The growth of the non-wetting liquid ^4He on Cs

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Abstract

It was theoretically predicted that the heavy alkali metals provide the only surfaces non-wetted by superfluid ^4He below a certain temperature T_w . This was experimentally proven both for Cs and for Rb. However, investigations have shown that the non-wetting thin-film state for the He–Cs system is extremely dilute for $T \ll T_w$, yet close to T_w it can be much thicker and of the order of monolayers. Using the photoelectron tunneling method we have sensitively measured the growth of the non-wetting thin-film state of ^4He on a quench-condensed Cs surface. It turns out that far from co-existence there is little adsorption of helium. In contrast, close to co-existence a rapid growth up to two to three monolayers of helium is observed, but the surface is still non-wet under the usual convention.

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1. Introduction

Since the prediction [1] that the alkali metals Cs, Rb and K are the only surfaces non-wetted by superfluid ^4He at $T = 0$ K, it was experimentally proven that Cs [2–4] and Rb [5] indeed show non-wetting. For ^4He on Cs the wetting temperature T_w is about 2.0 K. At very low temperatures a macroscopically thin ^4He film exists on Cs showing a two-dimensional (2D) gas like behaviour [6], but a thicker helium film may exist close to T_w [7]. On Rb it was shown that for $T < T_w$ the non-wetting thin ^4He film is about 0.3 monolayers (ML) thick and superfluid [5].

These systems are excellent for studying the phenomena of non-wetting and pre-wetting, difficult to do with classical fluids. However, it turned out that the wetting behaviour, determined by measured contact angles [4,8,9] is strongly influenced by the method used to prepare the cesiated substrate: quench-condensed Cs, as it is obtained by evaporation at low temperature, (expected to cause a rough surface) shows weaker non-wetting than Cs grown from the liquid state (which allows for a smooth surface to develop). This difference

in wetting behaviour is mainly attributed to the microscopic roughness of the Cs [10].

We have studied the non-wetting thin film state on a quench-condensed (and hence rough) Cs surface using the photoelectron tunneling method [11]. Close to liquid-vapour coexistence there is a sharp drop in photocurrent which indicates a transition from a monolayer to a multilayer film. Even in this regime the Cs surface shows non-wetting behaviour, according to the usual definition. From the temperature dependence of this transition a phase diagram for the monolayer and multilayer regime, respectively, can be constructed.

2. Experimental setup

The experimental setup consists of two different types of measuring techniques, which can be used simultaneously. On one hand we determine the ^4He thickness, from one monolayer up to a thick saturated film, by means of the Surface Plasmon Resonance (SPR), a technique which is described in detail in Ref. [12]. This method allows to determine the *average* thickness of the adsorbate (averaged over regions on the scale of the wavelength of light), and is not further discussed here, because we would like to focus on that fraction of the

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Fig. 1. Sketch of the experimental setup: D, cesium dispenser; E, electrodes; I, electrometer; L, achromatic lens; M, mirrors. The inset on the right side shows a top view on the Cs substrate evaporated onto the glass prism.

surface which is in the non-wet state (from our previous experiments [12] we conclude that on rough Cs surfaces there exist microscopic areas which are wet and others which are dry). For this reason we have used for the present measurement a complementary technique, photoelectron tunneling, which allows to very sensitively resolve the thickness of an adsorbed helium film on the Cs from a submonolayer up to about four layers. The experimental setup is sketched in Fig. 1. Light coming from a monochromator (in the figure to the right) is focused via an achromatic lens onto the cesiated surface. The achromat can be positioned in the xy -direction using stepping motors whereby the focus can be scanned over the whole cesiated area. To check the position and the focus of the beam on the surface a CCD camera is mounted on the opposite side. The area of the focused beam on the surface is about 1 mm^2 , whereas the whole cesiated area is about 1 cm^2 .

The method used to prepare the Cs film was as follows: we first cooled down the evacuated cell to 4 K and afterwards slowly heated up the Cs dispenser. Meanwhile we pumped on the cell to make sure that the impurities desorbed from the dispenser were removed. By opening a shutter then a quench-condensed thin Cs film was deposited onto the Ag substrate.

We used the method of photoemission also to probe the chemical state of the Cs surface, i.e., by locally measuring the work function to check for chemical impurities before helium was admitted into the sample cell. This was done by measuring the photocurrent while scanning the wavelength of the incoming monochromatic light from 350 to 850 nm. For the Cs used in these studies the work function is about 1.9 eV [12]. Measuring the wavelength dependent photocurrent also allowed us to find the wavelength at which the photocurrent had its maximum value. This wavelength was then used during the adsorption measurements.

Our adsorption data were taken in the range of 1.3 to 2 K. At fixed temperature we measured the intensity of

Fig. 2. Normalized photocurrent as function of the reduced pressure ($I_{\max}$ is the current at the beginning of the run).

the photocurrent while ^4He gas was very slowly condensed into the cell and adsorbed on the Cs surface. Several runs were done at different temperatures, and the decay of the photocurrent as saturated vapour pressure, P_0 , was approached showed qualitatively the same behaviour during each run, see Fig. 2. Before the gas adsorption was started we pumped the cell in each run until the photocurrent increased to a final maximal value, indicating that most of the helium was removed from the cell and so the Cs should be dry. It turned out that at higher temperatures we got a slightly higher value of the photocurrent maximum. This is understood as the last fraction of helium layer adsorbed on the Cs is better removed by pumping at higher temperatures (i.e., $P/P_0 \rightarrow 0$).

