Prewetting of ^{4}He on Rb: The Coexistence of Two Superfluid Films?

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We have found the prewetting line and estimated the wetting temperature for ^{4}He on Rb. $T_w \approx 309$ mK is much lower than that found for Cs. This low value of T_w indicates that Rb is likely to be the only other element besides Cs that is not wetted by ^{4}He. The prewetting line is not consistent with theory nor with previous data. We argue that the *thin* state of ^{4}He on Rb appears to be superfluid for $\Delta\mu$ near coexistence; if this is so then on the prewetting line there are two superfluid films of disparate thicknesses.

It has now been clearly established that ^{4}He does not wet a Cs surface below a temperature of 2 K [1–4]. However, no other elements to date have shown nonwetting behavior, although composite substrates with a Cs film as the outer layer do so [5]. The next most promising element is Rb which is predicted to be a borderline case between wetting and nonwetting, i.e., having a wetting temperature T_w around 0 K [6]. The prediction has an uncertainty due to the difficulty of calculating the depths of the ^{4}He-alkali metal potential wells. The experimentally determined wetting temperature of Cs at 2 K is half that originally predicted. Experimental studies [7,8] of ^{4}He on Rb have only been down to $T = 0.8$ K, and they have indicated a prewetting transition but it has been concluded that Rb is wetted by ^{4}He at all temperatures at least for the samples of Rb reported so far [7]. This is an important point as the wetting behavior of surfaces is extremely sensitive to their chemical and physical state, and the high reactivity of the alkali metals makes this a critical problem.

The system of ^{4}He on Cs and H$_2$ on Rb has enabled prewetting to be seen and studied [9,10]. Prewetting is a first order phase change in two dimensions (2D) between a *thin* and *thick* film state. This phenomenon has been predicted in 1977 [11], but no examples were found until recently when it has been seen with the quantum systems of H$_2$ and ^{4}He and for the binary liquid methanol-cyclohexane mixture [12]. For classical systems the effect occurs too near the vapor-liquid coexistence line to be easily distinguished, but alkali substrates are predicted to show prewetting more readily [13].

There is no question that the state of the *thick* film is other than a liquid which is similar to bulk liquid at a distance a few layers away from the substrate. Indeed the *thick* film becomes infinitely thick at T_w. However, much less is known of the *thin* film state. The *thin* film state does not just occur on the prewetting line, but occupies the space in the $\Delta\mu$-T plane on the low temperature side of the prewetting line (see, for example, Fig. 4). The *thin* film seems to be tacitly considered to be a 2D gas which it certainly is at low temperatures, $T \ll T_w$. Consistent with this idea, it has been proposed that the prewetting transition is a 2D-gas–2D-liquid phase change

[4]. With a 2D quantum gas such as ^{4}He the question arises whether the 2D gas can be superfluid. However, it has been concluded [6] that the temperature-dependent density is too low, so that any potential Kosterlitz-Thouless transition is always at a temperature lower than that of the 2D gas. In particular, for ^{4}He on Cs, it is considered [3,5,6] that the prewetting line separates a normal fluid *thin* film state from either a superfluid *thick* state for $0 < \Delta\mu \le -3$ mK or a normal fluid *thick* state for $-3 < \Delta\mu \le -52$ mK.

In this Letter we report a wetting transition for ^{4}He on Rb at $T_w \approx 309$ mK together with the prewetting line close to coexistence. The gradient of this line is quite different from the theoretical prediction. We find that the *thin* film state appears to be a superfluid and so, on the prewetting line, it is possible that there is coexistence of two superfluid films of widely disparate thicknesses.

To investigate wetting and superfluidity in *thin* films at $T < 1$ K we have developed a technique which measures small flows of adsorbed ^{4}He on alkali metals. Consider a closed cell with a vertical post rising from the cell base as shown in the inset in Fig. 3. Around this post is a continuous band of Rb metal. Above this band on the post there is a thin film Cr heater, the wires to which run up the inside of the post and are sealed into it. Opposite the heater is a thin film Zn bolometer which is supported directly from the cell base. When the heater is pulse heated some of the ^{4}He atoms on it are evaporated and these atoms are detected by the bolometer as they give up binding energy and kinetic energy, which is typically 10 K per atom. The heater pulse energy is $\approx 1.5 \times 10^{-10}$ J, which can evaporate ≈ 0.1 layer of ^{4}He from the heater film.

