Journal of Low Temperature Physics, Vol. 106, Nos. 5/6, 1997

Rapid Communication

Measurement of the surface tension of superfluid ^{4}He at low temperature by capillary wave resonances

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(Received August 12, 1996; revised November 4, 1996)

The surface tension of liquid ^{4}He is determined from the frequencies of micron wavelength capillary waves. The extrapolated zero temperature value, $\sigma = 375 \pm 3\ \mu Jm^{-2}$, is in agreement with the pioneering static capillary rise determination but 6% higher than the more recent surface tension gravity wave measurements. Flow in the meniscus in this latter experiment is shown to mimic a surface tension correction to the dispersion relation there used which is of the same sign and magnitude as the discrepancy.

1. INTRODUCTION

The surface tension σ of liquid ^{4}He is a fundamental quantity for surface and interface properties. The variations of σ with temperature have been extensively studied and measured with high accuracy.[1,2] Up to 0.7 K, these experimental measurements are well explained by the ripplon contribution to the surface free energy as first proposed by Atkins[3] in 1953. Surprisingly, the absolute value of σ at T=0 remains poorly known as the principal determinations, the capillary rise method of Zinov'eva and Boldarev,[4] giving $\sigma = 378\ \mu Jm^{-2}$ and the surface tension gravity wave method used by Iino et al,[5] giving $\sigma = 354\ \mu Jm^{-2}$, exhibit a significant 6% difference. This large discrepancy has recently taken on greater importance with the growing interest in the problem of wetting on weak-binding substrates, in which the

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0022-2291/97/0300-0565$12.50/0

contact angle and the wetting temperature are directly related to σ.

Within this context, a clarification of the experimental situation appears necessary. The new determination of σ presented in this paper would seem to satisfy the conditions for accuracy and reliability. The analysis of our own data has led us to present also a critical examination of the previous experiments.

2. EXPERIMENTAL SETUP

The experiment was originally designed to study ripplon propagation in the micron wavelength range; the experimental cell is shown in fig. 1 of ref.[6] and the methods are described in detail there.

The measurements are done on a superfluid film covering a horizontal array of interdigital capacitors (IDC's) which consists of interlocking fingers, of width 2 μm separated by 2 μm, patterned in chromium on a monocristalline sapphire substrate. This geometry defines a fundamental periodicity of wavelength $\lambda_0 = 8\mu m = 2\pi/k_0$ in the (x,y) plane. Each finger is 2 mm long and the total IDC array consists of 1000 fingers.

The structure is also used to manipulate the liquid using the dielectric ponderomotive effect. When it is biased with a constant voltage V_{DC}, the electric field around the fingers attracts the dielectric helium. The equilibrium thickness $d(x)$ of liquid above the capacitor satisfies[9]

$$(h+d) - \frac{(\epsilon-1)\epsilon_0}{2\rho g}E^2 - \frac{\kappa^4}{d^3} - \frac{\sigma}{\rho g}\nabla^2_{x,y}\, d = 0 \qquad (1)$$

E is the electric field just below the liquid surface, g the acceleration due to gravity, ρ and ϵ the helium density and dielectric constant and σ the surface tension ($\rho = 145\, kgm^{-3}$, $\epsilon = 1.057$ at 0K). The parameter κ, describing the substrate to liquid helium van der Waals (vdW) interaction is approximately $0.7\,\mu m$ and $h \approx 8\, mm$ is the distance of the capacitor plane above the bulk helium surface.

The periodic potential produced by the IDC may be expressed in terms of the fundamental wavevector $k_0 = 2\pi/\lambda_0$ by

$$V(x,z) = V_{DC} \sum a_p \sin((2p+1)k_0 x) e^{-(2p+1)k_0 z}$$

,from which we may write the electric field intensity to order a_2 :

$$\begin{aligned} E^2(x,z) &= k_0^2 V_{DC}^2 (a_0^2 + 9a_1^2 e^{-4k_0 z} + 25 a_2^2 e^{-8k_0 z}) e^{-2k_0 z} \\ &+ k_0^2 V_{DC}^2 (6a_0 a_1 + 30 a_1 a_2 e^{-4k_0 z}) e^{-4k_0 z} \cos(2k_0 x) \\ &+ k_0^2 V_{DC}^2 (10 a_0 a_2) e^{-6k_0 z} \cos(4k_0 x) + \cdots \end{aligned} \qquad (2)$$

The coefficients a_p are determined by the basis cell of the structure. For finger width equal to separation, $a_{2n+1} = 0$ and $a_0 \simeq 1/2$.

