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Critical Casimir forces and anomalous wetting

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Abstract. We present a review of critical Casimir forces in connection with successive experiments on wetting near the critical point of helium mixtures.

Keywords. wetting, critical, Casimir

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1. Introduction

In 1978, M. Fisher and P.G. de Gennes [1] considered a critical system whose fluctuations are confined between two plates a distance L apart. They noticed that, with two identical plates, there is a singular contribution of order $k_B T/L^2$ to the free energy per unit area of the system so that there is an attractive force between the plates which should be of order $-2k_B T/L^3$ near the critical point at the temperature T_c. In analogy with the standard Casimir effect, which originates in the confinement of electromagnetic fluctuations between two electrodes, the phenomenon considered by Fisher and de Gennes is now known as the "critical Casimir effect". It has been studied by several groups and recently reviewed by M. Kardar and R. Golestanian [2] and by M. Krech [3] (among others). Despite all the work already done, it seems to us that it is not yet fully understood: the amplitude of the critical Casimir force has not yet been calculated with the boundary conditions corresponding to experimental situations; furthermore, Garcia and Chan demonstrated the existence of this effect with a series of two remarkable experiments [4,5], but some of their quantitative results appear somewhat puzzling to us.

In order to interpreat their experiments on wetting by helium mixtures, Ueno *et al.* [6] related the critical Casimir effect to another critical phenomenon, which is known as "critical point wetting" and was first predicted by J.W. Cahn [7]. This relation had first been proposed by M.P. Nightingale and J.O. Indekeu who had already noticed the potential interest of liquid helium as a model system for the whole issue [8,10]. Consider a binary liquid mixture below its critical temperature T_c: it is separated in two phases with an interface in between. The contact angle θ of this interface may be non-zero away from T_c, but Cahn predicted that, as T tends to T_c, θ should vanish and the wall be completely wet by one of the two critical phases. Several theoretical studies have confirmed that critical point wetting is very general and it has been observed in several experiments with different systems [11–13]. However, de Gennes pointed out the importance of long range forces in Cahn's situation and opened the possibility of exceptions to critical point wetting if such long range forces are present [14].

T. Ueno *et al.* studied ^{3}He-^{4}He liquid mixtures in contact with a wall in a series of two experiments: a first one in Kyoto [15] where magnetic resonance imaging (MRI) was used, and a second one in Paris [16] where the contact angle was measured optically. These two experiments showed that, apparently, an exception to critical point wetting had been found in the physics of helium mixtures. In their third article, Ueno *et al.* [6] proposed that critical Casimir forces were the long range forces responsible for this exception. By using Garcia's measurements, Ueno *et al.* found that the critical Casimir forces had the right sign and the right magnitude to explain the non-wetting behavior found in the Paris experiment.

Together with further developments of the theory, these interesting findings urged us to extend and confirm our experimental results. One of the important questions had been raised by Kardar and Golestanian [2] and concerned a new contribution to the critical Casimir force. Indeed, they explained that such a force could exist even if the medium was not close to a critical point. In the case of superfluids, the order parameter has a phase, so that so-called "Goldstone modes" fluctuate whatever the temperature, in the whole temperature domain where long range correlations exist. A similar effect was predicted for liquid crystals [17]. For liquid helium, a force originating in Goldstone modes should thus exist in the whole temperature region where it is superfluid. As a consequence, a liquid helium film adsorbed on a wall would be thinner if it is superfluid than if it is normal, even if T is much lower than T_λ, the superfluid transition temperature. In our context of wetting by helium mixtures, the Goldstone mode contribution to the Casimir force could have led to a non-zero contact angle of the ^{3}He-^{4}He interface with a wall at low temperature.

In order to test Kardar's prediction, we changed the geometry of our experimental setup, so that measurements could be done at lower temperature without too much difficulties with optical refraction effects. The results of this ongoing experiment are not yet published [18]. Its preliminary results indicate that the contact angle is in fact zero at low temperature (complete wetting). As we shall see, it does not mean that Kardar's predition is wrong, only that the magnitude of this effect is too small to be observed in an indirect measurement such as ours. Furthermore, we have tried to reproduce Ueno's former results near the critical point of helium mixtures, and, this time, we have found that the angle is very likely to be zero. This now means that there was probably an experimental artefact in Ueno's series of experiments. It also means that the amplitude of the critical Casimir force is probably smaller than what was deduced from Garcia's measurements. We thus hope that Garcia's experiment can also be reproduced, the comparison of its results with theory extended to the region below T_c, and its relation with the critical point wetting by helium mixtures more critically discussed.

The somewhat difficult goal of this review is to clarify the present status of this confusing situation, particularly the questions concerning the magnitude of the critical Casimir force and its connexion with critical point wetting.

