm for ideally pure He^4 at 0 K.) The observations are consistent with the dependence on σ suggested by Arkhipov,[12] namely, flow rate $Q = (\text{const}) \times \sigma^{1/2}$. Unfortunately, Arkhipov's theory also predicts a strong dependence of film thickness on flow rate, which is not observed,[13] and a much higher absolute flow rate. It is likely that another dissipative mechanism sets in before the one proposed by Arkhipov. If this were to involve vortex lines for

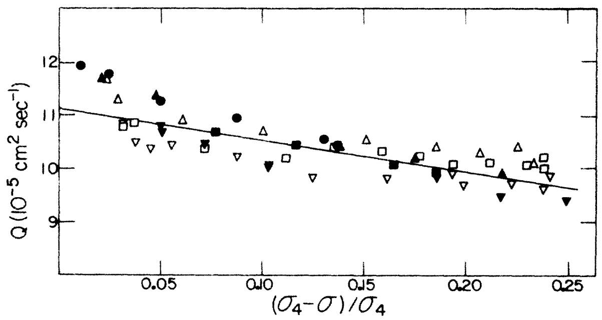

FIG. 2. Film flow rate Q below ~ 0.5 K vs fractional decrease in the surface tension. The He^3 concentrations are: ●, "pure" He^4(~ 0.2 ppm); ▲, 55 ppm; △, 110 ppm; □, 175 ppm; ■, 213 ppm; ▽, 300 ppm; ▼, 540 ppm. The closed symbols refer to the first series of measurements, the open symbols to the second. The curve represents $Q = (\text{const.}) \times \sigma^{1/2}$.

example, the dependence of Q on the surface tension through the vortex core radius might approximate that observed.

For concentrations higher than a few hundred ppm, the effect of the He^3 is appreciable at temperatures above 0.5 K, where the surface adsorption is relatively small. In this region we are probably seeing effects due to the He^3 dissolved in the film. The addition of He^3 seems to postpone the rise in the flow rate as the temperature is lowered near 1 K. There is a very rough correlation with the roton free path,[14] which is decreased by the He^3 and which becomes comparable with the film thickness at temperatures in this region.

For the highest concentration of He^3 (0.44%) there is a minimum in the flow rate at ~0.1 K. Below 0.1 K, the flow rate increases quite rapidly with decreasing temperature. It is tempting to view this minimum as connected with the beginning of Fermi degeneracy in the dissolved He^3. The Fermi temperature for this concentration is ~0.07 K,[15] and the He^3-He^3 collision rate can be expected to decrease markedly below this temperature. The He^3-He^3 free path is estimated to be about 2000 Å at 0.1 K.

THE FILM PROFILE

The profile of the film, i.e., its thickness as a function of height above the liquid, was studied by measuring the period τ of isothermal flow oscillations. This technique was first used by Atkins.[16] Our measurements of τ were carried out during the second series of experiments on the flow rate.

Following Atkins's derivation, and assuming, as he did, that the flow velocity and the superfluid fraction ρ_s/ρ are uniform throughout the thickness of the film and that the thickness d at height H does not depend on the flow velocity, we obtain for a film containing He^3

$$\tau = 2\pi \left\{ \frac{A^*}{2\pi r^*} \left[g - \frac{XA^*}{m_4 V^*} \left(\frac{\partial \mu_4}{\partial X} \right)_T \right]^{-1} \int_0^h \frac{dH}{(\rho_s/\rho) d} \right\}^{1/2} , \tag{1}$$

where A^*, r^*, and V^* are the reduced cross sectional area, radius, and liquid volume inside and outside the beaker $[A^* = A_{in} A_{out} / (A_{in} + A_{out})$, etc.$]$; $m_4 g$ is the weight of a He^4 atom; X and μ_4 are the molar He^3 concentration and He^4 chemical potential in the liquid; and h is the height of the beaker

