

Optical Measurement of Wetting by ^3He - ^4He Mixtures Near their Tri-Critical Point

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We have measured the contact angle of the interface of phase-separated ^3He - ^4He mixtures against a sapphire window using an interferometric method in an optical cryostat. Our measurement has been performed at saturated vapor pressure, along the phase separation line and near T_i , the temperature of the tri-critical point. We have found that the contact angle is finite, contrary to what was generally accepted up to now, i.e., complete wetting by the ^4He -rich phase. Furthermore, this angle does not tend to zero when the temperature approaches T_i . This is a remarkable exception to "critical point wetting," a phenomenon which is usually observed near ordinary critical points.

1. INTRODUCTION

It was generally accepted that, when a phase separated liquid mixture of ^3He and ^4He is in contact with a wall, the latter is completely wet by the ^4He rich liquid. This means that the ^3He - ^4He interface has a zero contact angle with the wall.¹ This was attributed to the van der Waals attraction which is stronger on the densest phase.² However, a recent measurement³ showed that this contact angle is non-zero on an epoxy surface. Since it was done with an MRI technique whose accuracy was not very high in the region of interest, it appeared necessary to repeat this measurement with a different technique. In this article, we present an optical measurement of the contact angle on a sapphire substrate. As we shall see, our measurements are more accurate than the ones previously obtained with the MRI

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technique. We have now obtained a clear evidence that, in the temperature range from 0.81 to 0.86 K, close to the tri-critical point at $T_t = 0.87$ K,⁴⁻⁶ the contact angle is non-zero. Moreover, it increases as the temperature approaches T_t instead of vanishing as one would expect if the usual phenomenon of "critical point wetting" occurred.⁷⁻¹⁰

Consider a binary liquid mixture which phase separates into a concentrated "c-phase" and a diluted "d-phase" below a certain critical temperature T_c . As is well known, the contact angle θ of the c-d interface against a substrate "s" is related to the energy σ_i of the c-d interface through the Young-Dupr e relation¹¹:

$$\cos \theta = \frac{\sigma_{sc} - \sigma_{sd}}{\sigma_i} = \frac{\delta\sigma}{\sigma_i}. \quad (1)$$

Here, σ_{sc} and σ_{sd} are the respective free energies of the interfaces between the substrate and the c-phase (resp. the d-phase). In his historical paper, Cahn⁷ explained that, as T_c is approached, the interfacial energy in the denominator vanishes more rapidly than the difference $\delta\sigma$ in the numerator of Eq. (1). This looks obvious if one admits that $\delta\sigma$ is proportionnal to the difference in concentration $\delta X = (X_c - X_d)$. Indeed, it is well established that, for ordinary critical points, the critical exponent of σ_i is 1.28,^{12,13} while the exponent of δX is 0.33. In our case of a tri-critical point at a temperature T_t , the exponents are respectively 2 and 1,^{13,14} so that the same argument could apply. As a consequence, as one approaches T_t , $\cos \theta$ should increase and reach the value 1 at a wetting temperature T_w lower than T_t . As for the contact angle θ , it should of course decrease towards zero at T_w , according to these arguments.

This is the behavior which had been observed by Heady and Cahn⁹ and later verified by Bonn *et al.*¹⁰ However, in 1981, de Gennes remarked that, in the presence of long range forces, "critical point wetting" does not necessarily occur.¹⁵ In 1985, Nightingale and Indekeu¹⁶ further explained that the contact angle could remain non-zero up to the critical point (partial wetting). Around the same time, Ross and Kornbrekke¹⁷ published measurements of a contact angle tending to 90 degrees as the temperature approached the critical point, but this was later considered as an artefact of their rather primitive observation technique by Moldover and Schmidt.¹⁸ We thus believe that we have found the first experimental evidence of an exception to "critical point wetting." The purpose of this article is to describe the experimental techniques which allowed us to obtain these results. We have also included some comments on the anomalous behaviour of helium mixtures, but quantitative interpretations are delayed for a later publication.¹⁹

