

# On the Structure of Thin $^4\text{He}$ Films on Solid Hydrogen

F. Mugele, U. Albrecht, and P. Leiderer

Fakultät für Physik, Universität Konstanz, 78434 Konstanz, Germany

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*We have investigated the adsorption of  $^4\text{He}$  onto a quench-condensed  $\text{H}_2$  film by means of surface state electrons. Oscillations in the surface state electron conductivity clearly reveal the layerwise character of the adsorption. At temperatures below 2K we have observed an anomaly in the conductivity around the completion of the first monolayer. This new feature is interpreted as an indication of a liquid-to-solid phase transition in the first layer.*

## 1. INTRODUCTION

Wetting phenomena in physisorbed films are a topic of considerable current interest. The degree of wetting is essentially determined by the interaction adsorbate/substrate in comparison to adsorbate/adsorbate. In this respect He films due to their weak interaction are of particular interest, and a considerable amount of theoretical and experimental work has hence been dedicated to this system.<sup>1,2</sup> Whereas most substrates give rise to complete wetting of He films, it has been shown in the last two years that some alkali metals are not wetted by He at low temperature.

Related is the fact that on most substrates the first one or two layers of a He film solidify under the action of the van der Waals forces.<sup>3,4</sup> Only for very "weak" substrates it is expected that also the first layer remains liquid. An intriguing case in this respect is molecular hydrogen as a substrate, because according to recent calculations<sup>4,5</sup> the first He layer on  $\text{H}_2$  is at or close to the borderline between a liquid and a solid state. Experimentally, however, the situation has been open so far.

In this letter we report on an investigation of  $^4\text{He}$  films on solid  $\text{H}_2$  by means of mobility measurements of surface state electrons (SSE). Our results show that upon completion of the first layer an anomaly in the SSE

mobility is observed, which we interpret as a signature that a phase transition from the liquid to the solid state does actually occur for the  ${}^4\text{He}/\text{H}_2$  system.

## 2. EXPERIMENTAL

A few systems with extremely low polarizability lend themselves to trap electrons in a state above the surface of the material.<sup>6</sup> For  ${}^4\text{He}$  and  $\text{H}_2$ , e.g., typical distances of the SSE are 78 Å and 17 Å, respectively, with binding energies in the ground state of 7.3 and 158 K. In this state the electrons are localized perpendicular to the surface, but are essentially free to move parallel to it. Various experiments have been carried out to obtain information about the 2-dimensional electron system thus formed (e.g. Wigner crystallization),<sup>7</sup> and the layering of He films on solid  $\text{H}_2$ .<sup>8,9</sup>

The experimental setup has been described in detail elsewhere.<sup>10</sup> It consisted of a closed sample cell in a  ${}^4\text{He}$  bath cryostat, containing two circular concentric metal electrodes covered by a glass plate. The  $\text{H}_2$  substrate in the present experiment was a thin film (thickness  $\approx 3 \mu\text{m}$ ) prepared by quench-condensation onto the glass plate at a temperature of about 1.8 K. After about 3 h of growth, the film was charged with electrons (density  $n_e = 3.4 \times 10^8 \text{ cm}^{-2}$  in the experiments discussed here) from a tungsten filament. Then we applied an AC voltage (30 mV, 50 kHz here) to the outer electrode and measured the contribution of the SEE to the complex capacitive crosstalk to the inner one.<sup>10</sup> This ‘‘Sommer-Tanner-technique’’<sup>11</sup> yields a value for the conductivity  $\sigma$  and for  $n_e$ . The initially low conductivity of the as-prepared film was improved by a proper annealing procedure.<sup>12</sup>

Finally, for each experimental run a fixed amount of He gas was admitted to the cell, a small fraction of which condensed on the  $\text{H}_2$  substrate as a physisorbed film with a (temperature-dependent) thickness  $d$ . Since the film was in equilibrium with the gas phase, its thickness was determined by the van der Waals interaction  $W(d)$  with the substrate and the chemical potential  $\mu_{\text{gas}}$  of the gas, according to

$$-\Delta\mu = k_{\text{B}} T \ln(p_{\text{sat}}/p) = W(d) \quad (1)$$

with  $\Delta\mu = \mu_{\text{gas}} - \mu_0$ . (Here  $\mu_0$  is the chemical potential of He at liquid-gas coexistence; in the following we will refer to  $-\Delta\mu$ , the deviation from coexistence, as the chemical potential.  $k_{\text{B}}$  is the Boltzmann constant,  $p$  the actual gas pressure and  $p_{\text{sat}}$  the saturated vapor pressure). In the thickness range where neither the short range repulsion of the substrate nor retardation effects are relevant,  $W(d)$  is simply given by

$$W(d) \cong \frac{\alpha}{d^3} \quad (2)$$

where  $\alpha$  is the van der Waals-Hamaker constant characteristic for the adsorbate/substrate combination. We note that theoretical studies mostly use the dispersion coefficient  $C_3$  of the van der Waals potential which is related to  $\alpha$  by

$$\alpha = \frac{C_3 - C_3^{\text{He}}}{d_0^3} \quad (3)$$

(Here  $C_3^{\text{He}} = 120 \text{ K}\text{\AA}^3$ , the interaction with a "He substrate," has to be subtracted.  $d_0 = \rho_{\text{bulk}}^{-1/3} = 3.58 \text{ \AA}^3$  is the thickness of one He layer at bulk density.)