The Rb surface is prepared in two stages. First Rb of nominal purity, 99.9%, is vacuum distilled and then transferred to a glass toroid and sealed off. The Rb fills only a small part of the volume of the toroid. Just before the experiment is performed, the Rb within the glass toroid is redistilled so that a fresh surface on bulk Rb is created at the top of the toroid. The experiment is then cooled to ≈ 1 K and the outer wall of the glass toroid broken away exposing the Rb to the pure ^{4}He gas [14].

The cell is then further cooled with a dilution refrigerator to a base temperature of ≈ 50 mK. Temperatures are measured with a calibrated Ge thermometer which is in the cell. The heater and bolometer are used, respectively, to evaporate ^{4}He atoms from the post and to detect them in the cell. The number of evaporated atoms can be calculated from the bolometer response, its sensitivity, and the geometry of the heater-bolometer configuration. The mass flow of ^{4}He over the Rb is then the number of atoms evaporated per pulse (A) multiplied by the pulse repetition frequency (f).

In a separate experiment we have investigated the behavior of normal and superfluid films of ^{4}He on a brass post. With this arrangement we can gauge the film thickness using the Kosterlitz-Thouless relation between the film thickness and the transition temperature T_{KT} [4] [see inset in Fig. 1(a)] and then measure the flow rates at a constant temperature above and below T_{KT} with the same ^{4}He film thickness. The amplitude of the bolometer signal is measured as a function of the pulse repetition frequency. The behavior is qualitatively different for normal (n) and superfluid (s) ^{4}He films as can be seen in Fig. 1 where we plot the flow (Af) as a function of frequency.

To understand these contrasting behaviors we must consider the processes by which ^{4}He gets onto the heater. When the film is superfluid there is ample time between heater pulses for the film on the heater to be restored to its dynamic equilibrium thickness, which is only a little less than the static equilibrium thickness; it is slightly less in order to create the gradient in chemical potential which drives the ^{4}He up the post. As the frequency increases so does the flow rate, until eventually there is significant dissipation. At yet higher frequencies the dynamic film thickness on the heater and its surroundings is reduced, and this causes the amount of ^{4}He evaporated in each pulse to drop as we see in Fig. 1(b). In contrast, when the film is normal there is always a resistance to the flow onto the heater between heater pulses. At low enough frequencies (<1 Hz), there is enough time between pulses for the film on the heater to equilibrate with that on the surroundings and so the amount evaporated per pulse is independent of frequency. This means at low frequencies, the flow increases linearly with frequency. However, at higher frequencies the amount of ^{4}He that can flow onto the heater, and so be evaporated, is proportional to the time available between pulses, i.e., $1/f$. The amount is also proportional to the thickness of the ^{4}He film in the vicinity of the heater, but this is expected to remain fairly constant. As $A \propto 1/f$, the flow (Af) remains constant as the frequency is increased as is shown in Fig. 1(a). The difference between n and s behavior can be seen with different film thicknesses.

The situation is similar but with an added factor when there is an exposed Rb band around the post. The amount of ^{4}He that is evaporated by the heater is determined again by the local conditions of the ^{4}He film near the heater but
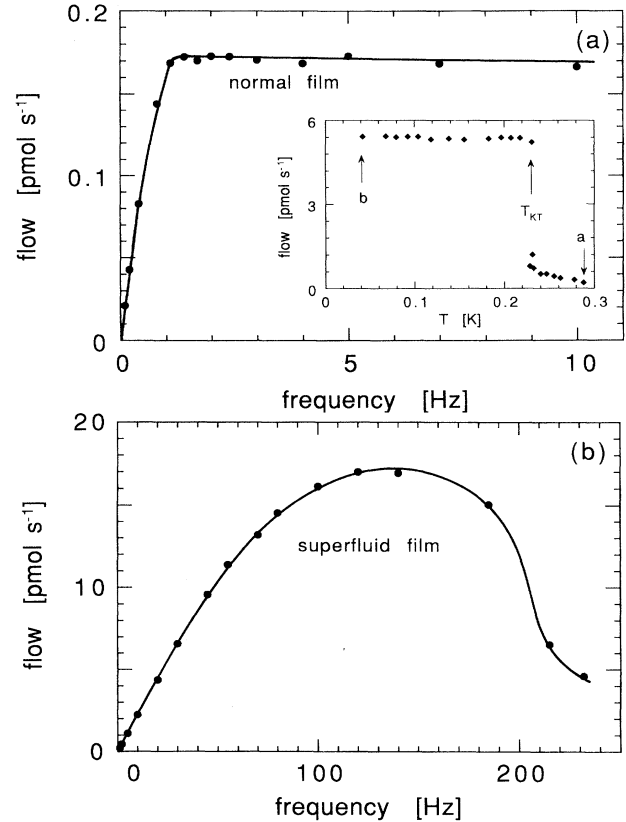