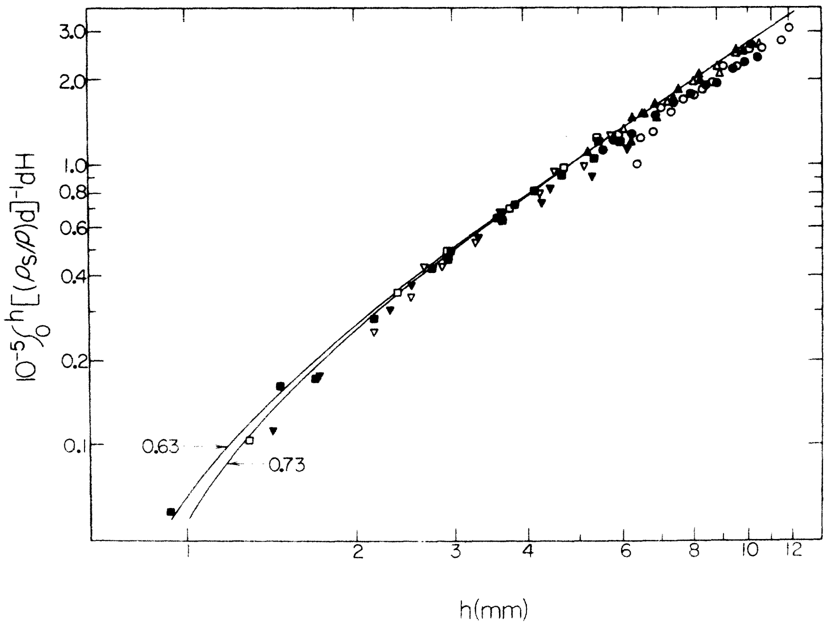

FIG. 3. Logarithmic plot of $10^{-5} \int_0^h [(\rho_s/\rho)d]^{-1} dH$, obtained from measurements of the oscillation period τ and Eq. (1), vs the height of the beaker rim h. The concentrations and temperatures are: "pure" He^4 (~0.2 ppm), 0.036 K (○) and 0.7 K (●); 110 ppm, 0.036 K (△) and 0.7 K (▲); 299 ppm, 0.036 K (□) and 0.1 K (■); 604 ppm, 0.036 K (▽) and 0.1 K (▼). The theoretical curves, which have no adjustable constants, have been calculated for a substrate with a plasma frequency of 18 eV, which is our estimate for stainless steel, and two values of the classical meniscus height h_0, given in mm on the curves. The value $h_0 = 0.73$ mm corresponds to the surface tension of pure He^4, and 0.63 mm corresponds to the lowest surface tension for this group of measurements, for 604 ppm at 0.036 K. The scatter in the points includes that due to the "hysteresis" described in Appendix A.

rim above the liquid level. For our apparatus $A^*=0.386\ \mathrm{cm}^2$, $r^*=0.47$ cm and V^* varied from 3.3 to 4.2 cm^3.

The derivation of Eq. (1) is straightforward. The derivative $(\partial\mu_4/\partial X)_T$ appears because, as well as an oscillating difference in gravitational potential, there is an oscillating difference in He^3 concentration between the inside and outside of the beaker. The derivative $(\partial\mu_4/\partial X)$ is proportional to the concentration dependence of the osmotic pressure; for low concentrations, where van't Hoff's law is obeyed, $(\partial\mu_4/\partial X)_T=-kT$. For higher concentrations and low temperatures the necessary formulas and data can be found in Ref. 15.

Measurements of τ were made below 0.6 K for $h=1$ to 11 mm. Damping of the oscillations prevented measurements higher than 0.6 K for the higher concentrations of He^3. Quite often small but significant variations in τ were observed depending on whether the oscillations were preceded by critical flow into or out of the beaker. A discussion of this effect, may be found in Appendix A. Despite these small variations, the data for all concentrations are in quite close agreement when plotted in Fig. 3. Here we have shown $r^*\tau^2[g-(XA^*/m_4V^*)(\partial\mu_4/\partial X)_T]/2\pi A^*$, which by Eq. (1), should be equal to $\int_0^h[(\rho_s/\rho)d]^{-1}\,dH$, plotted against h for representative temperatures and concentrations. It is clear that there is no systematic dependence of the profile $d(H)$ on either temperature or concentration within the scatter.

The simplest theory of the film thickness for pure He^4, originally proposed by Frenkel[17] and Schiff,[18] depends on the fact that the surface of the film and of the liquid is both a surface of constant chemical potential and, for low temperatures, zero vapor pressure. The chemical potential is $[\mu_4(P,T)+m_4gH+V(d)]$, where P is the pressure and $V(d)$ is the van der Waals potential per atom at the surface. If $\mu_4(P,T)$, the intrinsic chemical potential near the surface of the film, is assumed to be independent of d and the same as in the bulk liquid, this gives for the relation between film thickness d and height H:

$$-m_4\sigma/\rho R+m_4gH+V(d)=0\,, \tag{2}$$

where σ is the surface tension, R is the radius of curvature, and we have replaced $(\partial\mu_4/\partial P)_T$ by m_4/ρ. Above the "classical" meniscus height $h_0=(2\sigma/\rho g)^{1/2}$, which is 0.73 mm in pure He^4, the effect of the curvature term can be neglected and we have

$$m_4gH+V(d)=0\,,\quad H\geqslant h_0\,. \tag{3}$$

Sabisky and Anderson[19,20] have experimentally verified this equation over a very wide range of d and H for both saturated and unsaturated films which were adsorbed on various substrates including cleaved fluoride crystals, Si, and SiO_2. Their measurements were made at 1.4 and 2.1 K. We refer to their papers for further details and for some account of earlier work on the film thickness.