The first term in Eq.(2) induces a uniform increase of the equilibrium depth d_0 of the liquid which can be varied from the vdW film thickness to a few microns by scanning V_{DC}.

The oscillatory term in Eq.(2) deforms the liquid surface for a discrete set of wavenumbers $2nk_0$. It has been shown[6] how this allows one to excite capillary waves using an AC voltage $V_{AC}e^{i\omega t}$ applied in addition to the bias voltage, $V_{AC} \ll V_{DC}$; the bias controls the depth d_0, whereas the oscillatory component of the ponderomotive force ($\propto V_{AC}V_{DC}e^{i\omega t}$) excites surface waves at an angular frequency ω. If it coincides with the angular frequency of capillary waves on the film at wavenumber $k = 2nk_0$, constructive interference leads to resonant creation of waves. A capacitance bridge measurement (fig. 2 of ref.[6]) shows a resonance in power absorption and an anomalous dispersion at this point.

Irrotational surface excitations of an inviscid incompressible fluid of uniform depth d_0 propagate according to the dispersion relation:

$$\omega^2(k, d_0) = (\rho g' + \sigma k^2)\frac{k \tanh(kd_0)}{\rho} \tag{3}$$

$$g' = g\left[1 + 3\frac{\kappa^4}{d^4} - \frac{(\epsilon - 1)\epsilon_0}{2\rho g} \cdot \left.\frac{\partial E^2}{\partial z}\right|_{z=d_0}\right] \tag{4}$$

Here g' is a gravity-like term incorporating vdW and electrostrictive forces.

In the present experiment, we concentrate on the two harmonics $n = 1$ and $n = 2$ corresponding to ripplon wavelengths $\lambda_1 = 4\ \mu m$ and $\lambda_2 = 2\ \mu m$. At the high wavenumbers of the present experiment, the surface term σk^2 in Eq.[3] is dominant. The resonance frequency then becomes a very direct measure of the surface tension σ. The small corrections due to finite depth and to g' will be considered below.

3. ANALYSIS OF DATA

Our experimental procedure makes it useful to write directly the frequencies in terms of V_{DC} and to extract from the asymptotic behaviour of $\omega(2nk_0, d_0(V_{DC}))$ the value of $\omega_\infty(2nk_0)$ in the bulk limit.

For that purpose, we consider the $kd_0 \gg 1$ limit. As d_0 and κ^4/d_0^3 are both negligible compared with h, the equilibrium film thickness equation (1) becomes:

$$h = \frac{(\epsilon - 1)\epsilon_0}{2\rho g} k_0^2 a_0^2 V_{DC}^2 e^{-2k_0 d_0} \tag{5}$$

Using this to write $\partial E^2/\partial z$, in the large depth limit Eqs.(3,4) become :

$$\omega^2(k, d_0) = (\rho g' + \sigma k^2)\frac{k}{\rho}(1 - 2e^{-2kd_0}) \tag{6}$$

$$g' = 2gk_0h \tag{7}$$

Combining (5) and (6) the dispersion relation can be expressed in terms of V_{DC} instead of d_0:

$$\omega(2nk_0, V_{DC}) = \omega_\infty(2nk_0)\left[1 - \left(\frac{2\rho gh}{(\epsilon-1)\epsilon_0 k_0^2 a_0^2}\right)^{2n} V_{DC}^{-4n}\right] \tag{8}$$

$$\omega_\infty(2nk_0)^2 = (\rho g' + \sigma(2nk_0)^2)\frac{2nk_0}{\rho} \tag{9}$$

This variation of ω with V_{DC} provides an accurate way of determining $\omega_\infty(2nk_0)$ from the linear extrapolation $V_{DC}^{-4n} \to 0$ of the plot $\omega(2nk_0)$ versus V_{DC}^{-4n}. Figure (1) shows that Eq.(8) is satisfied over a large interval of V_{DC} and leads to a precise determination of $\omega_\infty(2nk_0)$ without further assumptions. We used the fact that a given structure allows us to measure two modes with wavenumber exactly in the ratio 2:1.

