

Characterization of Quench-Condensed Cesium on a Nanometer Scale at Low Temperatures

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The alkali metals Cs and Rb are the only surfaces which are not wetted by liquid ^4He below a certain temperature. Especially for the system ^4He -Cs this has been observed many times. However, there exist discrepancies in the measured contact angles and accompanying hysteresis, and in the movement of helium contact lines. Surface roughness is expected to have a significant influence on the wetting properties, but the working mechanism is not clearly understood yet. For this reason we have developed a special low temperature setup, which allows simultaneously an in situ evaporation of the alkali metals and an investigation of the substrate surface using a scanning tunneling microscope (STM). A characterization of quench-condensed Cs substrates is given and a manipulation of their surfaces on the nanometer scale is demonstrated. These investigations contain the first STM images of such a Cs surface at low temperatures.

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1. INTRODUCTION

The wetting of alkali metal substrates by superfluid ^4He has 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], where T_w is the wetting temperature. For liquid ^4He on Rb a non-wet state has been discovered for temperatures less than 0.3 K [5].

It is also observed that differently prepared Cs surfaces result in a quantitatively different wetting behaviour of liquid ^4He . E.g., a low temperature quench-condensed Cs shows a more pronounced hysteresis in contact angle than Cs grown from the liquid state. A possible interpretation is, that the first method causes a rough Cs surface, whereas the latter should create a

smooth one [6,7]. However, *in situ* measurements of the topography of the various Cs surfaces are still missing.

In this paper we present first results of a nanometer scale surface characterization of *in situ* prepared Cs substrates at liquid helium temperatures. Such surfaces are commonly used for studying the non-wetting behaviour of liquid ^4He . For these studies we constructed an experimental setup which allows simultaneously the *in situ* preparation of alkali metal substrates and the use of a scanning tunneling microscope (STM) for surface characterization, described in section 2. In section 3 calibration measurements of our STM are shown and in section 4 the process of the cesium evaporation and its characterization are demonstrated. In section 5, we present for the first time a direct manipulation of the cesium surface via the STM.

2. EXPERIMENTAL SETUP

We have constructed a low temperature setup to investigate Cs surfaces which mainly consists of three different parts: the STM, a sample holder with positioning system, and an alkali metal dispenser. The main part of the STM is a 1/2" piezotube. On one hand this piezo actuator provides the scanning motion; on the other hand, utilizing a slip-stick technique for the tunneling tip holder, it works as a linear motor which is responsible for the coarse approach [8]. Using this kind of approach system, the resonance frequency f_{res} of the STM-setup could be kept very high which results in a good stability to external vibrations. For this setup f_{res} is around 5 kHz, whereas the typical scanning frequency is a few Hz. The maximum scanning range of our tunneling microscope is about $1300 \times 1300 \text{ nm}^2$ at 300 K and about $330 \times 330 \text{ nm}^2$ at 1.3 K. The STM is mounted on a piezo translation stage (see figure 1).

The second part of our setup is a metal sample holder which is also mounted on a translation stage. There is access to any position on the sample within a maximum area of $1.2 \times 1.2 \text{ cm}^2$, using both stages at the same time. The substrate itself consists of several HOPG (*highly oriented pyrolytic graphite*) or mica plates, which are tightly mounted on the sample holder. This attachment provides a good thermal conductivity and thus a very small temperature gradient between the substrate, the slip-stick table, and the heater, which is useful for cleaning purposes (see section 3). For future measurements, the metal sample holder may be replaced by a glass prism allowing measurements of the thickness of adsorbed quantum films using the surface plasmon spectroscopy technique [9].

