Adsorption of Thin Liquid Helium Films on Cs Measured via Photoelectron Tunneling

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The alkali metals Cs and Rb are the only surfaces which are not wetted by superfluid ^4He below a certain temperature. In our experiments, using the photoelectron tunneling method, we can highly resolve the growth of the non-wetting thin-film state of ^4He on a quench-condensed Cs surface. It turns out that far from coexistence there is little adsorption of helium. In contrast, close to coexistence a rapid growth up to two monolayers of helium is observed, but the surface is still non-wet under the usual convention.

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1. INTRODUCTION

The wetting of alkali metal substrates by superfluid ^4He gained a significantly increased interest during the last decade. In 1991, Cheng *et al.*¹ predicted that ^4He should not wet Cs, Rb, and K at $T = 0$. Experimentally it was found that Cs is not wetted at $T < T_w \approx 2\text{K}$ ²⁻⁴. It was also demonstrated that Cs is extremely dry at very low temperatures and a microscopically thin film exists forming a 2D gas⁵. The first order phase transition is observed at the wetting transition^{3,4} and the prewetting line has been found for the first time³. It is also observed that differently prepared Cs surfaces result in different wetting behaviour: low-temperature quench-condensed Cs shows weaker non-wetting than Cs grown from the liquid state. This is understood that the first method causes a rough Cs surface, whereas the latter should create a smooth one.

This paper focuses on the features of the non-wetting thin film state. We use the photoelectron tunneling technique to resolve the growth behaviour of thin helium films, especially in the non-wetting regime. The effect of different adsorbates^{6,7} on the tunneling of photoelectrons emitted from surfaces was studied a long time ago⁸. This method, however, has not been used so

far for helium as an adsorbate. The method provides an extremely high resolution for the first few adsorbed monolayers and so is complementary to the widely used technique of surface plasmon resonance (SPR).

2. EXPERIMENTAL DESCRIPTION

The experimental setup is shown in Fig. 1. Inside the cell a BK 7 glass prism is mounted onto which a thin Ag film (35nm) is evaporated. Measuring the helium film thickness we use the SPR technique in a Kretschmann configuration ⁹. The light spot coming from a laser diode ($\lambda_1 = 635\text{nm}$) is coupled resonantly into surface plasmons at a certain angle of incidence, θ_R . The adsorption of a ^4He film causes a detuning of the resonance minimum. Measuring the shift of θ_R allows us to resolve the thickness of the ^4He film in the range of 1 to 500Å with an accuracy of about 2Å.

Fig. 1. Experimental setup (schematically): D $\equiv$ cesium dispenser, E $\equiv$ electrodes, I $\equiv$ electrometer.

After cooling down the evacuated cell to 4K we slowly heated up the Cs dispenser while simultaneously pumping on the cell in order to remove the impurities desorbed from the dispenser. By opening a shutter we quench-condense a thin Cs film onto the Ag substrate. The thickness of the Cs film has been monitored online by measuring the shift of the plasmon resonance curve. The final thickness of the Cs film is 10nm.

Simultaneous to the SPR measurements we used the photocurrent tunneling technique by illuminating the surface with light at different wave-

lengths from a monochromator. From outside we can change the position where the focused beam strikes the surface and hence map the whole Cs area. The spot size on the surface is 1mm^2 . The Ag layer and the upper electrode (kept at $+100\text{V}$) are connected to an electrometer, which measures the photoelectrons leaving the Cs surface, and, if a helium film is present, those which tunnel through this film (the potential barrier for an electron through helium is $\approx 1\text{eV}$). We are not measuring the electrons collected by the upper electrode. It is worth mentioning this as this way avoids a complicated approach of considering the angle and velocity distribution of the transmitted photoelectrons and also gives a much higher signal to noise ratio.

3. RESULTS and DISCUSSION

First we have checked the onset of the ^4He film on the bare Ag substrate by taking adsorption isotherms. Several runs at different temperatures have been used to calibrate the helium thickness related to θ_R . As expected the shape of the adsorption isotherms on bare Ag show the typical wetting behaviour. After the Cs evaporation, we have done several wavelength scans (ranging from 350 to 850nm) to check locally the Cs film quality, on different places on the surface¹⁰. These wavelength scans have intensity maxima at $\lambda_{max} = 440\text{nm}$ and at a cut-off wavelength of about 650nm, see Fig. 2.

Fig. 2. Intensity of the photocurrent as function of wavelength before ($\circ$) and after ($\bullet$) the Cs evaporation.