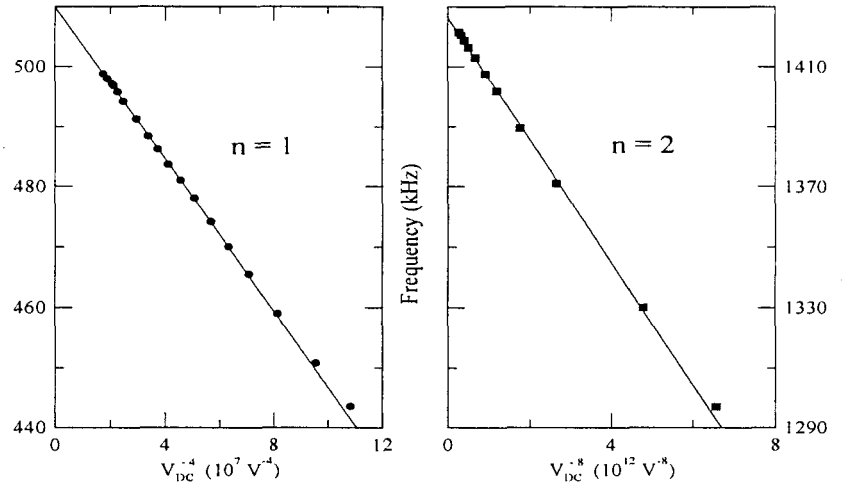

Fig. 1. resonance frequencies for $\lambda = \lambda_0/2$ and $\lambda_0/4$ ($\lambda_0 = 8\mu m$) versus V_{DC}^{-4} and V_{DC}^{-8} respectively. The asymptotic values $\omega_\infty(2k_0) = 510\,kHz$ and $\omega_\infty(2k_0) = 1426\,kHz$ are deduced from these curves.

Precise measurement of the two asymptotic frequencies as explained above leads to independant determination of the two unknown quantities g' and σ, the large surface term being obtained with better relative accuracy than the small gravity like term. We point out that our technique can provide the temperature variation of σ at least in the low temperature region ($T < 700\,mK$), where the ripplons are not overdamped.[6]

Fig.(2) shows our measured temperature variation of σ to agree well with the Atkins ripplon contribution law,[7] which we then use to extrapolate to zero temperature. This leads us to the $T \rightarrow 0$ value :

$$\sigma = 375\ \mu J m^{-2}$$

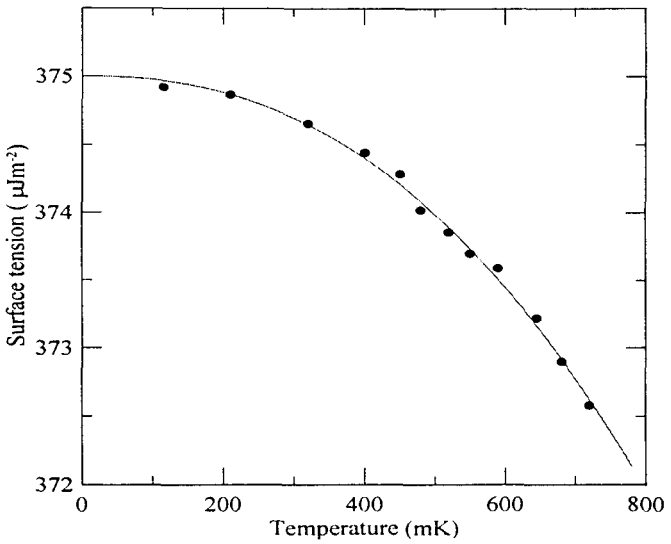

Fig. 2. Variation of the surface tension with temperature. The points are deduced from the resonance frequencies of the n=2 mode at fixed depth and at various temperature. The full line represents the Atkin's law.

