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# Apparent dewetting due to superfluid flow

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**Abstract.** – We have investigated the wetting behaviour of superfluid helium-4 on silicon. Surprisingly, we observe pseudo-dewetting: though a thick superfluid film covers the substrate, the meniscus diplays a finite contact angle which decreases from about 5° at low temperature down to zero at the superfluid transition. We show that this behaviour can be explained by a pressure decrease due to a superfluid flow, closely related to the Kontorovich effect.

Wetting properties of superfluid helium-4 have been under intense investigation since the theoretical prediction by Cheng *et al.* [1] that cesium is not wetted by helium-4 at low temperature. After the first experimental check by Nacher and Dupont-Roc [2], the contact angle at low temperature was found to range between 25° [3,4] and 48° [5]. More surprisingly, Alles *et al.* have measured recently a small but finite contact angle during the spreading of $^4$He on evaporated $SiO_2$ [6]. Herminghaus suggested that this effect could be due to the Bernouilli pressure [7]. This mechanism does not account for the contact-angle hysteresis observed by Alles *et al.*, and superfluid vortices are more likely to be responsible for this unconventional non-wetting behaviour, as proposed by Luusalo *et al.* [8].

In this letter, we report on apparent dewetting of superfluid helium-4. This is a quite unexpected situation where bulk liquid drops with a finite contact angle do coexist with a thick liquid film. This pseudo-dewetting situation is qualitatively different from the one observed by Alles. First, the value of the pseudo-contact angle $\theta$ is of the order of 5° at $T = 1.2\,\mathrm{K}$, while Alles reports values below 1°. Second, we do not observe any hysteresis. We think that this pseudo-dewetting is due to a superfluid flow, as proposed by Herminghaus. This flow is likely to be driven by residual temperature gradients in the experimental cell. We show that the experimental data are consistent with a pseudo-dewetting due to a kinetic effect, though a quantitative analysis leads to a critical superfluid flow larger than usually reported.

Our experiments are performed in an optical helium-4 cryostat equipped with an interferometer, so that the meniscus profile can be obtained from the interference pattern. The substrate is slightly tilted with respect to the horizon by an angle $\alpha$, which can be varied between 1 and 10°. Examples of interferometric images and profiles of the meniscus are shown

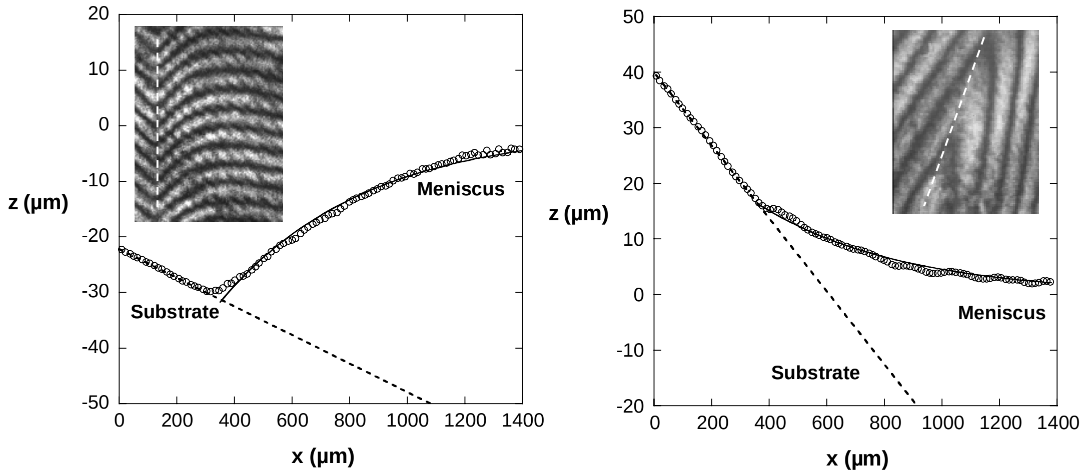


Fig. 1 – Interferometric images and corresponding profiles of the meniscus (actual size: 3.2 mm × 2.4 mm). Left: $T = 1.15\,\mathrm{K}$, $\alpha = 1.5°$, $\theta = 5°$, M2 substrate. Right: $T = 2.0\,\mathrm{K}$, $\alpha = 4.5°$, $\theta = 1.8°$, M1 substrate. The dashed line in the images is the position of the pseudo-contact line. The solid line is obtained by fitting the experimental profile with the exponential solution of eq. (1).