As an additional feature, the resistance over one of these mica plates

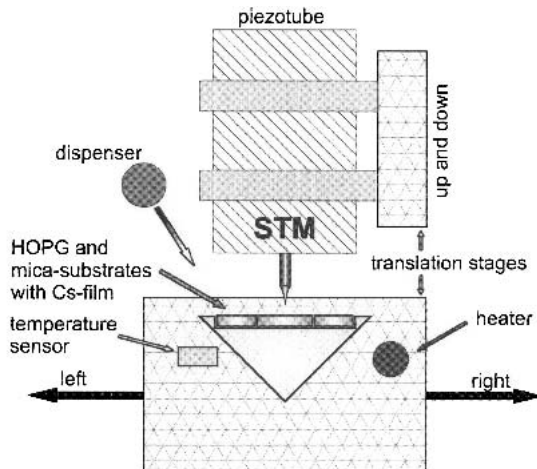


Fig. 1. Experimental setup (topview). Using the translation stages, the STM can be moved 1.2 cm in vertical and the substrate 1.2 cm in horizontal direction. The cesium dispenser is positioned left of the piezotube. The picture shows three different substrates attached onto the sample holder.

mounted on the sample holder can be measured during the evaporation process. The decrease in the resistance due to the growth of the alkali metal film on the mica plate allows the estimation of its thickness, although an accurate calibration has still to be supplied.

The third part of our setup is the alkali metal dispenser which is mounted close to the piezotube. The alkali metals (Cs, Rb, etc.) can be evaporated onto the substrate by simply applying a heating current I_{evap} to the dispenser. Typical values for I_{evap} are in the range of 6.5 to 7.5 A which leads to an evaporation rate of about 0.3 to 0.5 monolayers per second. Lower evaporation values can be achieved by decreasing I_{evap} . Figure 1 shows the setup schematically.

3. CALIBRATION MEASUREMENTS

For calibration of our STM in vertical direction we used atomic steps on a gold (111)-oriented surface, and a periodically structured surface for this purpose in lateral direction, respectively. These measurements are done both at room temperature and at low temperature to take into account the change in piezomechanical constant, which describes the correlation between

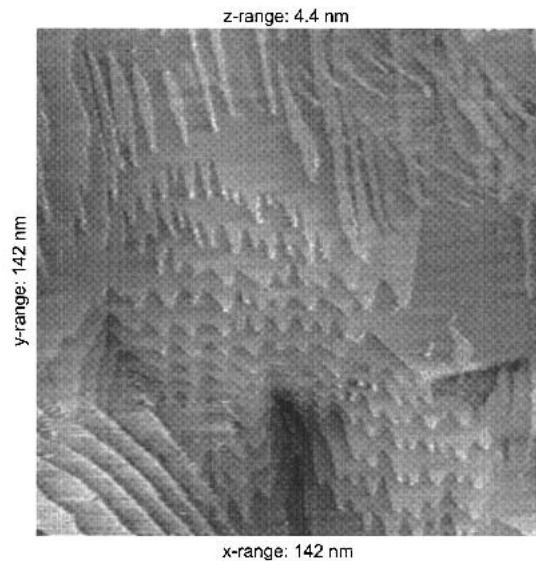


Fig. 2. Atomic steps on an annealed gold surface. The step height is 2.35 \AA as described in literature [12]. The thickness of the gold layer is about 100 nm . The scansize is $142 \times 142 \text{ nm}^2$, the z -range is 4.4 nm . $I_{\text{tun}} = 2 \text{ nA}$, $U_{\text{bias}} = 120 \text{ mV}$, $T = 300 \text{ K}$.

the applied voltage and the resulting deformation of the piezotube.

Figure 2 shows an image of atomic steps on a gold surface at room temperature. This kind of surface was prepared by slowly evaporating gold onto a cleaved mica plate, which after cleaving has been tempered for 24 hours at 400°C . The evaporation process itself has been done *ex situ* using a commercial vacuum evaporator. The epitaxial growth of the gold film was obtained using on one hand a slow evaporation rate ($\approx 3 \text{ \AA/s}$) and on the other continuously heating the mica substrate. The substrate temperature during evaporation was around 380°C . This procedure led to a monocrystalline, faceted surface on wide areas of the substrate, which is also reported many times in literature [10]. The crystal orientation of gold surfaces prepared under these conditions can be assumed to be (111) , like it has been proven using low-energy electron diffraction (LEED) techniques [11]. In addition, the single crystalline gold facets are stacked close packed which leads to a step height of 2.35 \AA [12]. These steps are typically used for the calibration of STM and AFM microscopes [13].