2. Critical Casimir forces

Fisher and de Gennes only gave an order of magnitude for the amplitude of the critical Casimir effect at the critical temperature T_c. One usually writes the critical Casimir force as

$$F(L,T) = \frac{k_B T}{L^3}\vartheta(L/\xi) \tag{1}$$

where the "scaling function" $\vartheta(L/\xi)$ depends on temperature and on the thickness L through its ratio to the bulk correlationlength ξ. Near T_c, ξ diverges proportionnally to $t^{-\nu}$ where $t = (T/T_c - 1)$ is the reduced temperature and $\nu = 0.67$ for ordinary critical points ($\nu = 1$ for tri-critical points). Following the seminal paper by Fisher and de Gennes, several theoretical works have brought important information about the critical Casimir force:

1- The sign of the force is given by the sign of the scaling function, and it depends on the symmetry of the boundary conditions. If they are symmetric, ϑ is negative and the force is attractive; on the opposite, if the boundary conditions are antisymmetric, ϑ is positive and the force is repulsive.

2- The magnitude of the force is generally considered as universal, in particular at the bulk critical temperature T_c, where its value is twice the "Casimir amplitude" Δ , which is the universal value of Θ (the similar scaling function appearing in the singular contribution to the free energy) at T_c. Furthermore the Casimir amplitude depends on the dimension N of the order parameter. From the work of Nightingale and Indekeu [9,10] and Krech and Dietrich [19], it appears that Δ is roughly proportionnal to N. For example, it is expected to be twice as large for a superfluid transition ($N = 2$) as for the phase separation of a usual liquid mixture ($N = 1$). It also depends on the boundary conditions, more precisely on their nature, not on the exact details of surfaces [19]. These conditions can be periodic, or the order parameter can vanish at the boundary ("Dirichlet" boundary conditions) or its derivative can vanish ("von Neumann" conditions).

3- With Dirichlet boundary conditions, the critical temperature in the film is significantly displaced with respect to the bulk critical temperature T_c, and the maximum of the scaling function $\Theta(L/\xi)$ is expected to be rather different from the Casimir amplitude Δ. In fact, as far as we know, there exists no calculation of the scaling functions both below and above T_c for Dirichlet boundary conditions. According to Krech and Dietrich [19], Δ is much smaller for Dirichlet boundary conditions than for periodic ones, but it does not mean that the maximum amplitude of ϑ is also much smaller, mainly that the temperature at which this maximum is reached is displaced (as far as we understand).

The calculation of $\Theta(L/\xi)$ has been performed above T_c by Krech and Dietrich [19], using an ϵ-expansion method. For periodic boundary conditions and below T_c, it has been more recently calculated by G. Williams in the frame of his vortex loop-model for liquid helium [24]. According to Williams, his calculation below T_c matches nicely with Krech's calculation above T_c, the Casimir amplitude being about -0.15, close to the maximum amplitude of the scaling function $\Theta(L/\xi)$.

In their first experiment, Garcia and Chan observed the thinning of a pure liquid helium film near the superfluid transition at T_λ. This film was adsorbed on a copper electrode and most of the thinning occured in a small temperature region near T_λ. They analyzed it in terms of the critical Casimir effect and extracted a scaling function $\vartheta(L/\xi)$ which was very similar in shape with calculations, for example the recent ones by Dantchev and

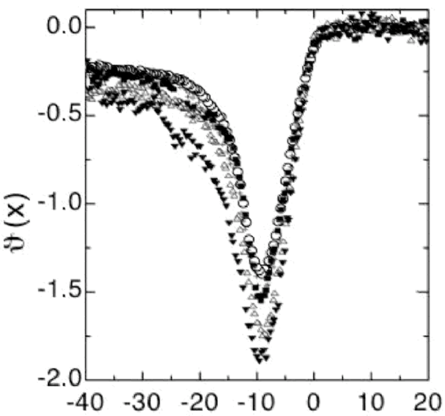

Figure 1. The scaling function $\vartheta(x)$ as measured by Garcia and Chan. The horizontal coordinate is $x = tL^{1/\nu}$ where t is the reduced temperature and L the film thickness, ν the critical exponent of the bulk correlation length ξ; it is measured in Å$^{1/\nu}$ units. These results could only be compared with Krech's theory above the critical temperature T_λ , i.e. for $x > 0$, where $\vartheta(x)$ has a very small tail. Different symbols correspond to different film thicknesses.