in fig. 1. Away from the pseudo-contact line, the height $z(x)$ is very close to the equilibrium profile (solid line in fig. 1) which is controlled by the balance between gravity and surface stiffness:

$$0 = -\rho g z + \gamma \partial^2 z / \partial x^2 \,; \tag{1}$$

$\rho$ is the liquid density, $g$ is the acceleration of gravity and $\gamma$ is the liquid-vapor surface tension (the zero-temperature value of $\gamma$ is 0.375 mN/m [9]). The capillary length is $L_\mathrm{C} \equiv (\gamma/\rho g)^{1/2} \approx 500\,\mu\mathrm{m}$. Note that the slope of the interface is small, so that the curvature can be approximated by $\partial^2 z/\partial x^2$. The pseudo-contact angle $\theta$ is defined by extrapolating the equilibrium profile down to the substrate. Close to the substrate, the meniscus matches smoothly a superfluid film. The rounding of the meniscus at the pseudo-contact line changes from one image to the other, because of residual vibrations and dust particles on the mirror. This blurs the edge of the meniscus, and prevents us from analyzing the shape of the matching region. Though the film is too thin to be measured, its presence is demonstrated by the fast formation of large liquid drops on dust particles. On a non-wetted substrate, such drops condense from the vapor phase, and their dynamics is much slower.

Two different runs have been performed. In the first one, the Si mirror (M1) was coated by 40 atomic layers of cesium evaporated at 20 K. The Cs coating was presumably very rough since it did not display any true dewetting. In the second one, we used a bare Si mirror (M2). We have not measured any significant difference in the value of the pseudo-contact angle. We have also checked that the value of $\theta$ is independent of the tilt $\alpha$ of the substrate. We could not detect any hysteresis. $\theta$ has been measured as a function of the temperature $T$ (fig. 2). We find $\theta = 5.5°$ at $T = 1.15\,\mathrm{K}$; $\theta$ decreases with $T$ and vanishes close to the superfluid transition ($T_\lambda = 2.17\,\mathrm{K}$).

Such a behaviour has never been reported previously. We think that it is observed in our experiment because of the thermal decoupling of the substrate: since earlier experiments (see ref. [3]), the cell has been modified so that the bottom of the cell, where the substrate lies, is now thermally decoupled from the cell walls. Thus the substrate is warmer than the

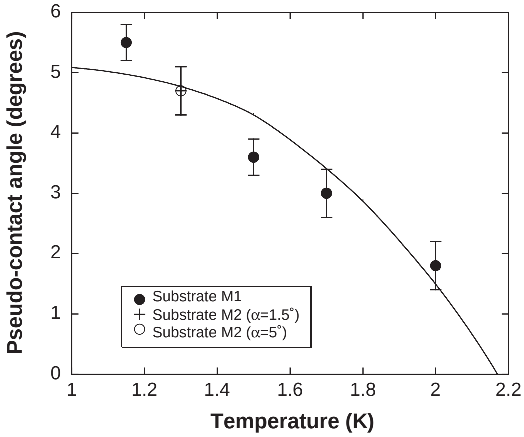


Fig. 2 – Temperature dependence of the pseudo-contact angle $\theta$ for various experiments. The solid curve is the pseudo-contact angle induced by the Bernouilli pressure, assuming that the flow current is equal to $2 \times 10^{-8}$ $m^2s^{-1}$ at low temperature and varies like the superfluid fraction.