After the substrate has been attached to the sample holder, it is heated

to about 80°C using a heater fixed onto the sample holder. Simultaneously the cell is pumped for 24 hours using a turbomolecular pump in order to get an adsorbent free, clean surface. At this temperature the pressure measured outside the cryostat (as close as possible to the cell) is less than 10^{-6} mbar. During the following cool down, the heater is used to maintain a temperature difference between the substrate and the rest of the cell of about 80 K. This technique avoids condensation of residual gases onto the surface of the substrate during this process. The cool-down itself takes about 2 hours from room temperature to liquid nitrogen temperature, and further 30 minutes to liquid helium temperature.

4. CESIUM EVAPORATION and CHARACTERIZATION

After reaching liquid helium temperature, cesium is evaporated onto mica and HOPG substrates. These substrates can be prepared almost ideally flat by cleaving and are thus quite suitable for surface characterization of adsorbed materials. The HOPG substrate was investigated at low temperatures before evaporation and appeared to be atomically flat, although lateral atomical resolution was difficult to ensure due to external vibrations. *In situ* investigations of the mica substrate before Cs-evaporation are not available due to the lack of conductivity.

The evaporation time we typically use is varied between 5 and 20 minutes resulting in a film thickness of about 150 and 600 monolayers (ML), respectively. The evaporation current is linearly increased by 0.2 A/min until the final current of 7.5 A is reached. This is the standard procedure as it is recommended by the manufacturer of the dispenser [14]. However, there are much lower evaporation rates reported [15,16] which can be achieved by lowering the evaporation current and increasing the distance between substrate and dispenser. The surface characteristics of such thin cesium films will be examined in future experiments.

The change of resistance measured over the mica plate during the Cs evaporation is shown in figure 3. The right part shows a magnification of the first variation of the measured resistance at an evaporation time of about 32 minutes. The decrease proves the growth of a conducting cesium path (percolation), whereas the following increase can be interpreted as a reopening of the path due to diffusion of the cesium atoms. These oscillations in the resistance show that the deposited cesium atoms primarily form an island structure. This process takes place at a substrate temperature of about 30 K. A coalescence of evaporated Cs islands to a closed Cs film can be associated with the rapid decrease of the resistance to its thick-film

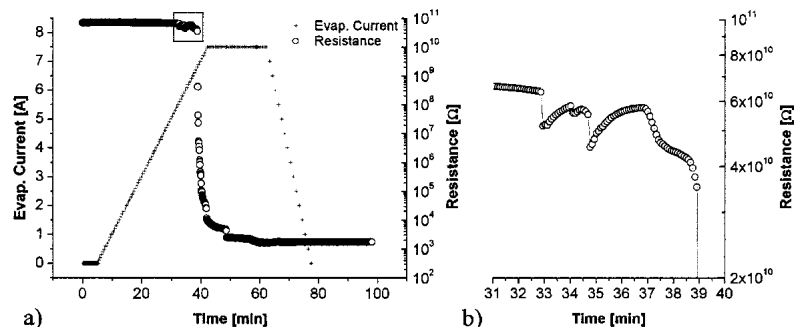


Fig. 3. a): Resistance measured over a bare mica plate during evaporation (\odot) versus evaporation current (+). b): Magnification of the marked region of interest (square in figure 3a).

value, which occurs at an evaporation time of about 39 min (see left part of figure 3). The finite resistance before cesium evaporation is due to leakage currents.

The surface topography of such a 600 ML thick quench-condensed cesium film as taken with our STM is shown in figure 4. Obviously the Cs surface shows a high roughness. The cesium islands have a relatively sharp size distribution, which is about 12 nm for their width and 5 nm for their height, respectively. As a comparison, gold evaporated at room temperature shows also a polycrystalline island structure with a similar height for the islands, but with a bigger average width (≈ 50 nm, [17]). A typical example of the Cs surface profile is shown in figure 5.

