Wetting studies of liquid ^4He on various Cs surfaces

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Cesium is a surface not wetted by liquid ^4He below a certain temperature, $T_w \approx 2\text{K}$. However, one observes that a thick helium film remains on the Cs substrate even if the surface is cooled below T_w from the wetted state. This leaves the question how the system will finally evolve from this metastable state to the thermodynamic equilibrium, represented by a microscopically thin film. We investigate the dynamic and static behavior of dewetting ^4He films on cesiated surfaces. Space and time resolved measurements of the thickness of the helium film are performed using a surface plasmon microscope. We have measured the work function of pure and oxidized Cs via photo current spectroscopy.

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1. INTRODUCTION

Liquid ^4He is extremely weakly bound to Cs and Rb surfaces and hence shows nonwetting at low temperature.^{1,2} For Cs the wetting temperature, T_w , is about 2K, i.e. for $T \geq T_w$ Cs is wetted by liquid ^4He but not for $T < T_w$, where a finite contact angle exists. At the wetting transition a clear first order phase change is observed³ and the long sought prewetting transition has been found in this system for the first time.⁴ Experiments at very low temperature have also shown that Cs is extremely dry and only a microscopically thin ^4He film exist forming a 2-dimensional gas.⁵ However most experiments show strikingly different hysteresis in film thickness and contact angle.⁶ When cooling a Cs surface below T_w which is initially covered with a thick ^4He film and in contact with a ^4He reservoir, i.e. along the coexistence line, then in most cases this film does not dewet but rather a metastable thick film remains. This hysteretic wetting behavior is attributed

to the surface properties of the underlying substrate, namely roughness and chemical impurities.

We have studied the dynamics of dewetting ^4He films on several Cs substrates. Using short-time laser pulses we could create dry patches on Cs and hence have triggered the dewetting of the metastable film. From the profile and movement of the contact line after nucleation we get information about stability and the mechanism of dewetting. For a rough analysis of our Cs surfaces we have measured the wavelength-dependent current of emitted photoelectrons from pure and oxidized Cs. In addition we have studied the dynamics of isolated superfluid ^4He drops on an otherwise dry Cs surface.

2. EXPERIMENTAL TECHNIQUES AND SETUP

To image the wetting behavior of ^4He on Cs we used a surface plasmon (SP) setup in the Kretschmann configuration.⁷ A thin Ag film ($\approx 30\text{nm}$) is evaporated onto a glass prism. The light is coupled resonantly into SPs at a certain angle of incidence, the resonance angle Θ_R , which leads to attenuated total reflection (ATR). A change of the dielectrical properties of the metal-dielectric interface due to, e.g., a He film causes a detuning of the SP resonance. Measuring the shift of Θ_R on the Ag film illuminated with a light spot ($\lambda_1 = 633\text{nm}$) allows to detect the thickness of the He film in the range of 0.1nm up to 500nm with an accuracy of less than 0.05nm in the lower and $\approx 5\text{nm}$ in the upper limit. Other metals (like Cs) cause a reduction in the resolution due to a higher damping of the SPs. To build up a SP microscope the sample is additionally illuminated with an expanded light source ($400\text{nm} < \lambda_2 < 800\text{nm}$) and the reflected light at a constant Θ_R is imaged via a CCD camera. Using this technique causes a reduction of the sensitivity (2 to 10nm) with respect to the above-mentioned setup but leads to a high spatial resolution and a time resolution only limited by the recording equipment.

Local photoelectron spectroscopy is used to characterize the Cs substrate by measuring the photocurrent when the substrate is illuminated with a light source ($300\text{nm} < \lambda_3 < 1000\text{nm}$). Scanning this light source over the substrate allows to map the work function on a length scale limited by the spot size. In addition, it is also possible to measure the scattered light from a laser reflected at the surface to characterize its roughness.

To study the dynamics of helium droplets deposited onto a surface and the developing helium films we used standard video equipment. A sketch of our experimental setup is given in Fig. 1. The sample cell contains the prism with the Ag film evaporated onto it before it was mounted into the cell.

Fig. 1. Experimental setup: He capillary (c); Cs dispenser (d); electrode (e) to measure the photocurrent; photodiode (PD); CCD camera; SP resonance angle Θ_R .

After cooling to 4K a 10 to 20 monolayer (ml) thick Cs film is evaporated onto the Ag film. A shutter is used to control the evaporation process out of the Cs dispenser. A capillary above the Cs surface allows us to supply liquid helium after its thermalisation with the bath onto the substrate.

3. RESULTS

We have prepared several Cs surfaces (which will be called A, B, and C). After evaporating the Cs the temperature is lowered and then standard ^4He gas is supplied to the cell through the capillary. The gas condenses in the capillary and from its end droplets fall onto the substrate.