It turns out that there are different values of the photocurrent maxima at different positions on the Cs, but λ_{max} remains constant. The scans have

been done both by increasing and decreasing the wavelength and at different scan speeds in order to check for equilibrium. No significant differences have been observed. Using these curves, and after normalizing, we determine the work function of the Cs film as (1.91 ± 0.05) eV, this is an average for the whole surface ¹¹.

We then cooled down the cell and, in the temperature range $1.3 < T < 2\text{K}$, have simultaneously measured the He film thickness and the photocurrent intensity while He gas is very slowly condensed into the cell and adsorbed onto the Cs substrate. Fig. 3 shows the typical behaviour of the photocurrent and the SPR change during a ^4He adsorption. From the SPR the adsorption isotherms on Cs look similar to those for bare Ag, which indicates that at those temperatures the Cs is still wet. However, as will be shown below, this is only the case for a certain area of the Cs.

The thickness of the helium film, via the SPR method, grows typically up to about $100\text{\AA}$, independent of temperature. In contrast, the photocurrent curves show a different adsorption behaviour. At the beginning of the adsorption isotherms (when no helium gas is in the cell) we start with a maximum value of the photocurrent at given temperature; the higher the temperature, the higher the initial photocurrent, which is due to the fact that the last helium layer adsorbed on the surface has been better removed by pumping at higher temperatures (P/P_o gets less as T increases). The curves have the same qualitative behaviour, namely as ^4He gas is condensed into the cell the photocurrent is decreasing. In addition, we find that as the temperature increases the steeper is the decay in current for low P/P_o . The photocurrent levels off as the relative pressure gets higher. The sudden drop of the photocurrent is a measure of the thin helium film onset on the surface. Assuming the photoelectron current having an exponential decay dependence as function of growing adsorbed helium thickness (as it is expected for a tunneling process) then we expect that at the very end of the adsorption isotherm the curve should approach asymptotically very low photocurrent values, i.e. goes to zero. This is not seen in our studies and is discussed on the basis of having a rough Cs surface which results in different wetting properties.

Usually quench-condensed Cs is rough on a microscale ¹², and so our evaporated Cs film not only has an inhomogeneous thickness but also presents island growth as well. In this case, the helium condensed into the cell will adsorb and hence wet those areas of the cesiated Ag-surface where the Cs film is too thin to show non-wetting. Far away from coexistence the adsorbed helium film can be still thin enough such that electrons can tunnel through. However, after the first few monolayers have been completed, the contribution to the current becomes too small and the only remaining area

for electrons to leave the surface are the Cs islands which are non-wet. So the signal should be quite constant or decrease very slowly as coexistence is approached. The latter is expected due to the growth of the thin non-wetting helium film, which is assumed to be a 2D-gas, but there are also signs of 2D-liquid behaviour¹³.

Fig. 3. Shown is the helium thickness on Cs measured via SPR (●) and the normalized photocurrent (◇) as a function of the reduced pressure. With the SPR technique also the adsorption behaviour of a wetted helium film on Ag (□) is shown for comparison.

We observe that after a decrease of the photocurrent of nearly one order of magnitude for very low values of P/P_0 the current does not change for a wide range of pressures and only when $P/P_0 \rightarrow 1$ the current drops by another order of magnitude. When slowly pumping out the cell the current increases back to its initial values without showing noticeable hysteresis. This we interpret as a non-wetting of the Cs islands but also as a growth of the thin non-wetting film to a final thickness of one or more monolayers. This behaviour will be described elsewhere.

This result is not in contradiction to the adsorption isotherms via SPR, which averages the film thickness over a bigger area and can not distinguish between wet and non-wet patches, i.e. thick and thin He films. The more helium gas is condensed into the cell the thicker the helium film grows between the Cs islands. But this does not decrease the photocurrent as the islands are still non-wet.

4. CONCLUSIONS

We have shown that with the photoelectron tunneling technique one can resolve very thin helium films adsorbed on cesiated surfaces which hence opens the way to study the features of a thin non-wetting ^4He film adsorbed on Cs. Photoelectron spectroscopy allows to investigate the chemical state and the homogeneity of thin Cs films. In addition using the SPR method one can measure the film thickness over a wide range, with monolayer resolution. This provides information to the overall adsorption behaviour, however, can not discern the differences between wet and non-wet areas on a microscopic scale. We have qualitatively explained the observed drop in photocurrent during helium adsorption due to the growth of the thin non-wetting helium film on a patchy wetted Cs surface. The latter results from surface roughness of the Cs film. Further experiments are in progress.

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