Parallel-Plate Capacitor Measurements of the Superfluid Wall-Film Thickness in a $^3$He/$^4$He Mixture of $^3$He Mole Fraction $x = 0.75$

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Abstract. A thin $^4$He-rich superfluid film coats solid surfaces in contact with bulk normal liquid $^3$He/$^4$He mixtures in a narrow region of the mixture phase diagram at saturated vapor pressure that adjoins the lambda line, the tricritical point, and the $^3$He-rich branch of the phase-separation curve. Recent arguments suggest that near the tricritical point, the thickness of the film is governed not only by van der Waals forces and gravity but also by Casimir and Helfrich forces that limit the thickness as the phase-separation curve is approached. Experimental evidence has been presented both showing and failing to show this phenomenon.

In the present work, parallel-plate capacitor measurements of a mixture having a $^3$He mole fraction $x = 0.75$ show a film thickness of from $\sim 14$ to $\sim 22$ nm as the phase-separation curve is first reached from higher temperature, depending on choice of background subtraction, whereas van der Waals forces limited only by gravity would be expected in our cell to yield a thickness of $\sim 31$ nm, rising in the first few mK below the phase-separation temperature to $\sim 43$ nm. Despite various uncertainties and anomalies, the results suggest that additional forces may be at work.

1. Background
The behavior of liquid helium near a wall is thought to be dominated by the van der Waals attraction of the helium by the wall. In addition to a rise of pressure that takes place in all cases, in $^3$He/$^4$He mixtures there is an increase in the concentration of $^4$He near a wall. This increase has the interesting result that in a narrow normal region of the $^3$He/$^4$He mixture phase diagram at saturated vapor pressure adjacent to the lambda curve, the tricritical point, and the $^3$He-rich branch of the phase-separation curve, a superfluid film exists next to the wall.

In the course of studying this film using a quartz crystal vibrating in shear mode at values of the $^3$He mole fraction $x$ both below and above the tricritical value of $x = 0.67$, we have made capacitance measurements of the film thickness at $x = 0.75$. The behavior of the thickness with temperature near the phase-separation temperature $T_{\sigma+}$, which in our case is 0.76 K, is of some interest because of a recent suggestion that near the tricritical point the growth of the film thickness as $T_{\sigma+}$ is approached from above might be limited by Casimir and Helfrich forces, leading to partial wetting of the solid surface by the $^4$He-rich phase. [1] At first, observations of nonzero contact angles between the phase-separation interface and a sapphire window seemed to support this suggestion, [1] whereas later observations were consistent with a zero contact angle and thus complete wetting. [2]
In the absence of gravity, local continuum theory [3] predicts that a simple van der Waals force (without relativistic correction) acting alone would lead to a film thickness increasing very nearly as \((T - T_{\sigma^+})^{-1/3}\) as \(T_{\sigma^+}\) is approached from above. [4] With gravity present, the film thickness at a height \(h\) above the bottom of the cell would increase very nearly as \((T - T'_{\sigma^+})^{-1/3}\), where \(T'_{\sigma^+}\) is an effective temperature slightly below \(T_{\sigma^+}\), the temperature at which phase separation first occurs at the bottom of the cell. [5] Thus the effect of gravity is to cause the film thickness to remain finite at the onset of phase separation, an effect that must be considered when considering other possible limitations on film thickness.

Some years ago, Sornette and Laheurte reported capacitance measurements of film thickness at \(x = 0.74\) as well as at two larger values of \(x\), using an interdigital capacitor prepared by photolithography of a chromium film evaporated onto a flat substrate. [4] They found that their thickness results could be fitted by a \((T - T_{\sigma^+})^{-1/3}\) dependence, the maximum thickness in their data reported being 7.2 nm. They mentioned possible problems with measurements close to \(T_{\sigma^+}\).

2. Experiment
The present results were obtained by combining measurements made in separate runs with two different horizontal parallel-plate capacitors. Measurements made with a capacitor having a gap of \(\sim 200 \mu\m\), in which film effects were negligibly small, served as a reference to which measurements made with a capacitor having a gap of 15.4 \(\mu\m\) could be compared. Each capacitor was made from glass microscope slides on which gold electrodes with underlayers of chromium were deposited by vacuum evaporation. Three-wire capacitance measurements were made using a General Radio Model 1615-A Capacitance Bridge at either 1 or 10 kHz. The cell temperature was regulated to 0.1 mK.

A reference capacitance \(C_{ref}(T)\), representing the capacitance that would have been measured with the narrow-gap capacitor were no film present and a uniform mixture extended all the way to the wall, was determined in two different ways. The first, yielding \(C_{ref1}(T)\), involved combining empty-cell and filled-cell measurements using the wide-gap capacitor, assuming negligible wall-film effects, with empty-cell measurements on the narrow-gap capacitor, for each capacitor making small approximate corrections for fringe fields in the glass. The second, yielding \(C_{ref2}(T)\), involved combining measurements of the dielectric constant of the mixtures by Kierstead, [6] again assuming negligible wall-film effects, with empty-cell measurements on the narrow-gap capacitor. Overall, these determinations were in quite good agreement, but in the detail needed for the present work there were significant differences as described below.