surrounding walls of the cell. The heat load to the substrate may have two different origins. In a preliminary report [10], we had suspected 300 K black-body radiation and illumination light to be responsible for heat absorption by silicon. However, we have added an IR filter without changing the situation. We have observed that the residual gas in the vacuum can is responsible for a heat leak between the cell and the 4 K thermal shield. The total heat input on the substrate is difficult to estimate, and we can give only a lower bound of 150 $\mu$W. At first sight, we thought that pseudo-dewetting was *directly* associated to some thermal gradient in the substrate, since the substrate below the bulk superfluid is presumably colder than the part of the substrate which is only covered by the film. It turns out that pseudo-dewetting can also be observed when a small diameter laser beam causes localized heating of the substrate (fig. 3). For a large enough heat input $\dot{q}$, isolated droplets are stable. In this

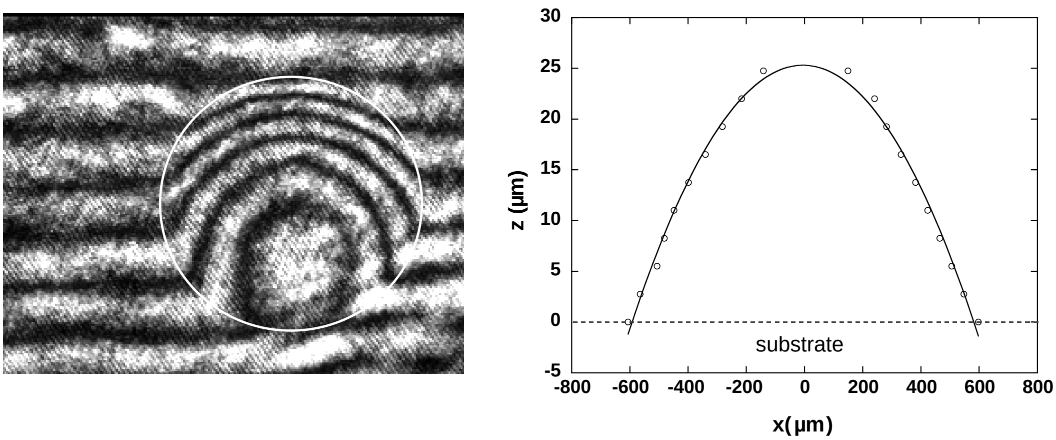


Fig. 3 – Image and profile of a sessile drop, which is stabilized with a localized heat source. The contact angle is of the order of 4.5°. Actual size of the image: 3.2 mm × 2.4 mm.

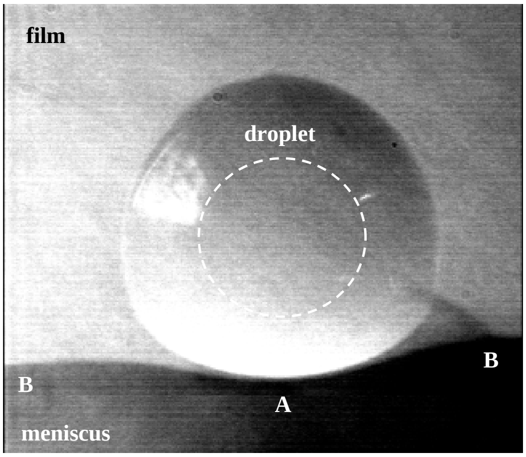


Fig. 4 – A sessile drop repels the meniscus! When a laser beam hits the substrate close to the meniscus, it creates a droplet and also causes a local increase in the flow outwards the meniscus. As a consequence, the contact angle in A is larger than in B. The dashed circle corresponds to the position of the laser spot. Actual size of the image: 3.2 mm × 2.8 mm.

case, the substrate covered by the droplet is warmer than the substrate covered by the film, and the temperature gradient is opposite to the one existing in fig. 1. However, one finds roughly the same pseudo-contact angle.