2. EXPERIMENTAL METHOD

We used a dilution refrigerator with optical access to cool down liquid ^3He - ^4He mixtures well below the tricritical point T_t . The cell could be seen from the outside of the cryostat through four windows. Two of them are infrared filters respectively attached to the 0.7 and 77 K shields. The two other windows at 4 and 300 K have an anti-reflection coating. The two infrared filters eliminated thermal effects such as possible thermal gradients in the cell. The cryostat is attached to an optical table which is standing on four air spring vibration absorbers. Interferometric images of the mixtures were obtained with a He-Ne laser and a beam expander, conventional imaging optics and a CCD camera. All our optics were on the table, outside the cryostat.

As shown in Fig. 1, our experimental cell was made of a copper body with two sapphire windows. It was surrounded by vacuum and thermally anchored to the mixing chamber of our refrigerator. The copper parts were

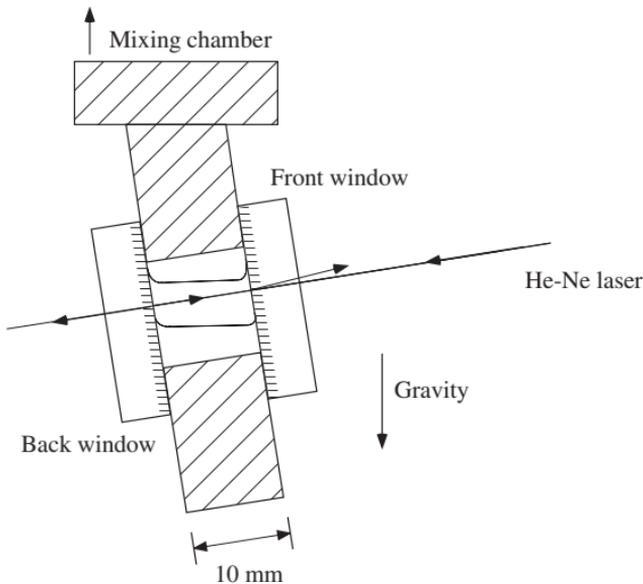


Fig. 1. A schematic drawing of the experimental cell. The cell body (hatched area) was made of pure copper and thermally anchored on the mixing chamber of the refrigerator. It was closed by two sapphire windows which formed an optical interferometric cavity. The surfaces of the windows were treated anti-reflection outside, and coated to have a 0.15 reflectivity inside. The sample space was $11\text{ mm} \times 11\text{ mm} \times 10\text{ mm}$ and was tilted by 8.78° with respect to the gravity. We did not fill the cell completely, so that, from top to bottom, we could see the gas phase, the c-phase which is concentrated in ^3He , and the diluted d-phase. The sample was illuminated by an expanded He-Ne laser beam.

used as a thermal link to the sample and the two windows formed an interferometric cavity. The inside of the windows were treated to have a 15% reflectivity. The surface coating was a dielectric layer of ZrTiO_4 , with a thickness of 47.3 nm, an rms roughness less than 5 nm, and an optical index equal to 2.01.²⁰ The outside surfaces had an antireflection coating. These reflectivities were chosen to observe well contrasted fringe patterns with a sinusoidal intensity. The observable sample space inside the cell was nearly cubic, 11 mm \times 11 mm \times 10 mm in size. The windows were sealed with indium and stainless steel clamps. Unfortunately, this induced inhomogeneous stresses which slightly bent the windows and the fringe pattern (see Fig. 2).

Near each window, the liquid interface formed a meniscus whose profile was analyzed to measure the interfacial tension σ_i and the contact angle θ . In order to distinguish one meniscus from the other, the cell was tilted by about 10° with respect to the gravity (a careful measurement of this angle showed that it was 8.78°). We sent the laser from above and observed the fringe pattern of the meniscus at the back window. By doing so, the path of light after crossing the ^3He - ^4He interface was shorter than if we had chosen the meniscus on the front window, and distortion of the fringe pattern associated with refraction effects were minimized. Images were captured with an ordinary CCD camera and stored in a computer. The light escaping through the back window was absorbed by a black screen and was not reflected back to the sample space, so that it did not affect the interferometric image. We avoided using a mirror in place of the back window, in order to minimize absorption of light near the meniscus and possible thermal gradients there. Furthermore, a mirror would have given less contrasted fringes.

