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THICKNESS OF THE STATIC AND THE MOVING SATURATED He II FILM*

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(Received 9 January 1970)

We have used a three-terminal capacitor incorporated in a flow vessel to search for the theoretically predicted difference in the thickness Δd between the moving and the static saturated He II film. The observations indicate that $\Delta d = 0$ to within a few Å.

Despite the considerable amount of effort that has been applied to determinations of the thickness d of both static and moving He II films,¹⁻³ the relation between theoretical predictions and experimental results in these matters remains unsatisfactory. An important example of this confusion concerns the difference in thickness Δd between the static and the moving film when other variables, such as the temperature and the vertical distance Z of the film above the liquid surface, are kept constant.

According to the now widely accepted ideas of Kontorovich,⁴ the following expression holds in regions of the moving film where no dissipation occurs:

$$(\rho_s/2\rho)v_s^2 + gZ - \gamma d^{-n} = 0, \quad (1)$$

where ρ_s/ρ is the superfluid fraction of the total density ρ , v_s is the superfluid velocity, g is the gravitational constant, and γ is the coefficient of the van der Waals attractive potential between the helium and the substrate in which n is usually taken as 3. For the static film ($v_s = 0$) we find from Eq. (1)

$$d = (\gamma/gZ)^{1/n}, \quad (2)$$

but when $v_s \neq 0$, the Kontorovich relation indicates that the film should be thinner by an amount

$$\Delta d = (1/n)(\rho/2\rho_s)(Q^2/gZd). \quad (3)$$

Here $Q = (\rho_s/\rho)v_s d$ is the flow rate per cm of container perimeter. Experiments using optical techniques by Gribbon and Jackson² (flow from a beaker) and by Pickar and Atkins³ (third sound)

have indicated Δd to be smaller than expected from Eq. (3), but scatter in the data has precluded a quantitative evaluation of Δd .

The main purpose of the investigation reported here was to devise a new method for measuring Δd more precisely than in previous work. During the course of the experiments additional features of the He II film became clarified.

As a probe of the film thickness a three-terminal capacitor (nominal capacitance ~ 19.5 pF) was constructed as shown schematically in Fig. 1. The cylindrical bucket (2.540 cm o.d., 1.925 cm i.d., and 8.25 cm deep) is a composite of stain-

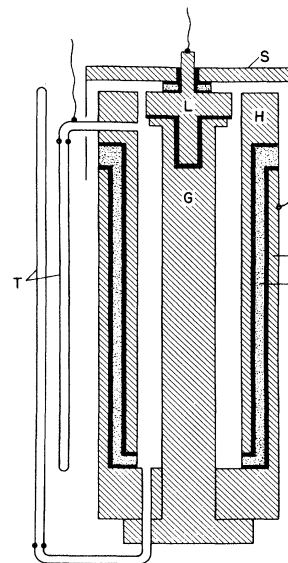


FIG. 1. Schematic drawing of bucket capacitor.

less steel and cloth-based laminated phenolic (P) bonded by Eccobond 26⁵ (E). The inner stainless-steel member serves as the flow surface and the high-voltage capacitance plate (H). Atop the central post (1.651 cm diam) and electrically insulated from it is a stainless-steel disk (0.381 cm long) acting as the low-voltage plate (L) and providing a gap of 0.010 cm between it and the flow surface. The central post, the outer stainless-steel member, and the top steel shield form a common ground (G). Two glass sight tubes (T) communicate with the bucket interior, the lower one for monitoring the liquid level in the bucket, the other for probing the relative chemical potential ($\Delta\mu$) of the film 0.64 cm below the rim.⁶

The bucket is rigidly mounted within a glass cylinder with metal ends. This is fitted with a pumping tube and a valve opening to the liquid-He bath which surrounds the can, so that He may be admitted either as purified gas or as liquid directly from the bath. Within the can is an externally operated plunger for adjusting the liquid level with respect to the bucket.