FIG. 1. Inset in (a) shows the ^{4}He flow on a dummy post below and above the Kosterlitz-Thouless transition $T_{KT} = 230$ mK with driving frequency $f = 25$ Hz. Arrows a and b indicate temperatures where the flow vs driving frequency is measured in the normal and superfluid state. (a) Flow of ^{4}He in a normal $T > T_{KT}$ shows an increase with frequency at low frequencies and is constant at higher frequencies. This behavior continues until the frequency is so high that the amplitude is too small to measure. (b) Flow of ^{4}He in a superfluid state $T < T_{KT}$ shows an initial increase with frequency, a maximum, and then a decrease of flow for higher driving frequencies due to dissipative processes. Note the difference in flow maxima in the two cases.

these conditions are controlled by the ^{4}He flow over the Rb band. To see this, consider that the ^{4}He is superfluid on both the Rb and the post and is flowing towards the heater to replace the ^{4}He that has evaporated there. The superfluid film on the Rb is thinner than the film on the post as Rb is a weaker binding substrate than glass. So when the flow rate is increased, dissipation will occur first in the film on the Rb. If the flow on the Rb becomes dissipative, then the film on the post becomes thinner until a new dynamic equilibrium is attained where the lowered chemical potential on the post drives the ^{4}He across the Rb. However, the thinner film around the heater causes less ^{4}He to be evaporated and so the new equilibrium has a lower flow rate than before. In this way we can determine what is happening on the Rb from the evaporation rate [15].

Two typical flow rates as a function of frequency are shown in Fig. 2 for $T \lesssim T_{pw} = 312$ mK. The behaviors
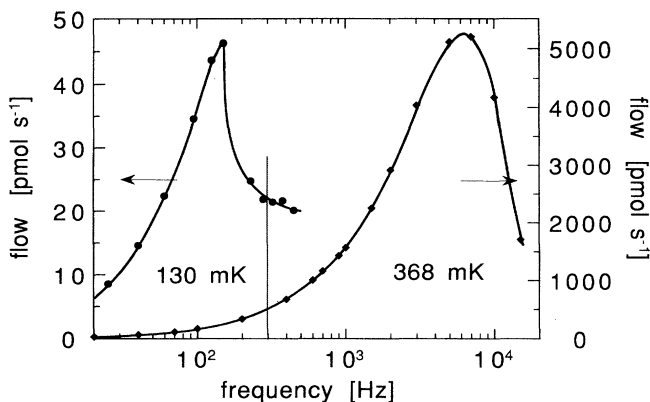
FIG. 2. ⁴He flow rate on Rb below ($T = 130$ mK) and above ($T = 368$ mK) the prewetting temperature ($T_{\mathrm{pw}} = 312$ mK) as a function of driving frequency of pulses into the heater on a post for $\Delta\mu = \mu - \mu_0 = -0.14$ mK. Note the increase in the flow of ⁴He for $T > T_{\mathrm{pw}}$ compared to $T < T_{\mathrm{pw}}$ which corresponds to the increase in film thickness on the Rb. The line at 300 Hz shows the frequency at which the temperature scans in Fig. 3 are taken. The critical frequency f_c, where the flow rapidly drops for $T = 130$ mK, is independent of the dynamic equilibrium but the vertical scale of the flow curve depends on the initial film thickness on the post for $f < f_c$ but is independent of it for $f > f_c$.

are independent of temperature in the two regimes. We believe that the transitions in the flow rates at ≈150 Hz for the thin film ($T < T_{\mathrm{pw}}$) and 7×10^3 Hz for the thick film ($T > T_{\mathrm{pw}}$) on Rb is due to the superfluid film on the Rb becoming dissipative at the high flow rates. The difference being that the thick film can sustain a higher flow rate without dissipation than the thin film.