We prepared a gas mixture with the critical concentration as follows. We first mixed ^3He and ^4He approximately with the right proportions. During a first run, we then accurately measured the ^3He concentration from the phase separation temperature and the temperature dependence of the height of the c-d interface. Then, after warming up, we accurately adjusted the concentration to the exact critical value $X_3 = 0.675$ and kept this mixture for the rest of the experiment. The sample space was filled at a constant regulated temperature (0.6 K), and the condensation process was carefully monitored optically. Then the amount of helium in the cryostat was kept constant till the end of the measurements. Since some of the sample space was kept gaseous in the whole temperature range, the mixture remained at saturated vapor pressure. The temperature was measured with a calibrated Ge thermometer attached to the bottom of the cell and regulated within ± 0.2 mK during one measurement, typically 20 minutes. The equilibrium of the phase separation was confirmed by the observation

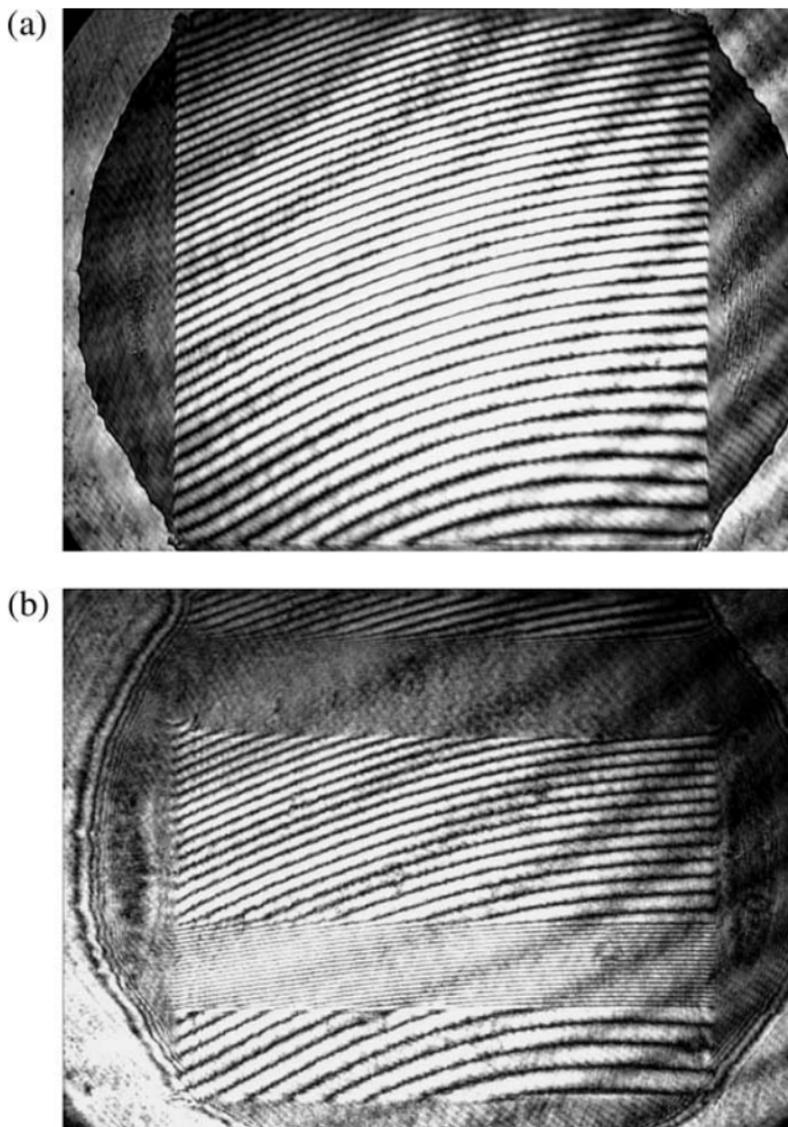


Fig. 2. Interferometric pictures of the sample space. The upper picture (a) shows the sample space before filling and the lower picture shows the same space once filled with phase-separated mixtures at 0.852 K. Due to inhomogeneous stresses the sapphire windows were slightly bent and the observed fringes were not straight. The lower picture (b) shows the existence of two interfaces. The upper interface separates the gas phase from the ^3He rich "c-phase," and the lower interface separates the c-phase from the diluted "d-phase." The gas to c-phase interface region appears dark because of large refraction effects. We used a lens to magnify images of the upper part of the c-d interface region and extract the profile of the meniscus and the contact angle of the interface on the back window.

that the c-d interface stopped moving. This took several hours very close to the tri-critical temperature $T_t = 0.87$ K.

3. RESULTS