Capacitance measurements through the Clausius-Mosotti relation may provide data on the atomic density of dielectric material in the gap at the bucket rim. These were made with a 1615-A General Radio capacitance bridge, using a 1 kc, 30-V signal from a 1311-A General Radio audio oscillator and a model 121 PAR lock-in amplifier as a null detector. Under operating conditions the long-term (~2-h) stability of the system was generally of the order $\pm 1 \times 10^{-5}$ pF, the sensitivity being about 2×10^{-6} pF. From the bridge conductance we calculate the power density deposited in the film as only $\sim 2 \times 10^{-2}$ erg/sec cm². Because, in addition, the film at the point of measurement is well shielded from thermal radiation, this method for determining d should perturb the film far less than that using optical techniques.

In order to calibrate the apparatus, increments of He gas were admitted to the initially evacuated can, and at each step the pressure P and the capacitance C were measured. This process generated an adsorption isotherm in the form of C vs P/P_0 (P_0 is the saturated vapor pressure) superimposed upon a much larger contribution to C from the vapor phase. Over the range $0.1 < P/P_0 < 0.5$ the adsorption isotherm is quite flat,⁷ so that here the slope of the observed curve represents changes of atomic density N in the gas phase alone. Consequently, data in this region together with gas-law calculations including the

second virial coefficient were used to determine the relation between ΔN and ΔC . Near $P/P_0 = 1$, a C vs P/P_0 curve rises steeply and then stops abruptly at $P/P_0 = 1$, C remaining constant because the further addition of gas condenses as bulk liquid and not as film. The difference between C at $P/P_0 = 1$ and C calculated for the gas extrapolated to $P/P_0 = 1$ is a measure of the number of atoms absorbed as film (with the substrate suspended in the vapor above bulk liquid); this may then be translated into values of d from knowledge of the apparatus geometry and the assumption that the film has the same density as the bulk liquid.⁸ The filling process was completed by admitting liquid from the bath into the can to the desired depth, C being observed to determine the accompanying change in d .

Two series of runs were made. In the first of these, performed with the flow surfaces of the apparatus only machined, albeit very carefully, the films produced were apparently quite thick. The center post was then removed from the bucket and replaced after the capacitor plates had been polished with 900-grit SnO₂. For experiments performed with the polished surfaces, d appeared to be about $\frac{1}{3}$ as large as in the first series of runs. This paper reports principally upon the results of the second series, although, with few interesting exceptions, the film behavior was qualitatively similar in the two series.

From seven curves of C vs P/P_0 , obtained at temperatures between 1.55 and 2.01 K, $\Delta N/\Delta C \equiv \alpha$ was determined to be 2.195×10^{22} atoms/cm³ pF with an uncertainty of about 1%.⁹ Correspondingly we found $\Delta d/\Delta C \equiv \beta = 4.98 \text{ \AA}/10^{-5} \text{ pF}$. These values have been used in reducing all the data for the second series (thin films).

As an indication of the power of the method described here, as well as of the stability of the film, we show in Fig. 2 some results for the static film thickness (at constant Z) as a function of temperature between 1.088 and 2.160 K. Experimentally, a run was started at a reference temperature T_0 at which a reference capacitance C_{T_0} was measured, the liquid levels inside and outside the bucket being equal. Then the system was successively brought to new values of T and the corresponding capacitance values C_T obtained. Temperatures were in all cases derived from the vapor pressure inside the can as read on a calibrated oil manometer. In the figure we plot $d_{\text{obs}} - d_{\text{calc}}$ vs T , where for a given T

$$(d_{\text{obs}} - d_{\text{calc}}) = (C_T - C_{T_0} - \Delta N_{T-T_0} \alpha^{-1}) \beta. \quad (4)$$