We propose the following interpretation. Because of the illumination or heat leaks, the heating of the substrate is not homogeneous. This creates a superfluid flow towards the heat sources, where the superfluid component is converted into normal fluid [11]. As long as the heat input is small enough, the flow is non-dissipative, and there is no temperature gradient. As suggested by Herminghaus, the superfluid flow decreases the pressure in the liquid and leads to pseudo-dewetting. As the Bernouilli pressure is quadratic in velocity, this scenario is consistent with a dewetting which does not depend on the direction of the flow. It is also consistent with the surprising observation that a sessile droplet repels the meniscus (fig. 4). When a laser beam hits the substrate close to the meniscus, it creates a droplet and also causes a local increase in the flow outwards the meniscus. This leads to a local increase of $\theta$. Such a situation shows clearly that the slope of the interface is not sensitive to the direction of the heat current. For an isolated droplet, the incoming flow can be easily estimated. The typical heat input due to the laser beam is of the order of $\dot{q} = 80\,\mu$W for a droplet of radius $r = 0.6$ mm. Assuming that the heat is carried away by evaporation at the surface of the droplet, the flow divided by the perimeter is found to be: $j \simeq 7 \times 10^{-9}$ m$^2$/s. This value is of the order of critical currents in films [11], so that finite-temperature gradients are likely to exist at the substrate.

Let us show that the change in pressure due to superfluid flow can explain the measured value of $\theta(T)$. In the two-fluid model [11], only the superfluid component can flow in a film, so that the change in pressure due to Bernouilli's law is $\Delta p = -\rho_S v_S^2/2$, where $\rho_S$ and $v_S$are, respectively, the density and the velocity of the superfluid component. For a film of thickness $h$, the experimental relevant quantity is the total flow current $j = (\rho_S/\rho)v_S h$, so that $\Delta p = -\frac{\rho^2}{2\rho_S} j^2/h^2$. The effective disjoining pressure $\Pi$ for a film of thickness $h$ is the sum of the usual van der Waals term and $\Delta p$. This leads to a thinning of superfluid films, known as the Kontorovich effect [12]. In this paper, we are interested in films whose thickness $h$ varies

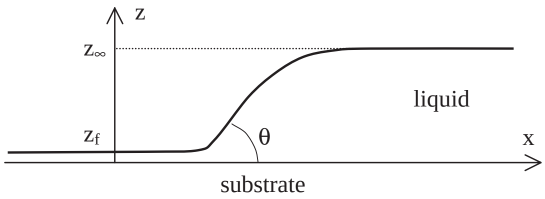


Fig. 5 – Schematic profile of the meniscus. For $z \gg 1\mu$m, both van der Waals interaction and Bernouilli pressure are negligible so that the profile is the same as the equilibrium one.

spatially. The experimental situation (see fig. 1) is rather complicated since the substrate is not horizontal. Fortunately, $\theta$ is found to be independent of the tilt angle $\alpha$ (this will be discussed later), so we shall assume $\alpha = 0$ in the following analysis. Thus, the local liquid thickness $h$ is equal to the height $z$ (fig. 5).

We can also safely assume that, for large $x$, the thickness $z_\infty$ is large enough to neglect van der Waals and Bernouilli effects, so that bulk liquid-vapor equilibrium is achieved. The equilibrium condition for the interface then reads

$$-\rho g z_\infty = -\rho g z + \gamma \partial^2 z/\partial x^2 + \frac{C}{z^3(z/d)} - \frac{\rho^2}{\rho_{\mathrm{S}}}\frac{j^2}{2z^2}\,. \tag{2}$$