Interferometric pictures of the whole sample space are shown in Fig. 2. The upper picture shows the cell before filling. Since the two windows were not strictly parallel to each other, there existed a fringe pattern even when the cell was empty. The averaged misalignment of the two windows was deduced from the interfringe distance and found equal to about 0.06° . Due to the slight bending of the windows, the fringes were not straight lines and their spacing varied slowly with position. In the lower picture of Fig. 2, the sample space was at 0.852 K and filled with a phase-separated liquid mixture. There were two regions corresponding to two different interfaces, the upper black one for the gas-liquid interface, the lower one for the c-d interface (hereafter c always refers to the phase which is concentrated in ^3He , and d to the diluted one). The large difference in the refractive indices caused a large refraction of the light at the gas-liquid interface so that no light from this region could come back to the camera and the upper interface appeared dark. Most of the c-d interface region corresponds to a flat horizontal interface. We had to look into the small transition region where the fringes changed shape in order to extract the profile of the meniscus of interest. As can be seen from Fig. 2, fringe lines in the lower right corner of the sample space were nearly horizontal and this made our image analysis rather difficult in this region. To avoid this problem, we kept the c-d interface as high as possible in the cell, and, for each temperature, we analyzed magnified fringe patterns at three fixed positions, respectively on the left, at the center and on the right of the contact line in the cell.

Figure 3 shows a typical magnified fringe pattern at 0.841 K and the c-d interface profile we extracted from it. The magnification of the picture was $\times 4.75$ and its size was 768 pixels \times 576 pixels. In the fringe pattern of Fig. 3, the upper part was the c-phase and the lower part was the flat c-d interface. The region of most interest lies where fringes deviate from straight lines. The contact line was almost horizontal on the picture and fringes corresponding to the flat c-d interface were also nearly horizontal.

From such fringe patterns, we extracted the profile of the c-d meniscus by studying the sinusoidal variation of the light intensity in the vertical direction.²² The interface profile has to be invariant by translation in the direction parallel to its contact line with the sapphire window. The phase of the light intensity was fitted with a sinusoidal function of the coordinate perpendicular to the line. The orientation of the contact line itself was