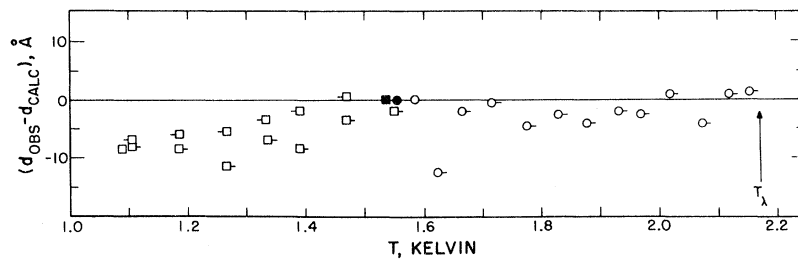


FIG. 2. Plot of Eq. (4) as a function of temperature showing $d(T) = \text{const.}$ See text for explanation.

Here ΔN_{T-T_0} is the change in atomic density of the saturated vapor between T and T_0 as calculated for the real gas. Hence, if d actually remains constant with T , any observed capacitance changes ($C_T - C_{T_0}$) should arise solely from changes in vapor density and $d_{\text{obs}} - d_{\text{calc}}$ should vanish. From Fig. 2 we see this to be the case over a wide temperature range. (In the figure the solid symbols represent the reference points for the respective runs, and the "arms" on the symbols point in the direction of the temperature change.) The systematic drift in the data below 1.5 K is probably not real and could be corrected by varying α well within the limits of its uncertainty. In any event it seems safe to conclude from these and other data we have obtained that, between 1.088 and 2.160 K and at constant Z , the equilibrium film thickness remains constant to within an atomic (statistical) layer. Some less precise earlier experiments have suggested these results, while others have indicated that as T decreases d becomes thinner.¹

For thickness measurements on the flowing film, we first established a gravitational potential using the plunger, and then measured the time dependence of the capacitance and of the liquid levels in the can and in the two sight tubes. This procedure provided simultaneous observations of d and $\Delta\mu$ near the rim as functions of Q and Z .

The principal result of this investigation is that within an uncertainty of $<5 \text{ \AA}$ no dependence upon Q was found for d in some 33 experiments about equally divided between fillings (inflows) and emptyings (outflows) of the bucket and covering the ranges $1.12 \text{ K} < T < 1.93 \text{ K}$ and $1.4 \text{ cm} < Z < 8 \text{ cm}$.

Consider, for example, an inflow at 1.12 K which equilibrated at $Z = 1.4 \text{ cm}$. Just prior to equilibration of the levels d was measured as 420 \AA and Q as $20.5 \times 10^{-5} \text{ cm}^3/\text{sec}/\text{cm}$; and according to Eq. (3), taking $n = 3$, d should have increased by 120 \AA when the flow stopped. During the flow the measured value of C had been slowly

changing, reflecting the $d(Z)$ relation; but when the liquid levels stopped changing so did C . In fact C remained steady to within $\pm 5 \times 10^{-6} \text{ pF}$, indicating substantially no change in d at the completion of the flow process. This behavior was typical of the runs, which were followed for at least 15 min after flow stopped. In order to ascertain whether d might remain constant in a metastable state, several runs were extended for one hour after level equilibration; a flashlight was then focused on the outside bucket wall; and finally the cryostat was vigorously tapped. None of these actions produced an observable effect upon d .

These null results are illustrated in Fig. 3, where d vs Z is plotted for several flow processes. Here Z is measured from the liquid level inside the bucket to a point midway on the inner plate, i.e., 0.190 cm below the rim; and C at this point is considered as the linear average value of C over the plate rather than an average according to the $Z^{-1/n}$ law—this introduces less than 1% error in d in the worst case ($Z = 1.4 \text{ cm}$). Because movement of the plunger at the start of a run sometimes perturbed C by as much as $8 \times 10^{-5} \text{ pF}$, d was always normalized to its value near $Z = 7 \text{ cm}$ as obtained from the initial filling process, during which the plunger was not moved. Equation (3), if correct, has three consequences for the data as graphed here: (1) As discussed above, the end points of the runs representing static conditions should depart from the curve obtained during flow; (2) experimentally, flow rates Q_I and Q_O for inflow (I) and outflow (O) were found to differ, and hence the $d(Z)$ profile for I and O should be different; and (3) the $d(Z)$ profile for these moving films should depart markedly from the static film profile.

