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HELIUM WETTING AND PREWETTING PHENOMENA AT FINITE TEMPERATURES

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Helium wetting and prewetting phenomena at temperatures $T > 0$ are addressed. Emphasis is given to the weak-binding substrates Cs and H_2, which have been predicted and observed to be nonwet and wet, respectively, by ^4He at low temperatures. Calculations of the wetting temperature T_W and the prewetting line near T_W for Cs, as well as the prewetting line near $T = 0$ for H_2, are given. Predictions concerning the interplay between superfluid onset and prewetting are made, and prewetting critical temperatures are estimated.

The recent prediction of ^{4}He non-wetting of Cs[1,2] and its subsequent experimental confirmation[3,4,5,6] have produced a high level of interest in the physics of helium adsorption on weak-binding substrate. Our earlier work[1,2] focused primarily on low temperature phenomena. Here we address helium wetting and prewetting phenomena at temperatures $T > 0$.

There are two generic situations at $T = 0$. The most common is that of wetting with prewetting. Here the thickness of an unsaturated film undergoes a finite jump followed by a smooth increase to macroscopic size as coexistence is approached. The solid H_2 substrate[7,8] will be our example here. The second, and most striking, case is that of non-wetting, where the Cs substrate provides the only verified example. In this case a transition to wetting, with prewetting, is expected for some T less than the liquid-gas critical temperature.[2,9] Very recently, this has been observed by Taborek and Rutledge[6] in experiments providing the first direct observation of prewetting.

We commence with a calculation of the wetting temperature T_W and the prewetting line near T_W for the case of Cs. Following the argument leading to the approximate criterion of Eq.(1) of Ref. 1, generalized to include

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the temperature dependence of the liquid-vapor surface tension $\sigma_{lv}(T)$, the condition for a wetting transition at temperature T_W becomes

$$\frac{\sigma_{lv}(T_W)}{\sigma_{lv}(0)} = \frac{0.6\rho_0(C_3D^2)^{1/3}}{\sigma_{lv}(0)} - 1. \tag{1}$$

Here D is the well depth and C_3 the van der Walls coefficient of the substrate-adsorbate potential.[1] Temperature dependence arising from excitations at the substrate-liquid interface has been ignored due to the relatively large energy cost of creating such excitations. Use of experimental data for $\sigma_{lv}(T_W)$[10] in Eq.(1) produces wetting transition curves in the $C_3 - D$ plane. Agreement with the observed value $T_W = 1.95$ K on Cs[6] can be achieved by adjusting the D for Cs to 8.6 K (from the value 4.41K used in Ref. 1). Similar adjustments put K in the wetting regime, leaving Rb in the nonwetting regime but with a wetting transition temperature under 1K.

We next look at prewetting for $T > 0$. A Clausius-Clapeyron derivation[11] of the slope of the prewetting line in the $\Delta\mu - T$ plane, where $\Delta\mu$ is the deviation in the chemical potential from its value at coexistence, gives

$$\frac{d\Delta\mu}{dT} = -\frac{s_{lv}}{\rho_0 l} \tag{2}$$

where $s_{lv} = -d\sigma_{lv}/dT$ is the liquid-vapor interfacial entropy per unit area. We have neglected the vapor density compared to ρ_0 and the thin film entropy compared to that (s_{lv}) of the thick film. Next, for the thick film, $l = [\Delta C_3/(-\Delta\mu)]^{1/3}$,[12] where $\Delta C_3 = C_3 - C_3^{He}$, and $C_3^{He} = 130\text{K\AA}^3$ is the van der Waals coefficient for the case of a helium substrate. Neglecting the temperature dependence of ρ_0 then permits integration of Eq.(2) to produce

$$\Delta\mu = -\left[\frac{2\sigma_{lv}(0)}{3\rho_0(\Delta C_3)^{1/3}}\left(\frac{\sigma_{lv}(T_W) - \sigma_{lv}(T)}{\sigma_{lv}(0)}\right)\right]^{3/2}, \tag{3}$$

valid for T sufficiently near T_W.

We estimate that the prewetting critical temperature T_c^{PW} for the case of a Cs substrate is given by $T_c^{PW} = a\epsilon$ where $\epsilon \equiv \sigma(l_{max}) - \sigma(0)$ is the barrier height in the $T = 0$ surface energy $\sigma(l)^2$ as a function of l, and a is a constant. Here l_{max} is the position of the maximum in $\sigma(l)$. For the choice $D = 8.6$ K, we find $\epsilon \approx 1.6$ K. The observed[6] value of T_c^{PW} is 2.5K, corresponding to $a = 1.56$. In Fig. 1(a) we plot Eq.(3) from $T_W = 1.95$K to the observed $T_c^{PW} = 2.5$K. The data of Ref. 6 is also shown; the agreement between theory and experiment is very good, given the noise in the data.