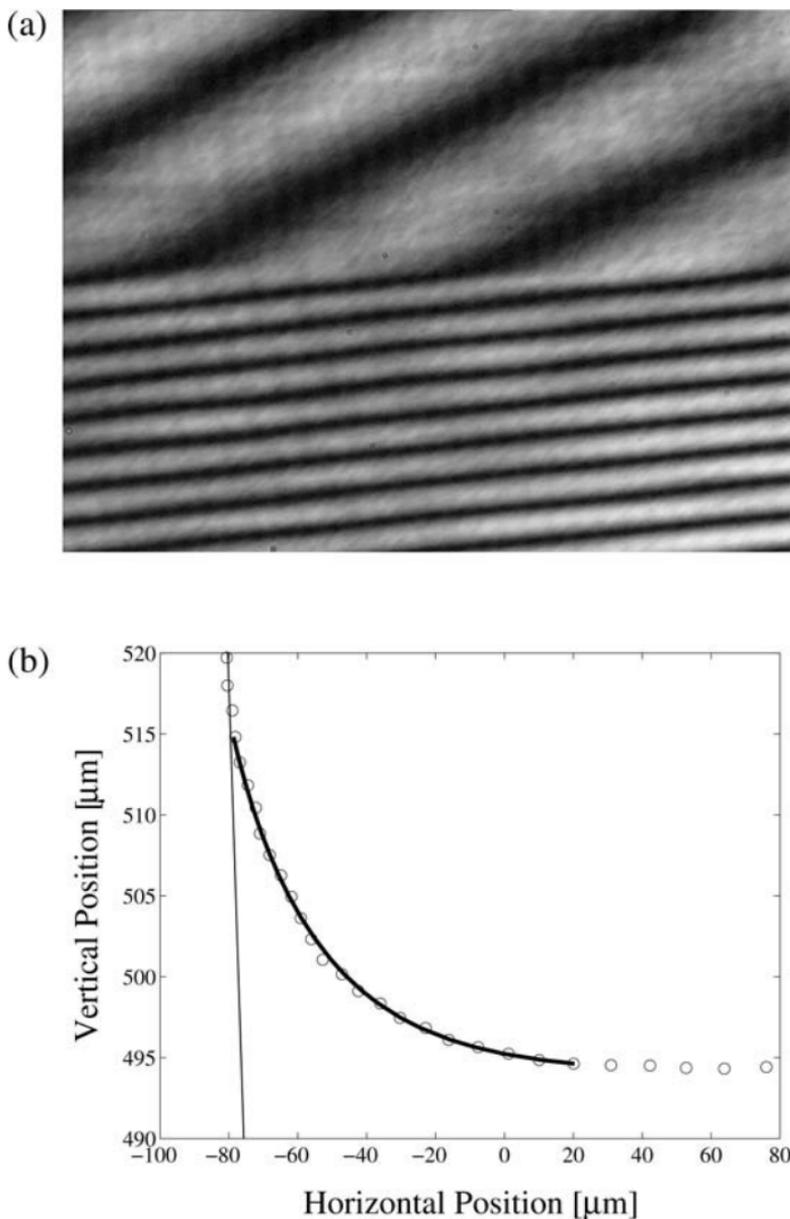


Fig. 3. Fringe pattern of the c-d interface at 0.841 K and its extracted profile. Circles are experimental measurements of the position of the c-d interface and the thick solid line is a fit with a solution of the Laplace equation. The thinner solid line shows the position of the back window. Note that the c-d interface profile in the graph is expanded in the vertical direction.

determined by optimizing the fit. A simple relation between the fitted phase change $\phi_{\text{fitted}}(d, T)$ and the c-d interface profile $h(d, T)$ is given by:

$$\phi_{\text{fitted}}(d, T) = 4\pi \frac{n_c(T) d \tan \alpha}{\lambda} + 4\pi \frac{[n_d(T) - n_c(T)] h(d, T)}{\lambda}, \quad (2)$$

where d is a distance measured perpendicularly to the contact line from an arbitrary position along the back window, T is the temperature, $n_c(T)$ and $n_d(T)$ are the refractive indices of the c-phase and of the d-phase, α is the small wedge angle between the two windows and λ is the wavelength of the He-Ne laser light, i.e., 632.8 nm. The first term of the right-hand side of Eq. (2) describes the background pattern due to the misalignment of the windows, and the second term comes from the c-d interface itself. In order to subtract the background, we made a polynomial fit of its pattern. We used the values of $n_c(T)$ and $n_d(T)$ near T_i from the measurements by Kierstead.²³ After subtraction of the background, the phase was constant above the contact line, despite some noise in its measurement. As shown in Fig. 4, a fit with a straight line through the data points in this region allowed us to determine the exact position of the window surface. The typical error bar on this position was found to be 5 microns. Below the contact line and sufficiently far away from the window the c-d interface has to be horizontal. This is because the capillary length ℓ_i is less than about 300 μm , its value at the absolute zero,²⁴ that is always small compared to the depth of the cell. As we shall see below, it becomes very small close to the tri-critical point at T_i . Thanks to a careful adjustment of the temperature T_i and of the exact tilt angle of the back window (8.78°), the image analysis gave a horizontal c-d interface far from the wall. For the tri-critical temperature, we found $T_i = 0.8695 \pm 0.0005$ K, in good agreement with other data.^{4, 5, 23, 25}