The measurements were performed with ^{4}He purified by the heat flush method ($X_3 < 10^{-8}$) and it was shown in two different ways that there was no effect of ^{3}He condensation at the surface : Any ^{3}He condensation at low temperature should decrease σ. The fact that the surface tension agrees fairly well with the Atkins law as shown in Fig.(2) (monotonic increase with decreasing temperature) is an indication that we can rule out any effect of isotopic contamination at the ^{4}He surface. Second, upon purposely adding ^{3}He to the cell up to $X_3 = 3\,10^{-3}$, we did not observe any change on the ripplon frequencies at low temperature. This feature seems to indicate that the transport of ^{3}He through the superfluid ^{4}He film linking the free surface to the thick film is very slow and thus that the ^{3}He atoms actually never reach the region above the bias IDS, even after a waiting time of one day.

4. DISCUSSION OF ERROR SOURCES

The three determinations give

σ	$= 378 \pm 5\ \mu Jm^{-2}$	Zinov'eva et al[4]
σ	$= 354 \pm 0.4\ \mu Jm^{-2}$	Iino et al[5]
σ	$= 375 \pm 3\ \mu Jm^{-2}$	Present experiment

We first examine the various sources of error involved in our experiment and then we will discuss the expected accuracy in the previous determinations. In our experiment neither the absolute value of the gravitational acceleration g nor that of the vdW constant κ is relevant as their contributions to the error are entirely negligible. The fundamental wavenumber k_0 is a geometrical factor which was first determined in the early stages of fabrication of the structure. It was also independently measured by optical diffraction from the grating formed by the IDC. At room temperature, we measured $k_0 = (784.0 \pm 1) mm^{-1}$. This value becomes $k_0 = (784.5 \pm 1) mm^{-1}$ near 0 K according to the thermal expansion of monocristalline sapphire. Uncertainties in the resonant frequency result from the width of the resonance. For the $\lambda = 2\mu m$ mode, this width was less than 2 kHz, in good agreement with the expected quality factor defined by the length of the structure. $\delta\omega/\omega$ is taken to be conservatively $2\ 10^{-3}$, including the error arising from the extrapolation to ω_∞. Summing all these independent contributions gives the error estimate.

In the static capillary rise experiment of Zinov'eva and Boldarev, the rise h of the liquid in the capillary is simply related to the diameter b of the capillary by $\rho g h = 2\sigma/b$. The liquid level can be measured within a few micrometers giving a $\delta h/h$ accuracy of the order of $5 \cdot 10^{-3}$. The main difficulty comes from the determination of the diameter b of the capillary. It can be non uniform or not reproducible from run to run (as pointed out by Atkins and Narahara[1]) due to possible contaminant remaining after the temperature cycle. An accuracy $\delta b/b$ of the order of 10^{-2} seems reasonable. Then, this kind of experiment yields $\delta\sigma/\sigma \approx 10^{-2}$ which is comparable to the absolute fluctuation of σ from run to run mentioned in ref.[1].

It is more difficult to understand the 6% discrepancy with the recent determination by Iino et al.[5] The disagreement is so much greater than estimated statistical error that one must look for a more fundamental cause.

Their approach was to measure with high accuracy the resonance frequencies of successive surface wave modes in a cylindrical box. Using the usual dispersion relation for a flat surface of liquid of uniform depth d :

$$\omega^2 = gk \tanh(kd)(1 + k^2 l_c^2) \tag{10}$$

they deduce the capillary length $l_c = \sqrt{\sigma/\rho g}$ by measuring a set of 3 cor-

responding modes at each of 2 different depths on the hypothesis that the wavenumbers are the same. Thus, with no assumption about how the $\{k\}$ are related to the radius R, it is possible to deduce l_c (and $\{k\}$ and the two d). They argue that any meniscus effects are thereby eliminated by hypothesizing that the sole effect of the meniscus is to change the relation between wavenumber and radius and that this effect is independant of the depth. The procedure looks beyond reproach.

We argue differently : that the meniscus has no effect on the wavenumber but that it changes the dispersion relation, in such a way as to imitate a reduction in surface tension.