Krech [20]. Garcia's scaling function displays important features which deserve several comments:

1- The maximum amplitude of ϑ does not occur at the bulk critical temperature T_c. This was expected because the order parameter for superfluidity vanishes on both sides of the superfluid film, so that the superfluid transition temperature is depressed in the film: $T_c^{film} < T_c^{bulk}$. It occurs significantly *below* T_c, for $x = tL^{1/\nu} \approx -10$ (the reduced temperature is taken negative below T_c. Note also that, in both articles by Garcia and Chan, the horizontal coordinate $x = tL^{1/\nu} = (L\xi_0/\xi)^{1/\nu}$ is not dimensionless, but close to $(L/\xi)^{1/\nu}$ since L is taken in Å and the quantity ξ_0 is about 1 Å [27]). The magnitude of the scaling function *above* T_c is very small, as predicted by Krech and Dietrich [19]. Garcia and Chan claim that their measurement of ϑ agrees with the calculation, but this only concerns the small tail at $T > T_c$, where the signal/noise ratio is poor, while most of the observed effect occurs below T_c.

2- Garcia and Chan found that $\vartheta(L/\xi)$ reaches maximum negative values which vary from -1.5 to -2 as a function of the film thickness L. This is doubly surprising, firstly because a dependence on L seems to contradict the predicted universality, secondly because no calculation has ever found such large amplitudes for ϑ. In the various situations which have been calculated, the theoretical results are 5 to 50 times smaller. This is a serious problem which needs further studies: new experiments should identify the origin of the L-dependence, and ϑ should be calculated below T_c with Dirichlet boundary conditions.

3- They also found indications that the scaling function does not tend to zero in the low temperature limit, away from T_c. One possible explanation for this is the confinement of Goldstone modes invoked by Kardar and Golestanian [2]. The amplitude of the Goldstone mode contribution looked too small to explain the rather large negative value of $\vartheta(T \to 0)$ found by Garcia and Chan, but a more recent calculation by R. Zandi *et al.* proposes that, the film surface being mobile, a contribution from third sound modes at the surface of a

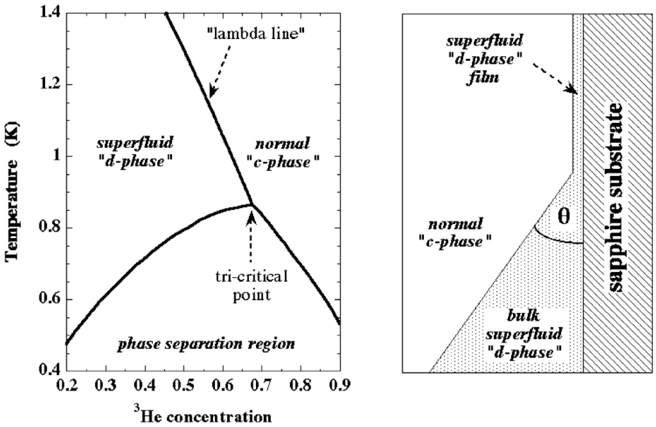

Figure 2. The phase diagram of liquid helium mixtures (left) and a schematic representation of the partial wetting of a wall by the phase separated mixture (right).

superfluid film should be added to the one coming from Goldstone modes, so that the total attractive force acting on the film surface is larger than first calculated [21]. One could even imagine that phonons also contribute to the force at low temperature [22].

3. Critical point wetting and Ueno's experiments

Following Cahn's argument [7], let us consider a binary liquid mixture near its critical temperature T_c. Below T_c, the mixture is separated into a concentrated "c-phase" and a diluted "d-phase." In the case of partial wetting, the contact angle θ of the cd-interface against a substrate "s" obeys Young-Dupré relation [25]:

$$\cos\theta = \frac{\sigma_{sc} - \sigma_{sd}}{\sigma_i} = \frac{\delta\sigma}{\sigma_i}, \tag{2}$$

where σ_{sc}, σ_{sd}, and σ_i are interfacial free energies between s and c-phase, s and d-phase, and c- and d-phases respectively. If one assumes that $\delta\sigma$ is proportional to the difference in concentration ($\delta X = X_c - X_d$), on finds that, for ordinary critical points, $\delta\sigma \propto \delta X \propto t^{0.33}$. Since the interfacial tension $\sigma_i \propto t^{1.28}$, one finds that the numerator in Eq. 2 vanishes more slowly than the denominator, so that the cosine increases as T approaches T_c, and the contact angle reaches zero at some temperature below T_c: a "critical point wetting" transition occurs. In the case of a tri-critical point, as is the case for liquid helium mixtures because phase separation occurs at the same temperature T_t as superfluidity (see Fig. 2), the (mean field) exponents are respectively 2 and 1 [26,27], so that the same reasoning should apply and complete wetting should occur as T approaches T_t. The above argument is too simple but more careful analysis have proved that is is qualitatively correct in the absence of long range forces [8].