The thick film on the Rb is many tens of layers thick so it is undoubtedly superfluid. We think that the thin film is likely to be a superfluid too for the following reasons: (i) The maximum flow rate that can be attained is at least as high as that with the superfluid film on brass. The breakdown of superfluid flow depends on the smallest perimeter on the post and it is comparable on the Rb and brass. (ii) The flow curves in Fig. 2 for $T \lesssim T_{\mathrm{pw}}$ have similar shapes although very different in maximum flow and critical frequency. This suggests that both films are superfluid and only differ in thickness. In both cases the bottleneck is the film on the Rb. (iii) If the film on the Rb were normal for $T < T_{\mathrm{pw}}$, then this would give a high flow impedance which is comparable with that for the normal film around the heater for the brass post (the geometrical forms are similar, i.e., length/width ≈ 1/4). This would reduce the thickness of the film on the post so much that it would also become normal at low frequencies (≈1 Hz) and so the flow rate would drop to that of the normal film on the brass post. As the flow rates for the Rb thin film are 2 orders of magnitude higher, we can exclude this possibility.

If we compare the flow rate of the superfluid film on the brass post with the flow rate of the thin film on Rb, and use this ratio to scale the thicknesses, we find that the thin

film thickness, averaged over different measurements, is of order of 1 layer.

To measure the prewetting line we make temperature scans at constant chemical potential ($\Delta\mu < 0$ where $\Delta\mu = 0$ at coexistence of the vapor and bulk liquid). The values of $\Delta\mu$ are fixed by gravity by having the surface of the bulk ⁴He in the cell at various distances (h) up to 290 mm below the Rb band. Then $\Delta\mu = -mgh$ where m is the atomic mass of ⁴He. The values of the height h above the liquid are determined by partially filling 1 cm³ pots connected to the cell by a capillary tube. This arrangement enables the liquid ⁴He level to be set to ± 5 mm.

The repetition frequency, for the temperature scans, has to be chosen so that a change in flow rate across the Rb can be seen when the film thickness on the Rb changes. We choose a frequency between the two peaks in Fig. 2 for the temperature scan. This frequency is above the critical frequency for dissipative flow on the Rb in the thin film state but is low enough not to exceed the critical frequency on the Rb in the thick film state. Before the temperature is scanned, the system is allowed to come to a dynamic equilibrium at the lowest temperature, with the heater pulsed at the chosen frequency. It is usually left overnight in this state. This gives sufficient time for the film on the post, above the Rb, to come to an equilibrium thickness which is considerably less than the thickness when there is zero flow.

Two temperature scans, at the same $\Delta\mu = -0.14$ mK, are shown in Fig. 3. The measurement is reproducible and clearly shows a transition at 312 mK. The increase in amplitude of the signal at this temperature is due to the ⁴He film on the Rb getting thicker so reducing the dissipation to a negligible amount. The loss of the impedance to flow on the Rb means that the film thickness on the post above the Rb can increase and so the amount of ⁴He evaporated per pulse increases.

Taking the temperature of the maximum gradient in the temperature scans as the thin-thick transition point, we can plot a prewetting line in the $\Delta\mu$-T plane. The data for two runs is shown in Fig. 4. The solid line is $\Delta\mu = -\{2[\sigma_{lv}(T_w) - \sigma_{lv}(T)]/3\rho_0(\Delta C_3)^{1/3}\}^{3/2} K$ [6], where ρ_0 is the atomic number density of ⁴He, ΔC_3 is the difference in the van der Waals constants for Rb and ⁴He, i.e., $\Delta C_3 = 754 - 130$ K Å³, σ_{lv} is the bulk surface free energy of the liquid-vapor interface of ⁴He, $T_w = 309$ mK, and K is an arbitrary constant. To our surprise this formula with $K = 1$ is not consistent with the data points for Rb as it is for Cs [6]. To get correspondence between the experimental data and the theoretical line we need $K = 10^3$.