The refraction of the laser at the c-d interface caused two problems. First, the optical path was longer than if the light had kept perpendicular to the back window everywhere. Secondly, since the light came out slightly tilted from the cell, the exact fringe positions were slightly displaced in the interface region. Since the refraction depends on the difference between $n_c(T)$ and $n_d(T)$, these distortions became larger as the temperature went down, causing an apparent variation of the angle of the back window as a function of temperature below 0.8 K. At low temperature, the c-d interface was also in the lowest part of the cell where the fringe bending made the image analysis difficult. Therefore we decided to exclude data points below 0.810 K from the present analysis. Above 0.810 K the refraction effects were negligible.

Circles in the graph of Fig. 3 correspond to the position of the c-d interface. In this graph, the thick solid line corresponds to a fit with a

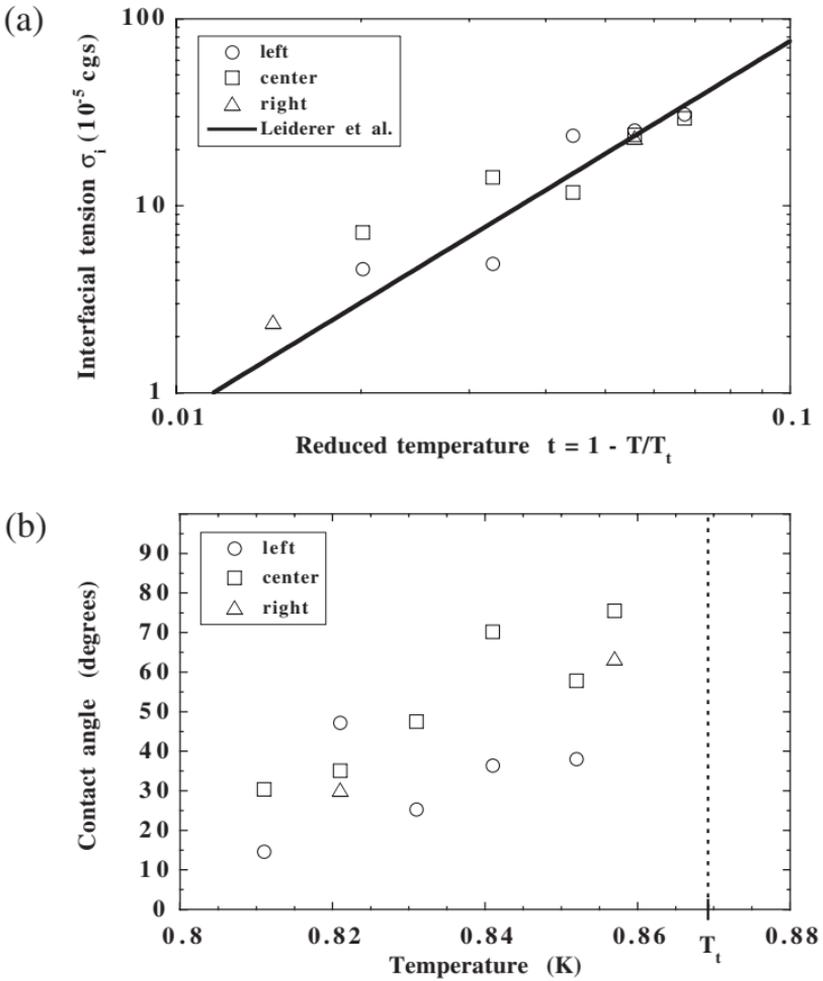


Fig. 4. Temperature dependence of the interfacial tension σ_i and the contact angle θ . The labels “left,” “center” and “right” correspond to different horizontal positions along the contact line in the cell. The solid line in graph (a) is a fit through the results of Leiderer *et al.*⁵

solution of the Laplace equation. From the fit, we obtained the capillary length ℓ_i and the contact angle θ . This capillary length is related to the interfacial tension σ_i of the c-d interface by²⁶:

$$\ell_i = \sqrt{\frac{2\sigma_i}{[\rho_d(T) - \rho_c(T)]g}}, \quad (3)$$

where $\rho_c(T)$ and $\rho_d(T)$ are the respective densities of the c- and d-phase and g is the acceleration of gravity.