Figure 3 clearly shows that not only do the points for static conditions fall directly upon the curves for the flowing films, but also that the points for inflow and outflow define the same $d(Z)$ profile. With respect to the latter, consider the

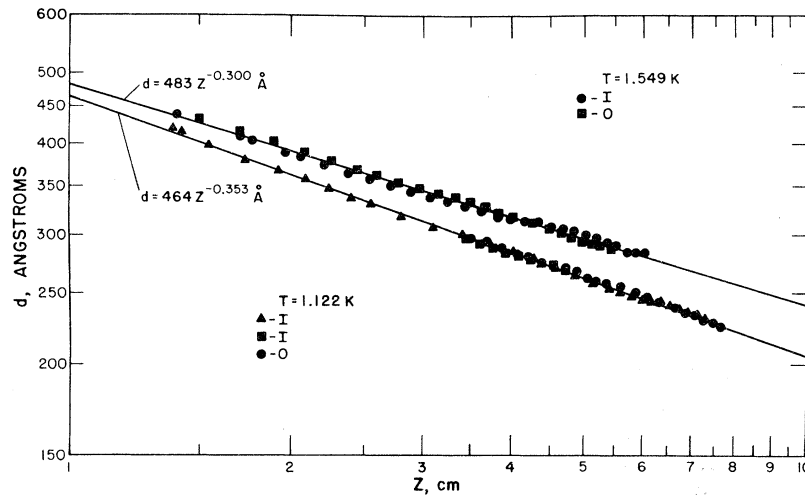


FIG. 3. Log-log plot of d vs Z for the moving and the static film at two temperatures and for both inflow (I) and outflow (O).

data at $T = 1.549$ K. At $Z = 5$ cm, $Q_I = 18.4 \times 10^{-5}$ and $Q_O = 19.0 \times 10^{-5}$ cm²/sec, so that according to Eq. (3) Δd should be 43 and 46 Å, respectively. However, at $Z = 2$ cm, $Q_I = 18.4 \times 10^{-5}$ and $Q_O = 23.0 \times 10^{-5}$ cm²/sec, and the corresponding calculated values of Δd are 80 and 125 Å. The observations obviously indicate that no such disparity exists.

The foregoing arguments provide sufficient evidence for us to conclude that the $d(Z)$ profile for the static and the moving films are equivalent. In addition, we note that in Fig. 3 the slopes of the curves, which provide the exponent in Eq. (2), are quite sensitive to the absolute values of d . In the present experiments, differences in d are much more accurately determined than d itself, the uncertainty in the latter being ± 30 Å. Within these limits, the curves (each of which represents a separate filling of the can) can be renormalized to give the value $n = 3$; and hence within experimental error the $Z^{-1/3}$ law is verified. On the other hand, for the "thicker" films we found $n \approx 2$. Whether these films are really thicker, or merely appear to be so due to surface roughness, cannot be extracted from the data. Similarly unclear is the extent to which surface conditions may affect the observations of d for the thinner films, i.e., a better-prepared surface might have resulted in even thinner films and perhaps a different value of n . It is interesting to note that the observed trend of n increasing with decreasing apparent d is contrary to the most careful theoretical treatment of this problem.¹⁰