The left-hand side is not zero because we have chosen $z = 0$ at the substrate. In the right-hand side, the first two terms are the same as in eq. (1). The disjoining pressure is the sum of the last two terms, respectively, the van der Waals potential and the Bernouilli effect. This equation is basically the same as the one derived by Herminghaus (eq. (2) in [7]). We have simply added the gravitational term, and we have used the retarded van der Waals interaction ($z^{-4}$ instead of $z^{-3}$ behaviour) [13]. Indeed, the crossover between those two behaviours occurs for a film thickness of the order of 10 nm, which is smaller than the typical values of $z_{\mathrm{f}}$ that we obtain. The values of the parameters $C$ and $d$ for helium on silicon can be estimated from the work of Sabisky and Anderson [14]; one finds $C = 4.9 \times 10^{-22}$ J and $d = 14$ nm. The only adjustable parameter is the flow current $j$, whose value is expected to be of the order of $10^{-8}$ m$^2$/s. Let us stress that eq. (2) is valid only if the temperature is homogeneous. Strictly speaking, this may be false: we have shown previously that the value of the superfluid current is of the order of critical currents in films. However, we have shown that the temperature gradient itself does not seem to change the value of the contact angle. It may happen that temperature gradients are localized in the region where the film is thin, and not in the vicinity of the pseudo-contact line, where $\theta$ is measured. This is a reasonable assumption, since it is known that the value of the critical current decreases when the film thickness gets smaller than a few hundred ångströms [11]. In the following, we will also make the assumption that $j$ is constant: we neglect mass transport through the vapor phase. Once more, this is certainly false at the scale of the whole cell, but this is reasonable in the vicinity of the contact line.

The disjoining pressure is negligible compared to $\rho g z$ as soon as $z$ is larger than the crossover length $H$ defined by $\rho g H = \frac{\rho^2}{2\rho_{\mathrm{S}}}\frac{j^2}{H^2}$. With $j = 10^{-8}$m$^2$/s and $\rho_{\mathrm{S}} = \rho$, one finds $H = 1.7\,\mu$m. So one expects the profile $z(x)$ to be nearly identical to the equilibrium one, as soon as $z$ is larger than a few micrometers. This is in agreement with the experiment (see fig. 1). This also means that it is not useful to integrate exactly eq. (2), since the accuracy of the profile measurement is not good enough to allow a comparison with the theoretical profile in the crossover regime $z \sim H$ . Solving eq. (2) in the limit $z \ll H$ is sufficient to compute $\theta$. First, let us rescale $x$ by $x_0 \equiv (\gamma\rho z_0^5)^{1/2}/Cd$ and $z$ by $z_0 \equiv (2Cd\rho_{\mathrm{S}})^{1/2}/\rho j$ ($z_0$ is the thickness of a homogeneous film at coexistence). One is left with

$$-\delta = z'' + \frac{1}{z^4} - \frac{1}{z^2}\,, \tag{3}$$

where $\delta = \left(z_\infty x_0^2\right)/\left(z_0 L_{\mathrm{C}}^2\right)$. Experimentally, $z_\infty$ is at most 50 $\mu$m (see fig. 1a); this gives $\delta \leq 6 \times 10^{-4}$. In first approximation, one can set $\delta = 0$, and one is left with an equation which is easily integrated. At large $z$ ($z \gg z_0$), one finds that $z$ varies linearly with $x$, so that the profile displays a pseudo-cusp very similar to the one found by Herminghaus in the non-retarded case. In this approximation, the minimum film thickness $z_{\mathrm{f}}$ is equal to $z_0$, and is found to be of the order of 15 nm. The pseudo-contact angle $\theta$ is given by

$$\tan\theta = \frac{2^{1/4}}{3^{1/2}} \frac{[(\rho^2/\rho_S)j^2]^{3/4}}{(Cd)^{1/4}\gamma^{1/2}}\,. \tag{4}$$

The change $\delta\theta$ in the contact angle due to the shift from coexistence $\delta$ can then be estimated. A calculation analogous to Herminghaus' leads to $\delta\theta/\theta = -\sqrt{3\delta}$. With $z_\infty \leq 50$ $\mu$m, the relative change in the contact angle is at most 4% smaller than the uncertainty.

