Absence of critical point wetting near the ^{3}He-^{4}He tri-critical point

T. Ueno [a,2], S. Balibar [a,1], T. Mizusaki [b] F. Caupin [a], M. Fechner [a] and E. Rolley [a]

[a] *Laboratoire de Physique Statistique de l'ENS, associé au CNRS et aux Universités Paris 6 et 7, 24 rue Lhomond,75231 Paris Cedex 05, France*

[b] *Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan*

Abstract

We have measured the contact angle of the ^{3}He-^{4}He interface on a sapphire window near the tri-critical temperature $T_t = 0.87$ K. We have found that it is finite and that it increases with temperature above 0.81 K. This behavior is opposite to the "critical point wetting" which is usually observed near critical points. Our results confirm earlier MRI measurements by Ueno et al[1]. We interpreat our observations as due to the Casimir effect which leads to an effective attraction of the ^{3}He-^{4}He interface by the substrate near the superfluid transition at T_t.

Key words: wetting, helium mixtures, critical phenomena, Casimir effect.

When a solid substrate is in contact with a phase-separated mixture of ^{3}He and ^{4}He, perfect wetting by the ^{4}He rich "d-phase" is usually expected, due to the van der Waals attraction by the substrate. However, we measured the contact angle of the ^{3}He-^{4}He interface on sapphire, and found that it is finite above 0.81 K, close to the tri-critical point $T_t = 0.87$ K. In the case of ordinary critical points, a wetting transition usually occurs slightly below the critical temperature. On the contrary, here, we have observed that the contact angle increases as T approaches T_t. These results confirm previous MRI measurements[1]. We interpreat our surprising observations as due to the Casimir effect[2].

We use a dilution refrigerator with optical access to cool down liquid ^{3}He-^{4}He mixtures below T_t. Our cell is made of copper with sapphire windows (Fig. 1). Interferometric images are obtained with a He-Ne laser beam and a CCD camera outside the cryostat. The inner surfaces of the windows have a 15% reflectivity, so that well contrasted sinusoidal fringes are obtained. The sample space is $11\times11\times10$ mm^3 in size. Near each window, the liquid interface forms a meniscus whose shape depends on both the interfacial tension σ_i and the contact angle θ. In order to separate the back-meniscus from the front one, the cell is tilted by 8.8°. We checked the mixture concentration by measuring its phase separation temperature. This phase separation was completed when the c-d interface stopped moving. This took several hours very close to T_t. Temperatures are regulated within 0.2 mK.

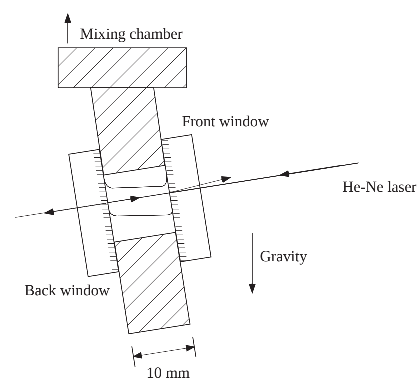

Fig. 1. The copper cell is closed by two sapphire windows which form an interferometric cavity. From top to bottom, one sees the gas phase, the ^{3}He rich "c-phase", and the "d-phase".

[1] Corresponding author. E-mail: balibar@lps.ens.fr

[2] Present address: Research Laboratory of Electronics and MIT-Harvard Center for Ultracold Atoms, MIT, Cambridge, MA 02139, USA

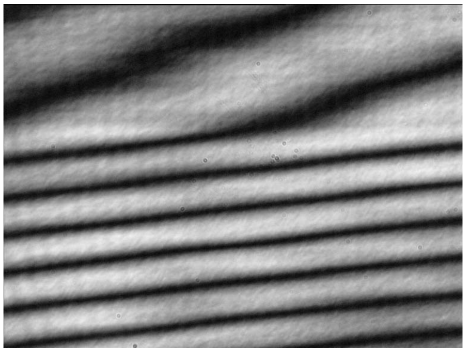

Fig. 2. Fringe pattern of the c-d interface at 0.856 K.