The thinner cesium film (150 ML) showed a reduced roughness with an average width of the islands of about 4 nm and a height of about 2 nm. Although other parameters like evaporation temperature and evaporation time may have influence on the shape of the film, the roughness seems to be dependent on the evaporated film thickness, which is planned to be investigated in future measurements.

Previous lateral resolved measurements, on a mesoscopic scale, of the surface work function show that cesium is evaporated with a high chemical purity [18]. This suggests, that the obtained images can be interpreted as topographical information of the cesium surface. However, measurements of the work function on a microscopic scale, using tunneling spectroscopy, still have to prove this assumption. The high values for the tunneling current stem from the low value of the work function of Cs ($\Phi_{\text{Cs}} \approx 1.8$ eV [18]). To achieve a common distance between the tip-apex and surface nuclei of a

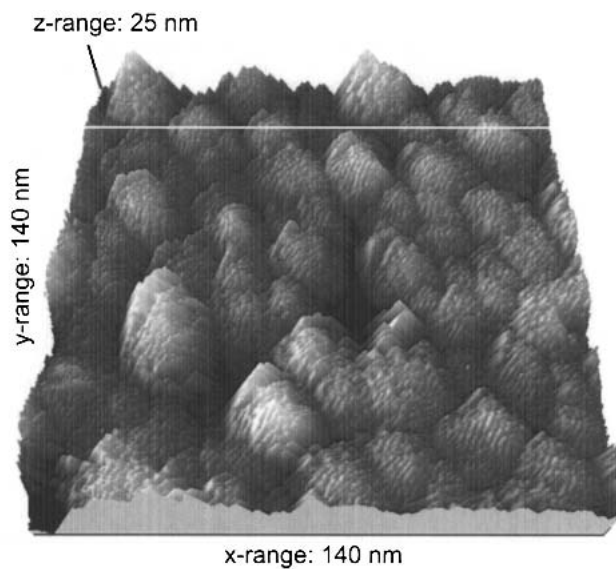


Fig. 4. Cs surface after quench condensation on HOPG. The film thickness is about 600 monolayers. The small ripples are scanning artefacts produced by external vibrations. The white line represents the linescan in figure 5. The scansize is $140 \times 140 \text{ nm}^2$, the vertical scale is 25 nm from minimum to maximum. $I_{\text{tun}} = 30 \text{ nA}$, $U_{\text{bias}} = 120 \text{ mV}$, $T = 4.2 \text{ K}$.

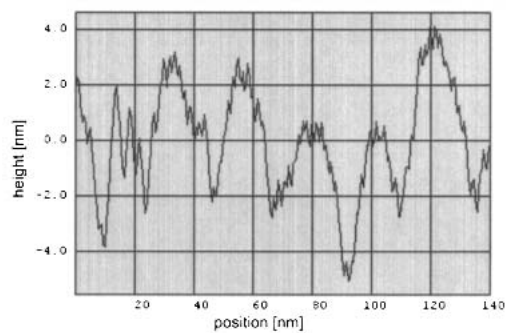


Fig. 5. A typical linescan in figure 4. The cesium islands have an average width of 11.5 nm and an average height of 5.3 nm. The periodicity is about 15.8 nm.

little less than 1 nm, the tunneling current on Cs has to be approximately one order of magnitude higher compared to that one on gold ($\Phi_{\text{Au}} = 5.31$ eV on a (111)-surface). This can be calculated using Binnig and Rohrer's expression [19] for the tunneling current density j_{tun} :

$$j_{\text{tun}} = \left(\frac{1.025e^2\Phi^{1/2}}{8\pi^2\hbar s} \right) U_{\text{bias}} \exp(-1.025\sqrt{\Phi}s) ,$$

where $\Phi = \frac{1}{2}(\Phi_{\text{tip}} + \Phi_{\text{sample}})$ is the average work function (in eV) of the tip and the sample, s the tunneling distance, and U_{bias} the applied bias voltage.