For $T = 0$ prewetting Eq.(2) shows that the prewetting line starts out with zero slope at $T = 0$. For a H_2 substrate, we have predicted that prewetting occurs at $\Delta\mu = -3.59$ K with a thickness jump of 0.43 layers.[1, 2] In this case the substrate potential is so strong that the jump is nearly a two-dimensional liquid-gas transition; indeed the actual 2D condensation density at $T = 0$ is 0.56 layers.[13] Thus we expect that T_c^{PW} is of order the 0.8K

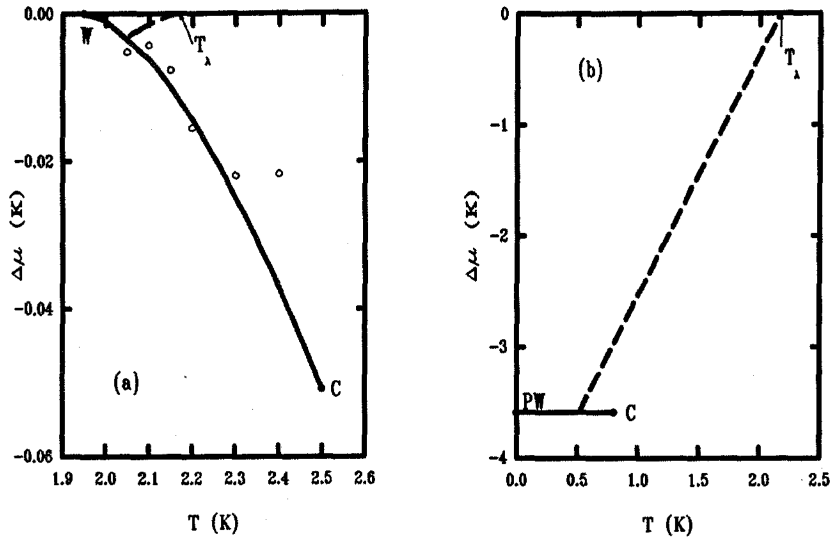

Fig. 1 (a) Phase diagram for ^4He on Cs, showing the prewetting transition line (full) connecting the wetting transition W with the prewetting critical point C, and the Kosterlitz-Thouless transition line (dashed). Open circles are data of Ref. 6. (b) Phase diagram for ^4He on H_2 showing the prewetting transition line (full) connecting the $T = 0$ prewetting transition PW with the prewetting critical point C, and the Kosterlitz-Thouless line (dashed).

binding energy of 2D helium. The prewetting line based on these results is shown in Fig. 1(b), where we ignore deviations from zero slope due to thermal excitations in the film. For H_2 the observed coverage jump of about 0.5 layers[7] at low T is consistent with our calculated prewetting jump.

It is important to understand the interplay between superfluid onset and prewetting. We begin by asking whether the thin prewetting film at the prewetting transition can be superfluid. Treating this film as a 2D gas with density ρ_2 (at $T = 0$ it has thickness $l = 0$, corresponding to an infinitely dilute gas) in equilibrium with the 3D vapor gives

$$\rho_2 = 0.2\rho_2^{KT} e^{-\beta(\epsilon_0 - \mu_0 - \Delta\mu)} \tag{4}$$

where ρ_2^{KT} is the density at superfluid onset[14] via the Kosterlitz-Thouless transition, ϵ_0 is the binding energy of a single ^4He atom on the substrate and $\mu_0 = -7.15$ K is the chemical potential at bulk coexistence. For Cs, if $D = 8.6$ K, $\epsilon_0 \approx -3.6$ K, and Eq.(4) predicts $\rho_2 \ll \rho_2^{KT}$ for all circumstances of interest and hence no superfluidity in the thin prewetting film. For H_2, $\epsilon_0 \approx -14$ K and $\Delta\mu = -3.59$ K, so that once again Eq.(4) predicts no superfluidity in the thin wetting film.

We next estimate the superfluid transition temperature on the thick film side of the prewetting transition for the case of Cs. Here, as $T_W = 1.95$ K is near the bulk λ-temperature $T_\lambda = 2.17$ K, we use the experimental result[15]

$$l = l_0 \left(\frac{T_\lambda}{T_\lambda - T_c}\right)^{\hat{\nu}} \tag{5}$$

where T_c is the superfluid transition temperature of a film of thickness l, $l_0 = 12.3$ Å, and $\hat{\nu} = 0.52$. Relating l to $\Delta\mu$ for thick films then gives

$$\Delta\mu = -\frac{\Delta C_3}{l_0^3}\left(\frac{T_\lambda - T_c}{T_\lambda}\right)^{3\hat{\nu}} \tag{6}$$

which appears as a dashed line in Fig. 1(a).

For H_2 we employ the Kosterlitz-Thouless relation $T_c = 0.82\pi\hbar^2\rho_2/2mk_B$. Use of our $T = 0$ result $\rho_2 = 0.033$ Å^{-2} (= 0.43 layers) gives $T_c = 0.52$ K, substantially below the estimated prewetting critical temperature of $T_c^{PW} = 0.8$ K. The superfluid transition line connecting T_λ with $T_c = 0.52$ K has been sketched as a dashed straight line in Fig. 1(b). Recent third sound studies[8] showing two modes do not seem to fit into the simple picture of Fig. 1(b) and are perhaps not related to wetting phenomena.

Finally, we note that the phase diagrams shown in Fig.1 imply that there is a minimum temperature at which the transition to superfluidity is via the Kosterlitz-Thouless mechanism. For lower temperatures superfluid transitions occur via first order prewetting transitions. It would be of great interest to test these predictions experimentally.

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