In their two successive experiments, Ueno *et al.* did not find Cahn's critical point wetting when studying liquid helium mixtures. Below their tri-critical temperature T_t, these

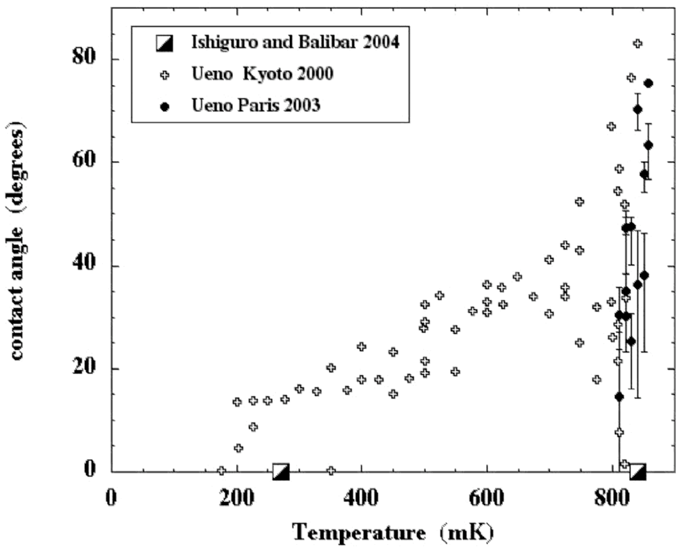

Figure 3. The contact angle θ of the ^{3}He-^{4}He interface with a wall was found non-zero in Ueno's experiments in Kyoto (crosses). They used a magnetic resonance imaging (MRI) technique whose accuracy was not good near the tri-critical point, in view of the large scatter of data points in this temperature region. Agreement with this anomalous behaviour was found in a later experiment by Ueno *et al.* in Paris. However, in their more recent experiment, Ishiguro and Balibar found θ = 0 both at low temperature and near the critical point.

mixtures are separated in a "c-phase" which is concentrated in ^{3}He, and a "d-phase" which is diluted in ^{3}He, consequently rich in ^{4}He. In the Kyoto experiment [15], Ueno *et al.* measured the profile of the cd-interface near a wall made of epoxy glue. They used a magnetic resonance imaging technique (MRI) and found a non-zero contact angle (see Fig. 3). In the Paris experiment [16], Ueno *et al.* measured the interface profile with an optical interferometric technique. They also found that the contact angle θ was non-zero below T_t; moreover, they found that θ increased as T approached T_t(see Fig. 4). This was not compatible with critical point wetting, so that, in their theoretical article [6], Ueno *et al.* reconsidered Cahn's argument after identifying three long range forces which are present in their experimental situation.

The van der Waals force is attractive on atoms and ^{4}He atoms occupy a smaller volume than ^{3}He atoms because their quantum fluctuations are weaker (their mass is larger). As a result, the van der Waals attraction on the d-phase is stronger than on the c-phase, and a c-phase is always separated from a solid wall (in Fig. 2, a sapphire window) by a film of d-phase. Being attractive on atoms, the van der Waals field induces an effective force which is *repulsive* on the film surface. In the absence of other long range forces, a finite thickness film would only exist off-equilibrium, but as equilibrium is approached the film thickness would diverge and complete wetting by the d-phase would occur. Romagnan *et al.* [28,29] found some experimental evidence for this, but their measurements were limited to thicknesses up to about 80 Å only. The sketch in Fig. 2 corresponds to a situation where another long range force acts on the film. Being attractive, this other force

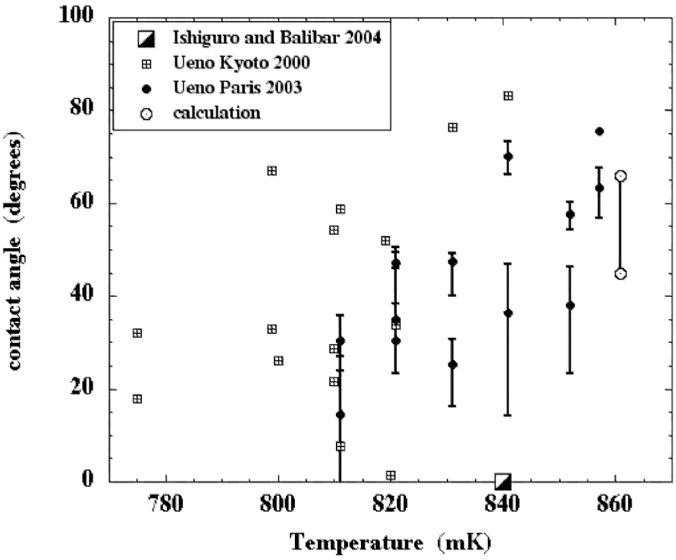

Figure 4. The contact angle near the tri-critical point. This is an enlargement of the graph shown in the previous figure. The experiment by Ueno *et al.* in Paris (2003) showed that θ increased instead of tending to zero as T approched the tri-critical temperature T_t. However, this anomalous behavior was not confirmed in the more recent experiment done by Ishiguro and Balibar, who found θ = 0.

is able to counterbalance the van der Waals field and to limit the film thickness, so that the macroscopic contact angle is non-zero.

