# Experimental Survey of Wetting and Superfluid Onset of <sup>4</sup>He on Alkali Metal Surfaces

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We present a survey of the wetting behavior of <sup>4</sup>He on evaporated films of rubidium, potassium and sodium. <sup>4</sup>He wets these surfaces at all temperatures. Off coexistence on rubidium and potassium there is a prewetting transition that is closely coupled with superfluid onset and possibly a surface tricritical point where the prewetting transition and superfluid onset intersect. Sodium has a prewetting critical temperature well below 1 K. Wetting and superfluid onset phase diagrams for <sup>4</sup>He on rubidium and potassium are presented and compared with that of <sup>4</sup>He on cesium.

#### **1. INTRODUCTION**

The study of helium films on strong substrates such as gold, glass, or mylar is a classical subject of low temperature physics.<sup>1-3</sup> <sup>4</sup>He wets these substrates so the thickness of liquid films can be continuously adjusted from the submonolayer regime to macroscopic thickness by varying the chemical potential. The only phase change that occurs in these films is the transition from normal to superfluid, which takes place via the Kosterlitz-Thouless (KT) mechanism.<sup>4-5</sup> Recently, it has become clear that alkali metals represent a qualitatively different type of weak substrate. Weak substrates typically exhibit a prewetting transition at which the film thickness jumps discontinuously.<sup>6</sup> Specifying the phase diagram of <sup>4</sup>He on a weak substrate therefore requires locating the first order prewetting line (with its associated critical point  $T_c^{pw}$ ) as well as the KT line.

Figure 1 illustrates the types of phase diagrams that are possible as the substrate potential is varied from the weak to the strong limit. For a very

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Fig. 1. Sketches of likely phase diagrams for helium films on various substrates of increasing attractiveness (a to d). Coordinates are chemical potential offset from bulk coexistence and temperature. The dotted line represents the KT line and the solid line is the prewetting line where thin and thick unsaturated films coexist.

weak substrate the prewetting line meets the coexistence curve  $\Delta \mu = 0$  at a finite temperature  $T_w$  as shown in Fig. 1a. The KT line meets the prewetting line at a critical endpoint and terminates there. It is likely that for the weakest substrates the thin film phase is always too thin to be superfluid.<sup>7</sup> As the substrate strength is increased, both  $T_w$  and  $T_c^{pw}$  decrease. For sufficiently strong substrates the prewetting line will not intersect the coexistence curve even at T=0, as shown in Fig 1b. We also expect that for a range of substrate strength  $T_c^{pw}$  will lie on or near the KT line, and the two lines will meet at a tricritical point (Figure 1b). As the substrate attractiveness is increased further, the prewetting and KT phase boundaries will not intersect at all (Fig 1c) In this regime, the KT line might bend into the thin film region or branch (Fig.1b,c) so that superfluid films with two different thicknesses might coexist along the prewetting line. For strong substrates, even  $T_c^{pw}$  is less than zero, and there is no remnant of the prewetting transition as shown in Fig 1d. The alkali metals provide an experimental means of tuning the substrate strength through the various regimes illustrated in Figure 1. Cesium, the weakest substrate known, has been extensively studied  $^{8-10}$ , and corresponds to the phase diagram of Fig 1a. The stronger binding alkalis Rb, K and Na have received less attention  $^{11-16}$ . This paper presents adsorption isotherm data for helium films on these intermediate strength alkali metals with particular attention to the interaction between prewetting and superfluidity.

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## 2. EXPERIMENT

The quartz microbalance technique is well suited to surveying the phase transitions of <sup>4</sup>He films. The surfaces of the microbalances resonate in a nearly pure shear mode. As a result superfluid onsets are revealed by a sharp drop in the frequency and a peak in the dissipation. However, a small admixture of longitudinal motion in the mode provides some coupling to the superfluid component of the film so that even at low temperatures, thickness changes in the <sup>4</sup>He film are detectable. The mass sensitivity is 0.25 Hz per normal layer and typically 0.025 Hz per superfluid layer. Alkali metal surfaces are prepared by evaporation from a pure metal source onto the gold electrodes of the microbalance. After depositing 75 layers on each side, the surfaces are annealed at 77 K for thirty minutes. While evaporating and annealing, the experiment cell is held below 7 K to prevent contamination of the surface.

We measure the resonant frequency and dissipation of the microbalance while varying the <sup>4</sup>He pressure. The results are presented as plots of the frequency shift,  $-\Delta f$ , and the change in the real part of the impedance,  $\Delta R$ , of the microbalance as functions of  $\Delta \mu$ , the chemical potential offset from liquid-vapor coexistence. Above 0.8 K we can record data while increasing or decreasing  $\Delta \mu$ . Examples for Rb are shown in Ref. 16; K behaves similarly. On Rb and K at low temperature the microbalance response is highly hysteretic. The isotherms are used to construct phase diagrams like the ones sketched in Fig. 1 for Rb and K.