The van der Waals potential at distance d from the substrate, $V(d)$, which is given by an equation due to Lifshitz,[21] can be represented over limited ranges of d by $-K/d^n$, where K is a constant characteristic of the substrate and n varies from $n=3$ (small d) to $n=4$ (large d). The value of K and the dependence of n on d for a metal substrate mainly depends on its plasma frequency. We estimate the plasma frequency for stainless steel to be 18 eV from the experimental values for Fe, Cr, and Ni.[22] From the calculations of Sabisky and Anderson,[19] this gives to a good approximation:

$$V(d)=(2.5\times10^{-12}\mathrm{erg\,cm}^{3.5})/d^{3.5}\,. \tag{4}$$

Substitution in Eq. (3) gives

$$d=(284\ \mathrm{A})\left(\frac{1\ \mathrm{cm}}{H}\right)^{1/3.5}\,,\quad H\geqslant h_0 \tag{5}$$

which, when substituted in the integral $\int_0^h[(\rho_s/\rho)d]^{-1}\,dH$ in Eq. (1) and putting $\rho_s/\rho=1$, gives the theoretical curves in Fig. 3. (Note that the contribution from the meniscus region $0\leqslant H\leqslant h_0$ to this integral is negligible). The agreement between theory and experiment is very satisfactory, since there are no adjustable constants in the theory.

The generalization of the theory to He^3-He^4 mixtures is quite straightforward. The van der Waals potential is the same for the two isotopes, and the He^3 and He^4 chemical potentials are uniform over the surface (although the concentration is not). After using the Gibbs-Duhem relation we obtain instead of Eq. (3),

$$\overline{m}gH+V(d)=0\,,\quad H\geqslant h_0$$

where

$$\overline{m}=(1-X)m_4+Xm_3\,. \tag{6}$$

The difference between $\overline{m}$ and m_4 is quite negligible for the concentrations in the present work. There are also changes in ρ and ρ_s/ρ, which again are very small. Even at the lowest temperatures, where there is as much as 1.5 monolayers of adsorbed He^3 on the surface, these changes will be of the order of 2%. More important is the change in h_0, which is illustrated by the two theoretical curves in the figure.

To examine the profile as a function of temperature and concentration with greater precision, three sets of measurements were made as a function of T keeping h constant so there was no supercritical flow between measurements. The oscillations were induced by a heat pulse to the liquid outside the beaker which produced a small, temporary difference in osmotic pressure. The results are displayed in Fig. 4 as a plot of the quantity

$$d_{10} = \frac{h^{(1+1/3.5)} - h_0^{(1+1/3.5)}}{(1+1/3.5)\int_0^h [(\rho_s/\rho)d]^{-1}\, dH} \qquad (7)$$

According to the theory d_{10}, which is the thickness of the film at $H = 10$ mm, should be equal to (284 ± 4) Å independent of h for small X and T. The data confirm this to an accuracy of 3%.

APPENDIX A: DIFFERENCES IN FLOW RATE AND PROFILE FOR INFLOW AND OUTFLOW

In this Appendix we describe a curious asymmetry in the film with respect to the direction of critical flow. Figure 5 shows a sequence of outflow rates followed by a sequence of inflow measurements plotted as a function of h, the height of the rim at the beginning of each flow. (As explained earlier, each flow measurement begins with the levels equal and at a distance h below the rim.) The data points are numbered in sequence and also have arrows to show whether they were outflows (increasing h) or inflows (decreasing h). After each flow we observed the oscillation period τ, which according to Eq. (1), is proportional to the square root of $\int_0^h [(\rho_s/\rho)d]^{-1}\, dH$. These points are also numbered in Fig. 5 and have arrows to show whether they were *preceded* by an inflow or outflow. Both flow rate and profile data are seen to lie on two separate curves, which we can think of as corresponding to two different states of the film, one characteristic of outflow and the other of inflow.

Although we have not made a systematic study of this effect, observations at a number of different concentrations and temperatures, all below 0.7 K, suggest the following generalizations. All measurements in a given state at fixed temperature and concentration are consistent with each other, but differences between the two states are not always observed. Sometimes the flow rate was different in the two states but not the profile and vice versa. The difference in the flow rate Q between the "inflow" and "outflow" states could be positive or negative and was unpredictable, but τ was always greater in the inflow state. At fixed temperature and concentration, observations of the period τ showed that a given state could persist for a period much longer than any of the equilibrium times of the apparatus, either for heat transfer or for He^3 transfer.

We believe it is very difficult to account for these observations and similar ones made by previous workers without invoking the idea that pinned vortex lines coupled with irregularities in the geometry of the substrate have some influence on both the flow rate and the profile. A reversal in flow direction also reverses the direction of the Magnus force on the vortex lines. This could change the interaction of pinned vortex lines with the irregular substrate. Without such a mechanism it would seem to be very difficult to explain the asymmetry with respect to the two directions of flow or the fact that this asymmetry can persist for long periods (up to an hour) in the oscillations of the film.