For the entire duration of every run the level height in the potential probe matched that of the

liquid inside the bucket, indicating that at all times the potential drop occurred in the film above the probe. (This was contrary to our experiences⁶ with glass beakers, where, especially in inflows, the potential drop often spread several cm down the inside wall.) Consequently we could not precisely define the dissipative region of the flow path and we cannot be sure of what kind of a mixture of dissipative (D) and nondissipative (ND) film flow the capacitor sampled, e.g., the D region may have been highly localized at the inside rim and therefore would have contributed negligibly to the average capacitance observed; or it may have been more spread out. Added to this, we have no independent way of telling whether d changes on passing from ND to D flow. Nevertheless we interpret the experimental results as indicating that at least for ND flow d does not depend upon v_s . In making this deduction we exclude as extremely improbable such possibilities as these: (1) The capacitor always sampled only D flow, and $\Delta d = 0$ for this but not for ND flow; or (2) $\Delta d \neq 0$ for both ND and D flow but with opposite signs so as always to cancel exactly.

It is then clear from these results that something is missing from the Kontorovich formalism⁴: We have no reason to doubt the validity of the $(\rho_s/2\rho)v_s^2$ term in Eq. (1) but instead suggest that at least one additional term is required to balance the kinetic energy contribution. Among the assumptions made in the derivation of Eq. (1) is that nondissipative film flow is both isothermal and isobaric. Possible departure from these conditions seems a likely source of the missing

elements, the search for which constitutes a separate work and will not be described here.

Experiments are in progress to determine what, if any, changes occur in d in the dissipative region of film flow. Results from these will be reported elsewhere along with a fuller account of the present investigation.

The author gratefully acknowledges helpful suggestions about this work gained from discussions with Dr. E. F. Hammel and Professor E. R. Huggins.

*Work performed under the auspices of the U. S. Atomic Energy Commission.

¹See, for example, the review of L. C. Jackson and L. G. Grimes, *Advan. Phys.* **7**, 435 (1958), and references therein.

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⁸All estimates of d take into account that film covers both plates of the capacitor and assume that only atoms in the gap defined by the inner plate contribute to the observed capacitance.

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POSSIBLE FLOW-DEPENDENT TEMPERATURE GRADIENTS IN He FILMS

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(Received 29 December 1969)

Various experiments, especially recent experiments by Keller, clearly demonstrate that the thickness of a helium film is independent of the flow rate Q . This is consistent with our current picture of the hydrodynamics of a helium film if there are temperature gradients in the film which produce a thermodynamic potential $\phi_T = \int SdT$ that cancels the $\rho_s v_s^2/2\rho$ term in the hydrodynamic potential μ . If such temperature gradients exist, they would produce unacceptably large evaporation rates unless the saturated vapor pressure p_{vap} is a function of μ rather than T . If $p_{\text{vap}} = p_{\text{vap}}(\mu)$, then there should be no thinning of the film with changes in Q , and the theory is in complete agreement with Keller's results.

Kontorovich^{1,2} predicted that the thickness δ of a helium film should be given by the relationship

$$\delta = \delta_0(1-q), \quad (1)$$

where

$$\delta_0 = (\beta/g\hbar)^{1/3}, \quad (2)$$

$$q = (\rho/6\rho_s)(Q^2/\beta)\delta_0. \quad (3)$$

The quantity δ_0 is the thickness of a stationary film calculated from a balance of gravitational and van der Waals' force potentials (h is the height above the bath and β is the van der Waals' coefficient whose numerical value for various substrates has been calculated by Schiff³). A change in the thickness $(1-q)$ due to a flow rate

$Q = \rho_s v_s \delta/\rho$ is assumed to arise because the additional term $\rho_s v_s^2/2\rho$ in the hydrodynamic potential should be balanced by a reduction in the van der Waals' potential $-\beta/\delta^3$.

Earlier experiments of Gribbon and Jackson² demonstrated that Eq. (1) predicts too large a thickness change. Recent experiments by Keller⁴ demonstrate that, to within distances of less than the thickness of an atomic layer, there is no change at all in the film thickness when the flow stops. Changes, predicted by Eq. (1) to be as large as 120 Å or roughly 30 atomic layers, did not occur.

Keller's results do, however, agree with the theoretical predictions of the thickness of a stat-