## 3. RESULTS AND DISCUSSION

Figure 2 shows isotherms of  $-\Delta f$  and  $\Delta R$  as functions of  $\Delta \mu$  measured at 1.6 K on Rb, K and Na surfaces. The data in Fig. 2 were taken as  $\Delta \mu$ was increased from low values to 0. On all three surfaces  $-\Delta f$  is large at  $\Delta \mu$ = 0, indicating that the surface is wet. The surfaces remain wet down to the lowest temperatures we have studied, 0.15 K in the case of Rb. Zero temperature wetting for the case of Rb is consistent with Ref. 11, but not Ref. 13. Among the alkali metals, only Cs has a finite  $T_w$  and a phase diagram like the first panel of Fig. 1.<sup>8</sup> On Rb, K and Na the adsorbed mass at low  $\Delta \mu$  is quite small, virtually 0 for Rb and K. As  $\Delta \mu$  is increased, in each case a region of relatively rapid increase of  $-\Delta f$  is reached. It lies around  $\Delta \mu = -0.07$  K, -0.18 K and -1.4K for Rb, K and Na, respectively. This trend is consistent with the expectation that the adsorption potential deepens as the alkali metal becomes lighter.<sup>6</sup> Below 1.9 K for Rb and 1.75 K for K the location of this region is hysteretic. It occurs at considerably lower  $\Delta \mu$  when the microbalance response is measured while decreasing  $\Delta \mu$ .



Fig. 2. Helium absorption isotherms at 1.6K on rubidium, potassium and sodium substrates. In each pair of graphs the left one is the frequency shift of the microbalance versus chemical potential offset. Frequency shift have been corrected to eliminate shifts due to pressure effects. The graphs in the right column are the change in the microbalance's dissipation. Notice that each substrate is plotted with different horizontal scales.

Hysteresis is the hallmark of a first order transition. In the temperature range it occurs, we associate the rapid increase of  $-\Delta f$  with prewetting and identify the temperature at which it disappears as  $T_c^{pw}$ . The value of  $\Delta \mu$  in the center of the rapid increase is plotted as a function of T for Rb and K in Fig. 3. Also shown as heavy closed circles are the prewetting critical points for these two metals. We have measured isotherms on Na down to 1.5 K and find no hysteresis. Consequently the prewetting critical point for Na must lie at a lower temperature. The increase in  $-\Delta f$  seen for Na in Fig. 2 is due to an enhanced two-dimensional compressibility, as seen with Cs above  $T_c^{pw}$ .<sup>8</sup>

In the  $\Delta R$  isotherms shown in Fig 2 there are sharp peaks at  $\Delta \mu = -0.065$  K, -0.1 K, and -0.95 K for Rb, K and Na, respectively. In the case of Rb and Na there are also sharp drops in  $-\Delta f$  at the same chemical potentials, while for K there is a small decrease in slope in the  $-\Delta f$  isotherm. We identify these features as superfluid onsets. The peak in  $\Delta R$  is the well-known finite dissipation peak characteristic of the KT transition and the drop in  $-\Delta f$  is due to the jump in the superfluid density at the transition. For Rb and K, the value of  $\Delta \mu$  at the maximum value of  $\Delta R$  is plotted as a function of T on the phase diagrams of Fig. 3.

In Fig. 3 for Rb the superfluid transitions lie just above the prewetting



Fig. 3. Complete phase diagrams for helium on rubidium (a) and on potassium (b). Triangles are prewetting transitions and circles are superfluid onset. The solid lines connect the forward isotherm prewetting locations. The KT line is shown with the dotted line. Error bars on all symbols, except prewetting locations on potassium, are smaller than the symbol size.

transitions. For both Rb and K, the superfluid onsets parallel the prewetting line below  $T_c^{pw}$  and turn abruptly upward at it. Surprisingly, when isotherms are measured as decreasing functions of  $\Delta \mu$ , we find that the positions of the superfluid onsets are also hysteretic and shift to lower values of  $\Delta \mu$ . To eliminate gross surface roughness as the source of the hysteresis, we have measured <sup>3</sup>He isotherms on the same Rb surface discussed here. Absolutely no hysteresis was seen.<sup>16</sup> The data might suggest that on Rb and K the prewetting and superfluid onsets are coupled into a single transition that retains some of the character of both prewetting and the KT transition. If this is the case, the heavy closed circles in Fig. 3 are actually tricritical points. It is clear that experiments that measure the total adsorbed mass would give valuable insight into the nature of this combined transition. It is known that critical points tend to "attract" second order phase transition lines and that the formation of tricritical points is therefore rather robust.<sup>17</sup> This might explain why both Rb and K, with considerably different prewetting chemical potentials, can display this behavior. It is clear that further work is needed to investigate this speculation.

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### 4. CONCLUSIONS

Rb, K and Na offer the opportunity to study the adsorption of superfluid films on substrates with adsorption strengths intermediate between the weak binding Cs case and the strong binding cases. The phase diagrams of Rb and K are in the category represented by Fig. 1b. Superfluid onset and prewetting are somehow coupled on these surfaces. Na is a considerably stronger substrate with a  $T_c^{pw}$  somewhere below 1 K. An interesting possibility is that the Na phase diagram resembles Fig. 1c. If this is the case prewetting may mark the coexistence of thin and thick superfluid films on Na.

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