In the presence of such long range forces, the way to calculate the surface energies, consequently the contact angle, is to integrate the so-called "disjoining pressure" $\Pi(L)$ which is nothing but the sum of the forces acting on the film surface [31]:

$$\cos(\theta) = \frac{\sigma_{sc} - \sigma_{sd}}{\sigma_i} = 1 + \frac{\int_{l_{eq}}^{\infty} \Pi(l)dl}{\sigma_i} \qquad (3)$$

In addition to the van der Waals force which is known, Ueno *et al.* explained that there is a critical Casimir force. This is because the d-phase film is superfluid, while the c-phase is normal. The order parameter of superfluidity is non-zero inside the film but it has to vanish on both sides. This symmetric vanishing should produce an attractive Casimir force on the film surface. Since there exists no calculation with such Dirichlet boundary conditions yet, Ueno *et al.* [6] used Garcia's measurement of $\vartheta(L/\xi)$ to calculate the contribution of the critical Casimir effect to the disjoining pressure. They also included the Helfrich force, which is repulsive, due to the cutoff of capillary modes at long wavelength by the presence of the nearby wall [30]. At a reduced temperature $t = -0.01$ below T_t, they found that the critical Casimir force was stronger than the two others in the thickness range from 0 to 400 Å (see Fig. 6). According to this calculation, the equilibrium film thickness was thus 400 Å, and Ueno *et al.* could calculate the contact angle by integration of the disjoining pressure (Eq. 3). They found 45 degrees, in good agreement with their measurement (see Fig. 4). Moreover, they argued that the Casimir force could be twice as

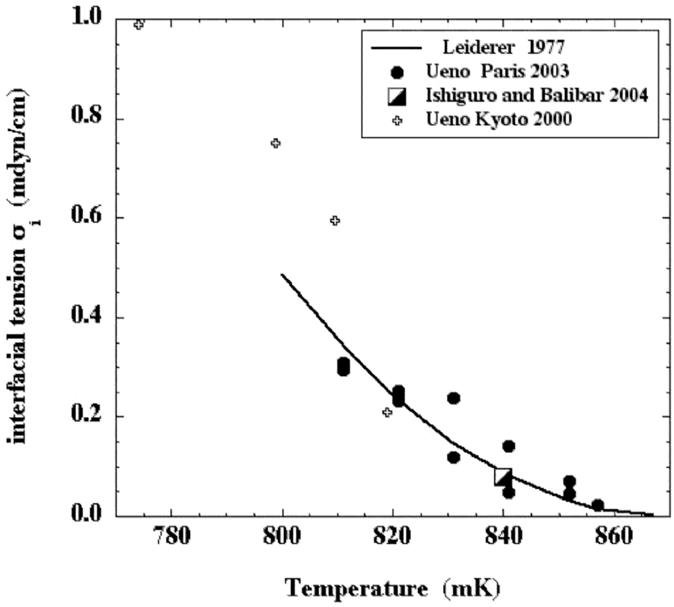

Figure 5. Various measurements of the interfacial tension σ_i between the c- and the d-phase in the vicinity of the tri-critical temperature T_t = 870 mK. Within the error bars, all data agree with the t^2 critical behavior measured by Leiderer *et al* in 1977.

large in their experiment as in Garcia's experiment, because it is a tri-critical point instead of an ordinary critical point such as the lambda point of pure liquid helium 4. According to this argument, they found that the contact angle was 60 degrees at $t = 0.01$, in even better agreement with their experimental results. Furthermore, Ueno *et al.* also extracted values for the interfacial tension σ_i and found good agreement with previous measurements (see Fig. 5). Although the support of experiments by theory and vice versa looked rather strong, we decided to repeat the experiment in a different geometry and with more careful analysis of the interferometric images. As we shall see, our new results show that there was probably an artefact in Ueno's experiment, also that the amplitude of the Casimir force might be smaller than what Ueno *et al.* deduced directly from Garcia's measurements.

4. The new experiment in Paris

In Ueno's experiment, the angle of incidence of the laser beam was large on the cd-interface, that is far from normal. As a consequence, as soon as the difference in optical index between the c- and the d-phase was large, refraction at the cd-interface induced a substantial difference in orientation between the ingoing and the outgoing beams. The fringe pattern was distorted and the calculation of the interface profile too difficult. This is the reason why Ueno *et al.* could not measure the contact angle below about 0.8 K. In order to do this, and be able to look for a possible effect of Goldstone modes on the wetting by helium mixtures, we rotated the cell by nearly 90 degrees, so that the incidence was now