The argument is simpler to present in rectilinear geometry; although numerical factors are slightly different in circular geometry, the physics is the same. We consider a box with infinite length in the y direction and vertical walls in the z direction at $x = -R$ and $x = +R$. The surface of the liquid is described by the equation $z = \eta(x) + d + \zeta(x,t)$ where d is here the uniform depth, $\eta(x)$ the meniscus shift closer to the walls and $\zeta(x,t)$ the deformation due to the excitation. If ϕ is the velocity potential, eigenstates of this box are

$$\phi = \phi_k \cos(kx) \cosh(kz) \tag{11}$$

with $k = n\pi/R$. The resonant wavenumbers are not affected by the meniscus as the continuity equations are satisfied on the walls and at the bottom. From the continuity equation at the surface we write $\phi_k = \dot{\zeta}_k/k \sinh(kd)$ to express the energy per unit length of the mode in the form

$$\mathcal{H}_k = \mathcal{T}_k + \mathcal{V}_k = \frac{1}{2}\mathcal{M}_k\dot{\zeta}_k^2 + \frac{1}{2}\mathcal{M}_k\omega_k^2\zeta_k^2 \tag{12}$$

where $\mathcal{T}_k$ is the kinetic energy, $\mathcal{V}_k$ the potential energy and $\mathcal{M}_k$ the hydrodynamic mass. The potential energy

$$\mathcal{V}_k = \frac{1}{2}(\sigma k^2 + \rho g)\zeta_k^2 R \tag{13}$$

is not modified by the meniscus to first order. The kinetic energy

$$\mathcal{T}_k = \frac{1}{8}\rho k \phi_k^2 \int_{-R}^{+R} [\sinh(2k[d + \eta(x)]) - 2k\eta(x)\cos(2kx)]\, dx \tag{14}$$

on the other hand is modified to order $k^2 l_c^2$ by the fluid velocity in the extra-volume enclosed by the meniscus. As the extension of the meniscus is of the order of l_c near the walls and $kl_c \ll 1$, we can consider that $\cos(2kx) \approx 1$ and the kinetic energy becomes

$$\mathcal{T}_k = \frac{1}{2}\frac{\rho}{k\tanh(kd)}\left[1 + \frac{l_c^2 k \tanh(kd)}{R}\right]\dot{\zeta}_k^2 R \tag{15}$$

This correction to the kinetic energy resulting from the motion of liquid in the volume of the meniscus can be interpreted as a correction to the hydrodynamic mass

$$\mathcal{M}_k = \frac{\rho R}{k \tanh(kd)} + \rho l_c^2 \tag{16}$$

It will be noted that it is only because the velocity is vertical in the meniscus region that the increment is simply the extra mass contained under the meniscus. In the Iino experiment, the depth of helium in the box is such that $\tanh(kd) \approx kd$ and the hydrodynamic mass correction modifies the dispersion relation to perfectly mimic a surface tension effect.

$$\omega_k^2 = gk \tanh(kd) \left[1 + k^2 l_c^2 (1 - \frac{d}{R})\right] \tag{17}$$

The analysis procedure used by Iino then extracts not l_c^2 but $l_c^2(1 - d/R)$. Using the values quoted by the authors, $d \approx 0.6\,mm$ and $R = 12\,mm$, they would have determined 0.95σ instead of σ. This is roughly the observed discrepancy. Our goal is not to find the exact bias but only to point out that a precise determination of σ cannot be made without taking into account the meniscus effect which depresses the apparent surface tension. It does so, however, in a way which is proportional to the surface tension itself, so that the relative behaviour with temperature, for example, is unchanged.

5. CONCLUSION

The present new experimental determination of the surface tension of ^{4}He at low temperature is made in conditions where the surface tension strongly dominates gravity. The high quality factor of the resonances and the accurate independent determination of the wavenumber lend confidence to the absolute value of the surface tension at low temperature. Setting the helium film thickness with ponderomotive forces offers high stability and reproducibility of the system and confident determination of the asymptotic bulk capillary wave frequencies. The result is in clear agreement with the pioneering capillary rise determination. It is of considerable interest that the ab initio functional density calculation[10] is in excellent accord with this new determination. We believe it is important too in providing a precise starting point for the growing field of helium wetting.

ACKNOWLEDGMENTS

We thank K. O. Keshishev, S. Balibar and J. Treiner for their helpful discussions.

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