Due to drifts of unknown origin, our filled-cell capacitance measurements with the narrow-gap capacitor \(C(T)\) on different days needed to be shifted by several times \(10^{-4}\) pF to bring them into agreement. In addition, we were forced to reject some of our data for \(C(T)\) that showed an anomalous temperature dependence between \(T_{\sigma^+}\) and 0.84 K. Fig. 1 shows an example of our data for \(C(T)\) from five series recorded on different days, involving shifts of as much as 0.0004 pF relative to one another. \(C_{ref1}(T)\) exceeded \(C(T)\) and \(C_{ref2}(T)\) lay below \(C(T)\) by several tens times \(10^{-4}\) pF. We chose to apply constant additive corrections to each of them so that the difference \(C(T) - C_{ref}(T)\) yielded a film thickness as described below of \(\sim 1.4\) nm at 0.84 K, as suggested by modeling based on the van der Waals force. [5] Neither of the resulting relationships between \(C(T)\) and \(C_{ref}(T)\) was highly satisfactory. \(C_{ref1}(T)\) fell below \(C(T)\) above 0.90 K, where film effects should be very small, by \(\sim 2\times10^{-4}\) pF. \(C_{ref2}(T)\) agreed well with \(C(T)\) above 0.90 K but yielded a somewhat peculiar dependence of wall-film thickness on temperature as described below. Fig. 1 shows portions of these two determinations of \(C_{ref}(T)\).

Film thickness was calculated from \(C(T) - C_{ref}(T)\) using the approximation that the film consists of a uniform mixture of mole fraction at the \(^4\)He-rich branch of the phase-separation curve and that the dielectric properties of the mixture are altered by the replacement of bulk...
mixture by such a film. This approximation neglects the small variations in $x$ and in pressure $P$ that take place outside of and within the film well away from the wall, and also the large changes in $x$ and $P$ that occur in a thin layer next to the wall, although these neglects are compensated for to a large degree by the shifts of $C_{ref}(T)$ described above. Modeling suggests that any error in thickness from this source increases as the film thickness increases but remains less than 1 nm. Fig. 2 shows an overall view of our results both below and above the phase-separation temperature, and Fig. 3 shows the details of the results above the phase-separation temperature, for both determinations of $C_{ref}(T)$.

The figures show a thickness that increases rapidly as the temperature is lowered to $T_{\sigma^+}$ from above, reaching values of \( \sim 14 \) and \( \sim 22 \) nm at $T_{\sigma^+}$ for $C_{ref1}$ and $C_{ref2}$, respectively. As the temperature is lowered further, and the mixture in the capacitor follows the $^3$He-rich branch of the phase-separation curve, the thickness for $C_{ref1}$ continues to increase, reaching a value of \( \sim \)
52 nm at $T = 0.65$ K, while that for $C_{ref2}$ remains relatively constant near 20 nm. These figures make clear the large sensitivity of our results to the choice of $C_{ref}$. The large slope at 0.84 K makes the results based on $C_{ref2}(T)$ seem less plausible than those for $C_{ref1}$ at temperatures above $T_{σ+}$. As shown in Fig. 3, the thickness $d$ based on $C_{ref1}$ can be fitted quite well there by the form

$$d = \frac{A}{(T - T'_{σ+})^{1/3}} + B$$

Here $A = 1.0 \text{ nm-K}^{1/3}$ and $B = -0.8 \text{ nm}$. $T'_{σ+}$ lies $\sim 0.3$ mK below $T_{σ+}$.

3. Discussion

The plane of the capacitor gap lies 3.8 mm above the very lowest part of the cell. If we assume that the film thickness is governed solely by van der Waals forces and gravity, we calculate that the film thickness at $T_{σ+}$, when the bulk $^4$He-rich phase first appears at the bottom of the cell, should equal $\sim 31$ nm, assuming a van der Waals potential that includes a relativistic correction of the form $\Omega(d) = -\alpha/(d^3(1+\beta d))$ with $\alpha = 33 \text{ K-(layers)}^3$ and $\beta = 8.2 \times 10^{-3} \text{ (layers)}^{-1}$. [7] One layer equals 0.36 nm. As the temperature is lowered from 1 to 3 mK below $T_{σ+}$, the phase boundary should rise 2.5 mm in a narrow neck of the cell, yielding a film thickness of $\sim 43$ nm. As the temperature is lowered further, the thickness should continue to rise, reaching $\sim 46$ nm at 0.65 K. Note that in our cell, the phase-separation boundary never rises to the level of the capacitor gap in the temperature region studied.

In the vicinity of $T_{σ+}$, our experimental values for the thickness based on $C_{ref1}(T)$ lie substantially below these estimates. If we were to lower the value of the reference capacitance to increase our value for the thickness of the film at $T_{σ+}$, we would increase our value for the thickness at 0.84 K by an even greater amount, inconsistent with van der Waals forces and gravity acting alone. The thickness based on $C_{ref2}(T)$, while larger than that based on $C_{ref1}(T)$ at $T_{σ+}$, has the wrong temperature dependence above $T_{σ+}$ to be due to van der Waals forces and gravity alone.

Thus despite various uncertainties and anomalies our results raise the possibility that, in addition to the van der Waals force and gravity, other forces, such as the Casimir and Helfrich forces proposed by Ueno et al., [1] may be at work in determining film thickness, although our value of $x$ may lie rather far from the tricritical value for these forces to be important. An analysis of our results in terms of such forces remains to be attempted. The value of A that we have determined in Eq.(1) is somewhat larger than the value of 0.67 nm$^2$K$^{1/3}$ derived by interpolation for $x = 0.75$ from similar fits made by Sornette and Laheurte. [4] Further experimental work would appear to be fruitful, in particular, measurements for values of $x$ nearer the tricritical value. Capacitive measurements near the tricritical point have also been made with interdigital capacitors by Nash et al. [8] They mention finding film thicknesses of several 100’s of nanometers at the $^3$He-rich branch of the phase-separation curve, possibly consistent with no other effects than gravity and van der Waals forces being at work in a capacitor much closer to the bottom of the cell than ours.

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