As explained above, we analyzed three pictures at three different positions for each temperature. This was done to verify that we had correctly subtracted the effect of the bending of the windows. In the three pictures each c-d interface was fitted to obtain σ_i and θ . The temperature dependence of σ_i and θ is shown on Fig. 4. For this, we calculated $\rho_c(T)$ and $\rho_d(T)$ from the Clausius-Mossotti relation, Kierstead's results²³ and an extrapolation of the phase separation curve from the data of Sydoriak.²⁷ Since T_i was 0.867 K in Ref. 5, we plotted σ_i as a function of the reduced temperature, $t = 1 - T/T_i$ in the upper graph of Fig. 4. Our measurements of σ_i agree well with Leiderer's results $\sigma_i = 0.076t^2$ (solid line on Fig. 4).⁵

As can be seen, we have found that the contact angle θ is finite in the temperature range from 0.81 to 0.86 K. The error bar in our measurements of the profile is about 5 μm . This uncertainty has to be compared with the capillary length which varies from 84 μm at 0.81 K to 33 μm at 0.86 K. Of course, it is the vanishing of both the capillary length and the difference in refractive indices $n_d - n_c$ which make measurements close to T_i rather difficult.

We have carefully studied the accuracy of our determination of the contact angle θ . We have found that, in the end, the main source of uncertainty

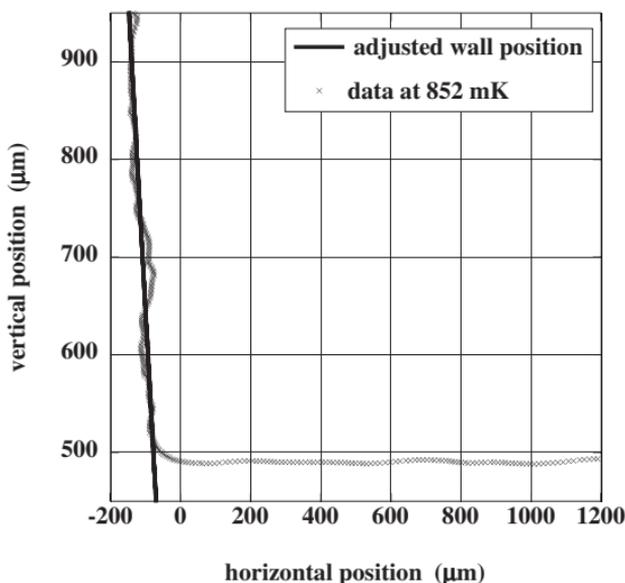


Fig. 5. The exact position of the wall is found from a fit through the data points above the meniscus. It is determined within 5 microns. Its tilt angle (8.78 degrees) is optimized so that, far away from the wall, the c-d interface is horizontal. The same tilt angle and the same wall positions were found at all temperatures. The region of interest for the contact angle measurement is the small rounded portion of the meniscus very close to the wall.

is the accuracy of the wall position. Since it is typically 5 microns, we have tried to fit our profiles not only with the wall at its adjusted position, but also after forcing the wall to be 5 microns away in the direction which would decrease the value of the contact angle. As shown in Fig. 6, we found a change from 38° with the wall at its right position to 24° when the wall was displaced by 5 microns. We thus believe that the typical accuracy of our measurements is 15° for the contact angle. Although this is clearly not as good as desired, it is definitely better than what was previously achieved with the MRI technique. In their previous work,³ Ueno *et al.* had found a large scatter of data points above 0.8 K, typically between 0 and 90° .