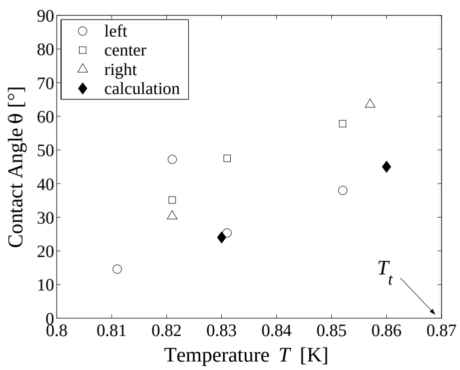

Fig. 3. Temperature dependence of the contact angle θ. The labels "left", "center" and "right" refer to three different positions along the contact line.

Fig. 2 shows a magnified image of a typical fringe pattern. The upper part corresponds to the ^{3}He rich "c-phase" and the lower one to the c-d interface. The interesting region is near the contact line, where fringes bend. From such patterns, we extract the profile of the c-d meniscus. From a fit with a solution of Laplace equation, we then obtain σ_i and θ. As T approaches T_t, the capillary length vanishes so that the region to be analyzed becomes very small. This is one limitation of our method. Moreover, refraction effects modify the pattern as soon as the index difference n_c - n_d is not small, so that the analysis is also difficult far below T_t. This is why we present results only from 0.81 to 0.86 K.

At each temperature, we analyzed three pictures at different positions along the contact line (see Fig. 3). Our measurements of σ_i agree well with previous results by Leiderer et al.[3]. As for the contact angle θ, we found that it was finite. Furthermore, it increases with temperature, instead of decreasing as the usual theory of critical point wetting would predict. Our present results are consistent with previous MRI measurements[1] ($\theta = 35 \pm 10\,^\circ$ on stycast at 0.8 K). Various experimental difficulties, such as the refraction effects, a slight bending of the stressed windows, and the small value of the capillary length near T_t, explain the rather large scatter of data points on Fig. 3.

Let us now briefly present a tentative interpretation for such results. Off-coexistence, a d-phase film exists between substrates and the c-phase. This is due to the van der Waals attraction which favors the d-phase where the volume per atom is smaller. One used to believe that, as coexistence is approached, the thickness of this film diverges to infinity. This would be true if van der Waals forces were the only ones acting on the mixture and it would imply perfect wetting by the d-phase. However, there is also a Casimir force which, being attractive on the film surface, prevents its thickness to diverge. As shown by Garcia and Chan[2], superfluid films of pure ^{4}He get thinner near T_λ. Our situation is similar: our d-phase film is just below its superfluid transition. At 0.86 K, we have calculated the Casimir attraction with Garcia's scaling function and we have found that it can be larger than the van der Waals force. Indeed, the latter is a small effective repulsion on the film surface, which vanishes linearly with (T - T_t). In order to calculate the "disjoining pressure"[4] Π(l) as a function of the film thickness l, we neglected short range forces and added a third long range force, the entropic repulsion by the wall which tends to increase the film thickness. The equilibrium film thickness l_f is given by the point where Π(l) crosses zero with a negative slope [5]. We found l_f = 400 Å. Following Ross et al.[4], we then integrated Π(l) from l_f to infinity, and obtained the interfacial tension of the sapphire to c-phase interface and finally a contact angle $\theta = 45\,^\circ$ at 0.86 K. At 0.83 K, we estimated $\theta\ \approx 24\,^\circ$, but we would need a better knowledge of the scaling function. These values agree with our measurements. Of course the Casimir effect vanishes away from T_t. We thus understand why the contact angle of the c-d interface is finite slightly below T_t and why it decreases as the temperature decreases away from T_t. This is a remarkable exception to the usual "critical point wetting". Very close to T_t, short range repulsive forces should become dominant because the correlation length becomes very large, and θ should tend to zero again. The wole effect obviously needs further measurements and calculations.

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