In one experimental run we have evaporated a homogeneous Cs layer (surface A) of about 15ml with a variation in thickness of $\pm 1\text{ml}$, averaged over an area of $\approx 20\mu\text{m}^2$. After cooling the cell to 1.3K we started to condense the He. The first drop partly covered the initially dry surface and formed the metastable thick film leaving a sharp contact line. This line is very stable ($> 30\text{min}$). The contact line only moves when a new He drop spills over this line. However, if a He drop falls on the wetted part then only the film thickness varies but not the contact line. Another possibility to keep the Cs surface wet at $T < T_w$ is by cooling from $T > T_w$, when again a thick He film remains on the surface. Some part of the Cs surface is covered

Fig. 2. (a) shows the SP signal of a Cs surface wetted by a 20nm thick He film at $T < T_w$ before a laser was focused onto it. The square and circle indicate the positions of the measurement of the film thickness depicted in the graph below. (b) shows the hole burnt into the He film (dark area). (c) represents time evolution of the film thickness (proportional to the ATR light intensity). The hole is stable for the whole observation time (> 30 min).

with a thick metastable ^4He film even far below T_w . We do not observe any dewetting of this film within our experimental time of several hours.

We have tried to artificially nucleate a hole in this metastable film by focusing a pulsed laser beam onto an area of about 3mm^2 . As seen in Fig. 2 a dry spot is created which remains free of the thick film surrounding the spot for hours. Hence the contact line does not move, i.e. the size of the dry spot remains constant, even if a He drop falls directly next the hole. If a drop hits the surface within the dry spot then only this surface remains wetted, the remaining area stays dry, i.e the size of the dry spot is reduced. In addition it is possible to give the hole a certain pattern by scanning the laser over the Cs.

This experiment was done in a glass cryostat where the temperature varied from 1.3K to 4K, due to transferring helium every 8th hour. As a consequence we observed that the Cs surface deteriorated after 2 days and the resonance angle and resonance width of the surface plasmons reduced to a value similar to bare Ag with some dielectric (but no additional metal) on the glass prism. Hence also the nonwetting behavior disappeared.

Fig. 3. Photocurrent spectras at different locations on Cs (surface C).

We then evaporated again Cs (surface B) onto the initial surface without opening the cell and without increasing the temperature above 80K. This time only one part of the new Cs surface showed the typical nonwetting behavior, whereas the other part (wetted) was always covered with a thick ^4He film. From the measured Θ_R , however, one would have expected a nearly homogeneous Cs surface. It was not possible to nucleate any stable dry spot in the wetted part. The nonwetted part of the substrate showed a strong film thickness hysteresis. We also observed that the surface was covered with some particles (perhaps ice or dust) which caused pinning centers for the thick He film.

In contrast to the finding above we observed in another experiment a *spontaneous* dewetting of the thick film into the equilibrium thin film state without any external disturbances at a temperature of 1.4K. When warming this substrate from the dry state the thick film covered again spontaneously the surface at 2.07K, which represents the wetting temperature of this particular Cs surface. So this particular Cs surface, although of the same thickness as before, showed stronger nonwetting behavior. To characterize the Cs surface due to chemical impurities, it is possible to do local measurements of the work function, Φ . Therefore we have measured the current of the photoelectrons from our Cs surface as a function of the cut-off light-wavelength. For surface A we determined $\Phi = (1.9 \pm 0.1)$ eV, as a value for the whole surface. For a third Cs surface (surface C) we determined $\Phi = (1.8 \pm 0.05)$ eV, see Fig. 3. This value is identical to what is found for pure Cs.⁸ However, we also observed that this Cs showed areas with a slightly higher work function when scanning the surface, although these areas seemed to be wetted at all temperatures. One possible explanation is that these parts of the surface were very rough which enhances wetting.

When warming the experimental cell we observed no significant change in Φ until $T \geq 30\text{K}$. Between 35K and 40K Φ decreased to values around 1.5eV, significantly lower than for pure Cs. For such a Cs it is expected to have a much more pronounced nonwetting behavior with a wetting temperature above T_λ .⁹ In this case also the wetting dynamics should resemble a rich behavior, close to T_w the metastable film would be in its normal state and comparable to a classical liquid, lowering $T < T_\lambda$ the film would be in its superfluid state. Experiments in this direction are under way.

4. CONCLUSIONS

We have observed an unexpected high stability of the metastable ^4He film against external disturbances on some Cs surface. On other Cs surfaces, however, a spontaneous disappearance of the thick metastable film below T_w was seen. Our results show that the dynamics of ^4He films on cesiated surfaces are strongly dependent on the film quality. From these experiments a strong influence of the chemical state of the Cs and its surface morphology on the wetting behavior of liquid ^4He can be inferred. As a consequence these effects should also have an influence on the interface free energies of this system, important for essential wetting parameters like the contact angle. Therefore the simultaneous characterization of the Cs surface is essential for further wetting studies on this otherwise ideal system. In addition, measuring the photo current versus the adsorbed film thickness should provide information about the equilibrium 'thin film' state.¹⁰

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