It is now clear not only that θ is non-zero but also that it increases as the temperature approaches the tri-critical point, instead of decreasing as the usual theory of critical point wetting would predict. The tendency to increase near T_i is not strong enough to show any critical point drying transition, since the contact angle does not seem to tend to 180° either.

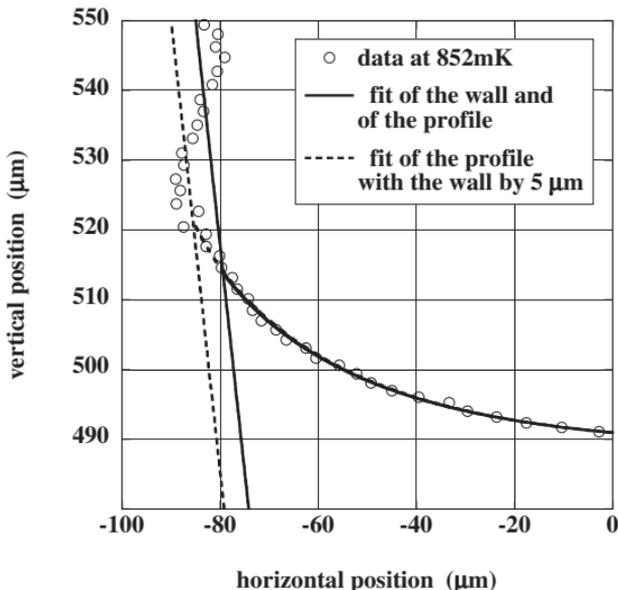


Fig. 6. An analysis of the accuracy in our determination of the contact angle θ . The data points correspond to $T = 0.852$ K. The solid lines correspond to the fit of the wall and to the fit of the profile with the wall at its fitted position. The broken lines correspond to another fit where the wall is forced to be 5 microns away from its fitted position. Away from the wall, the two fits overlap because they lead to values for the interfacial tension which are very close to each other. As for the contact angle, in the first case, we found $\theta = 38^\circ$ while in the second case, we found 24° . This means that the typical error bar on our measurements is about 15° at 852 mK, very close to the tri-critical point.

Although we had several experimental difficulties such as the refraction effects and the bending of windows, our determination of the interfacial tension σ_i agrees with previous ones, and we have carefully analyzed possible sources of errors, so that we believe that our measurements of the contact angle $\theta(T)$ are reliable.

4. DISCUSSION

We have found no complete wetting nor any critical point wetting transition near the tri-critical point of ^3He - ^4He mixtures. This is doubly surprising. Let us first comment on the existence of a finite contact angle. One previously thought that the d-phase wet solid substrates completely because of the experiment by Romagnan *et al.* followed by the interpretation by Sornette and Laheurte.^{2,28} Indeed, they showed that, when a ^3He -rich homogeneous mixture is cooled down towards T_{ps} where phase separation occurs, a ^4He -rich film forms between the substrate surface and the bulk c-phase. They measured the thickness l of this film and they found that l increased proportionally to $(T - T_{ps})^{-1/3}$. This behaviour is consistent with the existence of a van der Waals attraction by the substrate. However, they measured this behavior only up to 20 atomic layers, about 60 Å. If l really increased up to a macroscopic value, one should observe complete wetting by the d-phase at coexistence, that is when the macroscopic c- and d-phases are in equilibrium. However, if another long range force exists, and if this force is an effective attraction by the substrate of the c-d film surface, the increase of l might saturate at larger values, a few hundred Å for example. As will be explained in a future article,¹⁹ we think that this other force is due to the confinement of critical fluctuations in the film thickness. This "critical Casimir effect"^{29,16,30} is important in the region of the tri-critical point where the correlation length is large.

Finally, since the contact angle increases with temperature instead of decreasing, it means that the interfacial tension difference $\delta\sigma$ is not proportionnal to the concentration difference δX . We also believe that this can be explained by considering the critical Casimir forces. Of course, it would be important to repeat these experiments at higher pressure and see if the same partial wetting is observed. In future experiments we will also change the geometry in order to decrease the refraction effects and be able to measure the contact angle at lower temperature.

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