3. Results and discussion

In order to discuss our experimental results, we have first to address the question of how to interpret the data

which are obtained by means of the photoelectron tunneling technique.

As already indicated, we measure the photocurrent I emitted from the Cs surface upon irradiation with an external light source, and from the observed reduction of I as more and more He is adsorbed on the Cs get information about the He coverage.

In the coverage range *above one monolayer* a simple picture appears adequate, which has already successfully been used for the analysis of H_2 adsorption on Ag [11]: One can consider the adsorbed layer as an additional potential barrier on the metal surface, which the photoelectrons can only pass by a tunneling process. In the case of liquid ^4He the potential barrier which the helium surface provides for electrons is known to have a height of about 1 eV [13]. The transmission coefficient T_r through such a one-dimensional rectangular barrier of width d follows a relation $T_r \propto \exp(-2\chi d)$, where χ represents the decay rate and includes the potential barrier dependence [14]. Since the thickness of one monolayer of ^4He is about 0.35 nm, one obtains a drop in the transmission coefficient of roughly one order of magnitude per monolayer [11].

In the coverage range *below one monolayer*, on the other hand, this picture assuming an effective potential barrier which is distributed homogeneously across the Cs surface is most likely not appropriate. Still the presence of helium atoms on the Cs tends to reduce the photocurrent. Unfortunately the knowledge about the photoelectron mechanism in this region is sparse, and moreover a change in the effective work function of the Cs due to the presence of He might take place. In spite of this uncertainty in the interpretation, in the following we include the data also in this regime for completeness.

Fig. 2 presents the decay of the normalized photocurrent as a function of the reduced pressure at three different temperatures. One can clearly distinguish two decays of the intensity of the photocurrent separated by a region where the current is almost constant or is only slightly decreasing over a long range of the reduced pressure. The higher the temperature, the steeper the decay, and hence the plateau becomes broader. If we assume—as indicated above—that one order of magnitude in the decay of the photocurrent is associated with the growth of a helium monolayer on the surface, then the first decay represents the completion of nearly one monolayer of helium on Cs. The growth of this layer is relatively slow at 1.36 K and gets faster as the temperature is raised to 1.93 K. It should be noted that in this range, where the photocurrent measurements yield a helium coverage of less than a monolayer, the SPR technique indicates a much higher *average* film thickness in the nm range. This apparent contradiction can be resolved by assuming that due to the inhomogeneities of quench-condensed Cs only part of the surface is actually in the non-wet state [12].

Very close to coexistence (i.e., $P/P_0 \rightarrow 1$) we find a very rapid drop in photocurrent of nearly two orders of magnitude. Although this current gets close to the noise level of our measurement it is still finite at $P = P_0$. On the basis of the estimates given above the thickness of the helium film in this range is two to three monolayers.

At the end of the adsorption, we carefully pumped out the cell in order to check for hysteresis. The photocurrent signal followed the same path as for the adsorption without any indication of hysteresis effects.

In the submonolayer regime, far from coexistence, the adsorbed helium atoms are expected to behave like a 2D-gas. The surface coverage, $n_{2D\text{-gas}}$, in this regime is given by Klier and Wyatt [15]:

$$n_{2D\text{-gas}} \propto T \exp\left(\frac{\mu - \varepsilon_1}{k_B T}\right), \quad (1)$$

where ε_1 is the binding energy of a single ^4He atom to the Cs (with $\varepsilon_1 \approx 3$ K). In order to show our results in this regime more clearly we have plotted in Fig. 3 the photocurrent for several temperatures vs. chemical potential offset from coexistence $\Delta\mu = \mu_{\text{cell}} - \mu_0$ (where $\mu_0 = \mu(P_0)$), which related to the reduced pressure by

$$\Delta\mu = k_B T \ln\left(\frac{P}{P_0}\right). \quad (2)$$

As the system approaches the saturated vapour pressure (i.e., $\Delta\mu \rightarrow 0$) the surface coverage increases as well and gets very close to a monolayer completion, which is associated with the levelling off in the photocurrent signal. Close to saturated vapour pressure, the surface coverage exhibits a steep increase, which can be interpreted as the transition from one monolayer to the 2–3 monolayer regime. Yet, according to usual convention the system is still in the non-wet state. The maximum helium thickness in that range is hardly temperature dependent. However, the value of chemical potential $\Delta\mu$ where the drop in the photocurrent occurs

Fig. 3. Photocurrent I vs. chemical potential offset $\Delta\mu$.

Fig. 4. Regimes in the $\Delta\mu$ vs. T plane with coverages less than 1 monolayer (hatched) and greater than 1 monolayer, respectively, on a quench-condensed Cs surface.

depends strongly on temperature, as shown in Fig. 4. For increasing T this transition occurs closer and closer to coexistence. Besides, as is apparent from Figs. 2 and 3, the photocurrent data exhibit an increasing spread upon approaching the transition from below. This broadening reflects temporal oscillations in the photocurrent, indicating that the system becomes less and less stable close to the transition. The higher the temperature, the bigger the surface coverage oscillations are (see error bars in Fig. 4) and hence it becomes difficult to estimate the value of the chemical potential at higher temperature.

4. Conclusions

We have shown that the photoelectron tunneling method allows to study the growth of the non-wetting thin film state of ^4He on a quench-condensed Cs surface with high resolution. Far away from coexistence with bulk liquid the system is dilute with a helium coverage of less than 1 monolayer. Close to coexistence a transition

to a film with a thickness of a few monolayers is observed, but the surface is still non-wet according to usual convention. Detailed measurements of this transition and of the multilayer regime are in progress.

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