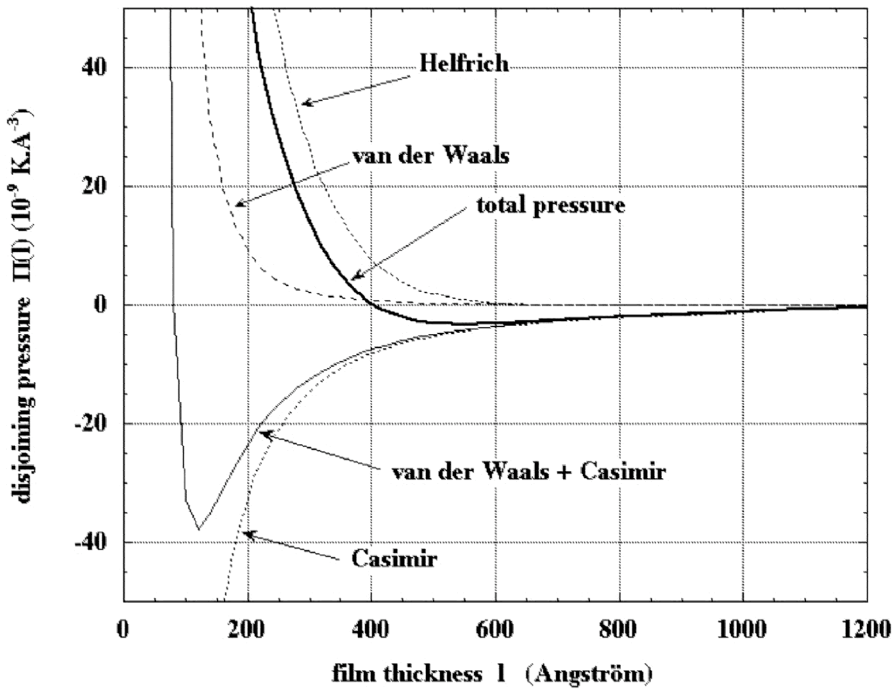

Figure 6. The 2003 calculation by Ueno *et al.* of the various forces acting on the film surface. The amplitude of the Casimir force was taken directly from the measurement by Garcia and Chan in 1999. It was apparently stronger than the van der Waals effective force which is positive, meaning repulsive on the film surface. After adding the Helfrich force, Ueno *et al.* found a total disjoining pressure crossing zero at l = 400 Å, the equilibrium film thickness. A finite film thickness means partial wetting and quantitative agreement was found with the measurements by Ueno *et al.* in 2003.

close to normal. We also tried to adjust the optical cavity more carefully, in order to obtain straight fringes. The interface profile is obtained by substracting the optical path without interface from the optical path with the interface (see Fig. 7). This is rather delicate and needs a very accurate knowledge of the fringe pattern in the absence of interface.

Close to T_t where the index difference is very small, any error in the substracted background produces large errors in the calculated profile. We believe that this is the origin of artefacts in the profiles found by Ueno *et al.* , whose fringe patterns where severely bent, due to inhomogeneous stresses on the two cell windows. In our new experiment, we have found zero contact angle in the whole temperature range. Two examples of profiles are shown in Fig. 8. At low temperature (271 mK) and also at 840 mK, near T_t, the interface bends upwards and meets the window tangentially. If Ueno *et al.* had been right, the interface would have bent downwards near T_t [note that, on the two figures, the vertical scale is not the same as the horizontal one, so that the window looks more tilted than in reality (20 degrees)]. A critical discussion of the profile extraction and a full presentation of results are delayed to a later publication where we shall explain more precisely why we now believe that there was an artefact in Ueno's work. For these conference proceedings, we only briefly discuss the meaning of our new measurements.

5. Discussion

The van der Waals effective force acting on a cd-interface is repulsive, proportional to the difference in the average volume per atom in each phase $V_c - V_d$:

$$\Pi_{vdW} = \frac{A_0}{L^3}\left(\frac{1}{V_d} - \frac{1}{V_c}\right) \tag{4}$$

with $A_0 \approx 1000$ K.Å^{-3}. A low temperature, V_c = 61.15 Å^3 and V_d = 46.56 Å^3 [32], so that, far below T_t= 0.87 K, the effective van der Waals force is about $5/L^3$ in K.Å^{-3} units. For partial wetting to occur in the low temperature limit, the contribution of the Goldstone modes to the Casimir force would need to be more negative than $-5/L^3$. In their review article [2], Kardar and Golestanian propose $-0.048k_BT/L^3$ which looks much too small. Following the recent work of Zandi *et al.* [21], one should also account for the existence of fluctuations at the film surface, i.e. third sound modes. Their contribution should add to that of Goldstone modes and lead to a total Casimir force which is three times more negative than previously thought, about $-0.15k_BT/L^3$. However, this looks still too small compared to the van der Waals field. In a sense, it is not surprising that we found complete wetting at low temperature, but in the first experiment done by Ueno *et al.* in Kyoto, partial wetting had been found and, since Garcia's measured value of the Casimir force is much larger than available calculations, it was worth checking that complete wetting occured at low T.