5. CESIUM SURFACE MANIPULATION

For future wetting experiments, an appropriate surface manipulation technique would be desirable. This would allow the investigation of surfaces with specific roughness in order to reproduce the different wetting behaviour as described in the introduction and to study the influence of roughness as pointed out in Refs. [6,7].

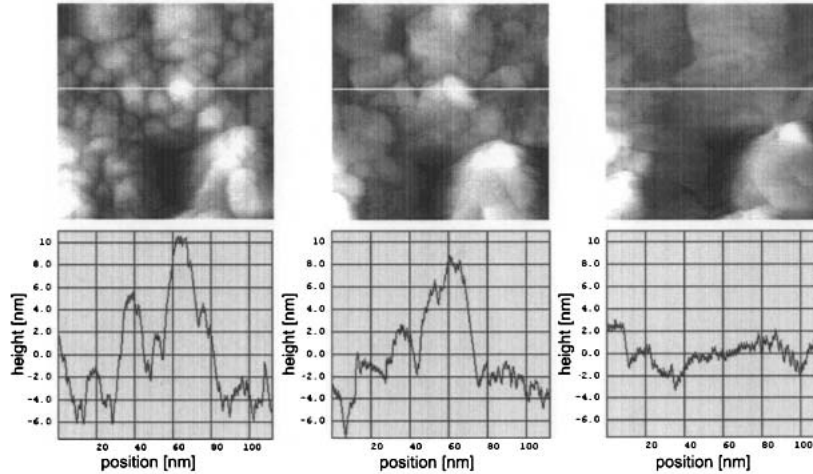


Fig. 6. Series of scans performed on a cesium surface evaporated on mica using a high tunneling current. The *rms*-roughness could be reduced by 35 %. All pictures are 112×112 nm² in size. $I_{\text{tun}} = 53$ nA, $U_{\text{bias}} = 120$ mV, $T = 4.2$ K.

Experience has been gained in our group regarding thermal annealing

on silicon using a ruby laser [20]. It would be conceivable using a similar technique for annealing a cesiated surface. In this way it should be possible to improve the surface by means of roughness reduction, which could be investigated using our experimental setup. This is planned for future experiments.

Additionally, it has turned out that surface manipulation on a nanometer scale can also be done by scanning tunneling microscopy using a very high tunneling current. Figure 6 shows a series of images taken on a cesiated mica plate. The surface roughness could be *reduced* by using a current of about 50 nA. The average root-mean-square (*rms*) roughness decreased to 65% of its initial value, see line scans in figure 6.

Whether this *nanomachining* technique is able to influence the macroscopic wetting properties is still an open question. Larger areas could be manipulated using the piezo slides and the STM simultaneously.

6. CONCLUSIONS

A low temperature setup has been developed, which allows an *in situ* evaporation and investigation of quench-condensed alkali metal films using a scanning tunneling microscope. Cesium films of different thickness have been evaporated at 4 K on mica and HOPG substrates. As expected, the cesium films (150 ML and 600 ML) show a high roughness. The appearance is qualitatively similar to a polycrystalline gold film, but the typical parameters like island size and height are different.

These thick metallic films have been useful for first scanning probe purposes. However, since many experiments so far have been done on quench-condensed alkali metal films of thicknesses of only a few ten monolayers, we are also aiming for thinner films. An investigation of surface properties of those thin films and its dependence on film thickness will be presented elsewhere. For future experiments, the combined possibilities of the setup will be used in order to investigate surface roughness and wetting properties simultaneously. For comparison, it would be very informative to investigate cesium surfaces grown from the liquid state [4], where a smoother surface is expected [6]. This would be a big advance to complete our understanding of the various results obtained so far on the wetting properties of cesium metal surfaces by liquid helium.

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