APPENDIX B: A NOTE ON THE FLOW RATE FOR VERY SMALL LEVEL DIFFERENCES

Atkins[23] and subsequent workers[6] have observed a decrease in the film flow rate as the levels approached within ~3 mm of each other at the end

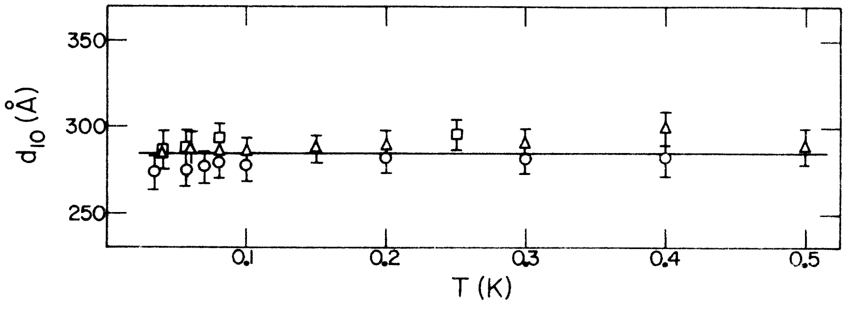

FIG. 4. The quantity d_{10} vs temperature obtained from measurements of the oscillation period and Eqs. (1) and (7) for three concentrations and rim heights: ○, 110 ppm, $h = 8.9$ mm; △, 306 ppm, $h = 10.2$ mm; □, 1510 ppm, $h = 8.4$ mm. The theoretical value of d_{10}, which is equal to the thickness of the film at a height 10 mm, is shown by the horizontal line.

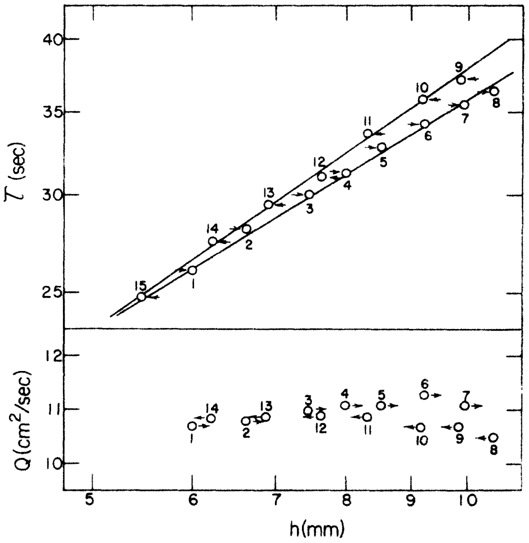

FIG. 5. Oscillation period τ and the critical film flow rate Q as a function of h, the height of the beaker rim above the liquid levels. The temperature is 0.1 K and the He^3 concentration 110 ppm. Both Q and τ have different values for inflow and outflow (see Appendix A).

of a flow measurement. This has been interpreted as a simple dependence of the flow rate on the level difference. In our experiment we observe the flow for very small level differences at the beginning of a flow measurement. Typically we observed the flow rate to have reached its full, critical value by the time the level difference was 0.05 mm. At no time was the level difference greater than 0.1 mm, before the critical rate was obtained. No transitions to a higher critical rate were ever seen as we increased the level difference up to 10 mm. The observed times required to accelerate the film to its critical rate (1–5 sec.) are in complete agreement with the values one would calculate using standard superfluid hydrodynamics.

Our observations are in agreement with the work of Duthler and Pollack,[24] who found that the transfer rate is nearly independent of the level of difference for distances to the rim, $h < 2$ cm. Their work was done on neon-coated beakers and did show a dependence on level difference for $h > 2$ cm. We are unable to compare results for $h > 2$ cm, since the size of our cell prevented us from making such measurements. More recently, Milbrodt and Pollack[25] have observed a significant dependence on level difference for h as small as 0.5 cm using solid-argon beakers.

Liebenberg[26] has studied the thermally driven superfluid film and has found that the flow rate is a function of the thermally induced chemical potential difference. He fits his data, as have Duthler and Pollack, and Milbrodt and Pollack, to the Langer-Fisher fluctuation theory[27] of intrinsic critical velocities. This theory predicts a superfluid critical velocity v_s that varies as $\rho_s / T(A - B \ln H)$. It is possible that in our experiments this weak logarithmic dependence on level difference was masked by some competing effect such as a slightly changing film thickness. However, without modification, the fluctuation theory does not seem to agree with either the temperature dependence or the dependence on level difference which we have observed at low temperatures.

[†] Work supported by Grants Nos. 27242 and 31650 from the National Science Foundation.

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