As for the vicinity of T_t, we have recalculated what should be the disjoining pressure if one assumed that, in our case, the magnitude of the Casimir force was only one fifth of what had been measured by Garcia and Chan [4]. There could be several reasons for this. Firstly, Garcia's results are indeed at least 5 times larger than any available calculation. Secondly, their measurement was done with a pure liquid ^{4}He film, while we are dealing with a mixture. Note that in a later experiment, Garcia and Chan also studied mixture films [5] but they measured the force acting on a liquid-gas interface, not on the cd-interface as we

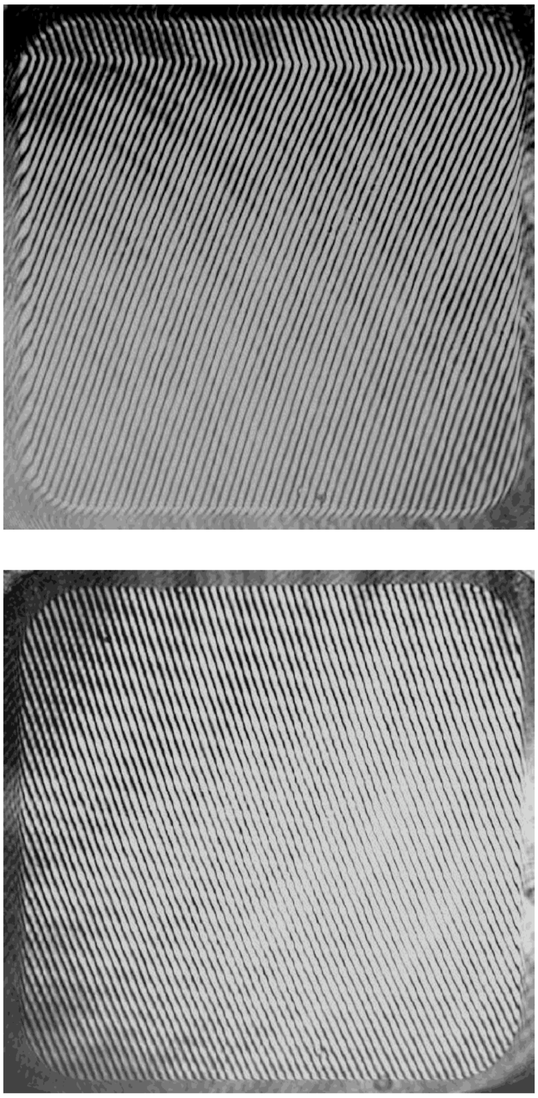

Figure 7. The profile of the cd-interface between phase separated liquid helium mixtures is calculated from the difference in optical path between a pattern with an interface (top image) and a pattern without interface (bottom image). These two fringe patterns were recorded at $T = 636$ mK.