We can calculate the thick film thickness when the Rb is wetted at a given $\Delta\mu$ from $\Delta\mu = -\Delta C_3/d^3$: Using $\Delta C_3 = 624$ K Å³ and 3.6 Å per layer we find the thickness of the thick film on Rb to be 46 layers for $\Delta\mu = -0.14$ mK. We finally compare the thickness of the ⁴He films on the Rb in the thick and thin states from the maximum flow rates. Although there are reasons in principle why it is incorrect [16], we assume that the dissipation
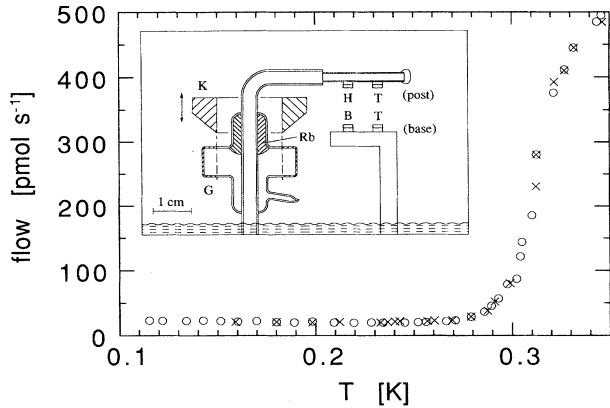
FIG. 3. Temperature dependence of flow of ^{4}He over the Rb for $\Delta\mu = -0.14$ mK with pulse frequency 300 Hz into the heater. The increase of the flow at ≈ 0.3 K indicates the transition from *thin* film to *thick* film state. The prewetting temperature $T_{pw} = 312$ mK is taken as the maximum slope of the curve. The temperature dependence of flow is reproducible: $\times$ and o are two measurements on different days. Inset: Schematic representation of the cell with the circular knife K exposing the Rb band in the glass toroid G at low temperatures and the heater H, bolometer B, and thermometers T with electrical wiring sealed into the post [14].

process is the same in the two thicknesses and so the critical flow rates just scale as the film thicknesses. The ratio of the maximum flow rates (see Fig. 2) for the *thick* to *thin* states is 110 for $\Delta\mu = -0.14$ mK. This gives an estimated *thin* film thickness of 0.4 layer relative to the calculated *thick* film of 46 layers. While we only expect the flow rates to give indications of the film thickness because of the assumptions that have to be made, it is a check on the self consistency of our results that the 0.4 layer is comparable to the 1 layer estimated above.

In conclusion, we have found a prewetting line and estimated the wetting temperature for ^{4}He on Rb. $T_w \approx 309$ mK is much lower than that found for Cs. This low
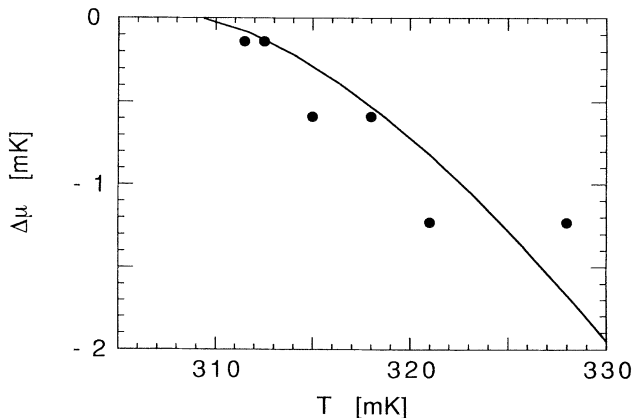

FIG. 4. The prewetting phase diagram for ^{4}He on Rb close to liquid-vapor coexistence. There are two measurements at each $\Delta\mu$. The experimental points show $T_w \approx 309$ mK. The full line is the theoretical prediction [6] multiplied by a constant 10^3. The prewetting critical point is not located.

value of T_w indicates that Rb is likely to be the only other element besides Cs that is not wetted by ^{4}He. This prewetting line is not consistent with theory [6] nor with previous data [17]. We have shown that the *thin* state of ^{4}He on Rb appears to be superfluid for $\Delta\mu \cong 0$, if this is so, then on the prewetting line there are two superfluid films of disparate thicknesses which coexist.

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[15] There are two characteristics of the flow curve for $T < T_w$ that indicate that the critical frequency f_c is due to the film on the Rb and not due to the area around the heater. First, at f_c the flow only drops by a factor of 0.5 and it is still 2 orders of magnitude greater than the flow of the normal film on brass. The second indicator is that it takes $\approx 5 \times 10^4$ pulses to halve the flow rate as f is increased beyond f_c. These pulses are necessary to reduce the film thickness on the post above the Rb to get to the new dynamic equilibrium. There is no such reservoir of ^{4}He to be removed if the transition occurs around the heater.

[16] The thick and thin films have the characteristics of three and two dimensions, respectively.

[17] Our "prewetting line" has such a steep slope that its extrapolation to higher temperatures does not pass through the prewetting points in [7,8] which are much closer to the $\Delta\mu = 0$ axis.