Let us now compare these predictions with experimental data. First, one finds that $\theta$ is weakly sensitive to the parameters $C$ and $d$ characterizing the van der Waals interaction. This is consistent with the fact that we measure the same contact angle on bare silicon and silicon coated with cesium. We find also that the change in $\theta$ due to the shift $\delta$ from coexistence is small. This shift is related to the asymptotic height $z_\infty$ of the bulk liquid with respect to the pseudo-contact line. In our experimental setup, this height depends on the tilt $\alpha$ of the substrate: $z_\infty$ varies from $z_\infty \simeq \theta L_{\mathrm{C}}$ for $\alpha = 0$ (as in fig. 5) down to $z_\infty \simeq 0$ for $\alpha = \theta$. Experimentally, we find that $\theta$ is not dependent on $\alpha$; this is consistent with the model which leads to a very small variation of $\theta$ in the experimental range for $z_\infty$. At low temperature, the normal component of the superfluid is almost negligible and $\rho \simeq \rho_{\mathrm{S}}$. Then eq. (4) provides a simple relation between $\theta$ and $j$. Using the experimental value $\theta = 5.5°$, one finds $j \simeq 2 \times 10^{-8}\mathrm{m}^2/\mathrm{s}$ [15]. This is larger than the flow estimated from the heat balance in the droplet case. This is not very surprising: the droplet case is certainly more complex. Direct temperature effects could play a non-negligible role, as the size of the heat source (*i.e.* the laser spot) is of the same order as the droplet diameter. What is more surprising is that we find a value for $j$ which is about 3 times larger than most of the values of critical flow reported in the literature [11].

At this point, we have to emphasize that the measured value of the contact angle is not sensitive to the heat input to the substrate: changes in the illumination intensity or in the residual pressure of the vacuum cannot lead to measurable changes in $\theta$. This is a strong indication that the superfluid flow in the film is critical at some distance from the meniscus (otherwise, one expects the superfluid flow to be proportional to the heat input [11]). In many experiments, the critical flow is found to vary with temperature roughly like the superfluid fraction $\rho_{\mathrm{S}}/\rho$ [11]. If we assume that, in our experiment, the flow reaches a critical value, and that this critical value varies like the superfluid fraction, we obtain from eq. (4) that the pseudo-contact angle $\theta$ varies roughly like $(\rho_{\mathrm{S}}/\rho)^{3/2}$. The corresponding variation of $\theta$ is plotted in fig. 2. Our model and hypothesis account for the vanishing of $\theta$ at $T_\lambda$. A precise comparison is difficult since our data are restricted to a limited range of temperature. It would be interesting to check whether $\theta$ saturates below 1 K, as expected from the temperature dependence of the superfluid fraction.

Many features of the experimental data are satisfactorily explained by the simple model originally proposed by Herminghaus and the assumption that the superfluid flow reaches a critical value away from the pseudo-contact line. This assumption is consistent with the fact that we have neglected the temperature gradient in the vicinity of the contact line. However, a quantitative agreement requires that, in our experiment, the critical flow is larger than usually reported for thin films. A precise test of the model would require a better knowledge of the superfluid current $j$. This could be achieved by controlling the heat current and using a subcritical flow, for which $\theta$ is expect to vary like $j^{3/2}$.

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[15] Herminghaus [7] finds about the same value for $\theta$ ($\theta = 4.6°$) for a superfluid flow $j = 10^{-8}$ m$^2$/s which is noticebly smaller than our value. This is not due to the difference between retarded and non-retarded van der Waals potentials, but mainly to some inconsistency in the evaluation of $\theta$. Herminghaus uses a typical experimental value of the critical velocity $v_C = 60$ cm/s as an additional parameter to determine the film thickness $h_0$ (denoted $z_0$ in this letter). Using $h_0 = j/v_C$, he finds $h_0 = 17\,$nm, which is not consistent with the definition of $h_0$: $h_0 \equiv A/3\pi\rho j^2$. This relation leads to $h_0 \simeq 80$ nm with $A/6\pi = 5.7 \times 10^{-22}$J, so that $\theta = 1°$ (note that $A$ should be replaced by $-A/6\pi$ on p. 445 of ref. [7]).