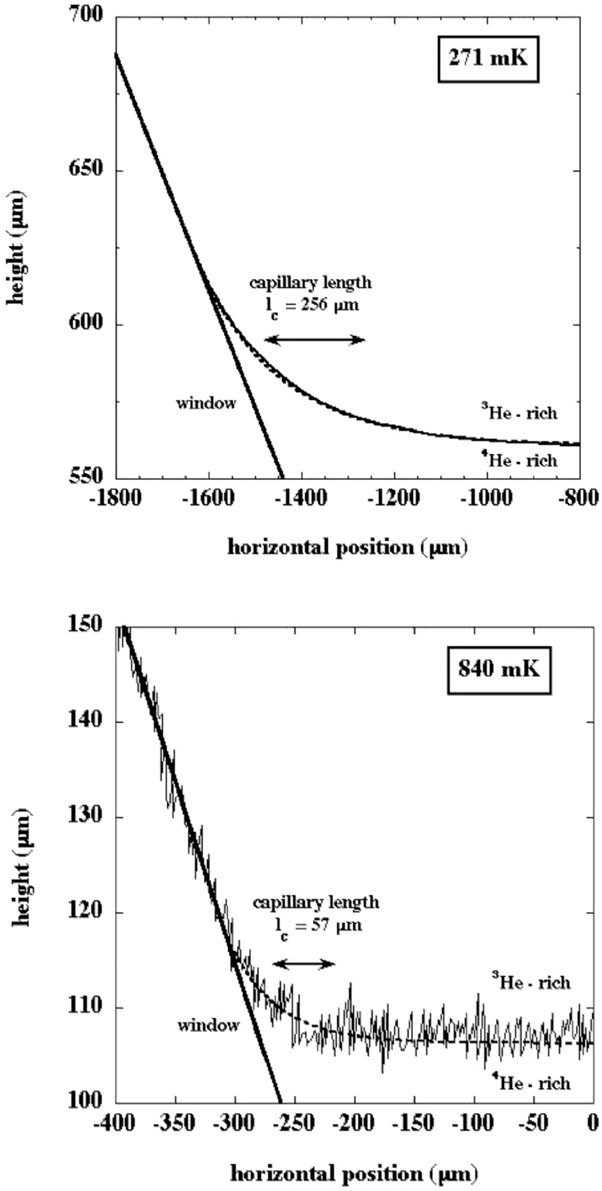

Figure 8. The profile of the *cd*-interface between phase separated liquid helium mixtures and a sapphire window at 271 mK (top) and 840 mK (bottom). The arrows indicate the magnitude of the capillary length and the broken lines are fits from Laplace's equation. The sapphire window is tilted by 20 degrees with respect to horizontal. We have found complete wetting by the ^{4}He rich d-phase: $\theta = 0$.

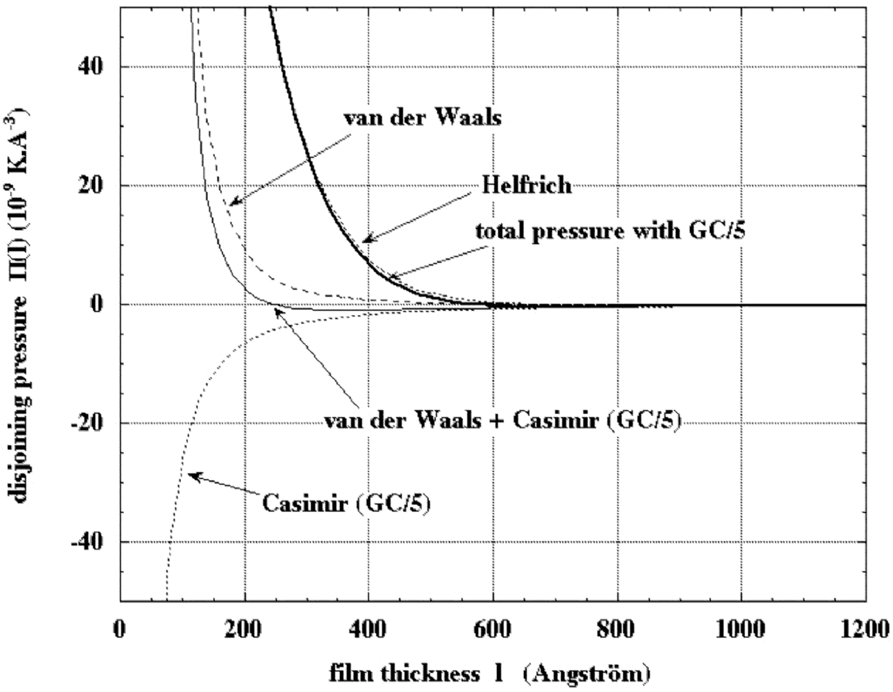

Figure 9. A modified calculation of the disjoining pressure acting on the film surface, where the amplitude of the critical Casimir force has been taken as one fifth only of the experimental result obtained by Garcia and Cahn. Contrary to what was shown in Fig. 6, one now finds that the van der Waals force always dominate the Casimir force, so that the disjoining pressure is always positive, leading to a macroscopic film thickness at equilbrium, and complete wetting.

do. In our case, a rigorous estimate should account for the existence of two competing effects. The confinement of superfluidity leads to an attractive force because the order parameter (the amplitude of the macroscopic wave function) vanishes symmetrically on both boundaries of the film. However, there are also fluctuations of concentration near T_t. Due to the van der Waals field, the film is more diluted in ^{3}He near the solid wall than near the cd-interface, so that , for concentration fluctuations, the boundary conditions are anti-symmetric. This is why, for an ordinary liquid mixture with no superfluidity, the Casimir force would add to the van der Waals field to favor wetting by the d-phase. Of course, superfluidity and concentration fluctuations are coupled near T_t, and the effect of superfluidity should dominate because the dimension of its order parameter is $N = 2$ instead of $N = 1$ for concentration. Still, we expect a rigorous calculation to find that the effect of concentration fluctuations decreases the effect of superfluidity fluctuations.

As shown in Fig. 9, when estimating the amplitude of the Casimir force as only one fifth of what was measured by Garcia and Chan, we find that complete wetting is restored. Compared to the results shown in Fig. 6, the disjoining pressure is now positive for all thicknesses. Given our most recent measurements [18], we now believe that, in our experiment, that is near a sapphire window, the critical Casimir force is too weak compared to the van der Waals field, so that complete wetting occurs, as usual. To find an exception to critical point wetting, one should try using a susbstrate exerting a much weaker van der Waals field than ordinary insulating materials, and this does not look easy to us.

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