### 3. RESULTS AND DISCUSSION

All the results presented here were obtained with one single  $H_2$  substrate, for which the SSE mobility  $\sigma/n_e$  had a rather high value of  $0.6 \text{ cm}^2/\text{Vs}$  on the bare hydrogen surface.

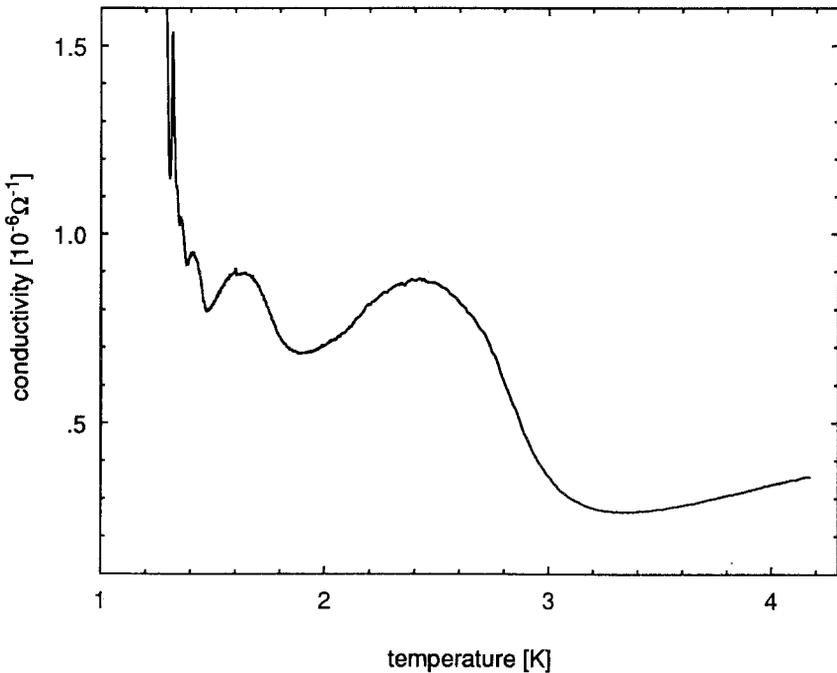


Fig. 1. Conductivity of SSE vs. temperature at a He gas density  $n_{\text{He}} = 0.9 \times 10^{19} \text{ cm}^{-3}$ . The maxima at 2.4, 1.65, 1.4 and 1.35 K, respectively, are assigned to the completion of the first to fourth monolayer.

An example of the conductivity as a function of temperature is shown in Fig. 1 for a He gas density of  $0.9 \times 10^{19} \text{ cm}^{-3}$ .<sup>(1)</sup> Starting at 4.2 K, where the He film thickness is below one monolayer,  $\sigma$  displays several pronounced oscillations as the film thickness increases upon lowering the temperature.<sup>(2)</sup> So far, more than one of these oscillations had never been observed on quench-condensed  $H_2$  films due to their roughness, but only on bulk  $H_2$  crystals carefully grown from the liquid phase.<sup>8,9</sup> They are a signature of layering since the SSE mobility is high on completely filled layers, whereas it is reduced for partially filled layers due to the enhanced scattering from thermally excited density fluctuations at the surface. The observation of layering indicates the high quality and smoothness of our  $H_2$  substrate.

The positions of the conductivity maxima indicate the completion of subsequent monolayers (see<sup>8,9</sup> and the discussion further below). Using Eqs. (1) and (2) we derive a value of  $\alpha = (21.0 \pm 2.2) \text{ K}$  from the positions of the second up to the fourth maximum. (The error bars quoted here and subsequently are statistical errors from several curves.) The chemical potential at the completion of the first monolayer is determined to be  $(7.8 \pm 0.3) \text{ K}$ , significantly less than extrapolated from the van der Waals relation. This reduction is due to the repulsive short range part of the substrate/adsorbate potential which mainly the first layer experiences.

The results agree very well with earlier measurements by various groups.<sup>8,9,14</sup> The experiments of Shirron and Mochel<sup>15</sup> also yield a value of  $\alpha = 21 \text{ K (layer)}^3$ , if the data analysis is corrected as discussed in the recent theoretical study by Cheng *et al.*<sup>16</sup> of several third sound measurements. On the other hand, Cheng *et al.* have also shown that all experimental results tend to overestimate the value of  $\alpha$  (by about 20%) due to neglecting the short range substrate/adsorbate repulsion for the second and subsequent monolayers. This accounts for our data, too. However, there is still a significant discrepancy between experiments and theory<sup>4,17,18</sup> which predicts  $\alpha = 5 \text{ K (layer)}^3$  or  $10 \text{ K (layer)}^3$ , the latter value being obtained when surface phonons are taken into account.

In the following we concentrate on an experimental feature which occurs at around the completion of the first monolayer, provided the temperature is below 2 K (see Fig. 2). In this case a small additional peak is

<sup>1</sup>The reduction of the gas density due to condensation of gas atoms in the film can be neglected here.

<sup>2</sup>The steep increase of the conductivity at the low temperature end of the curve corresponds to bulk condensation of liquid He. The origin of the very sharp peak in this curve, here appearing at 1.32 K, has already been discussed in the context of an earlier measurement (Ref. 13).

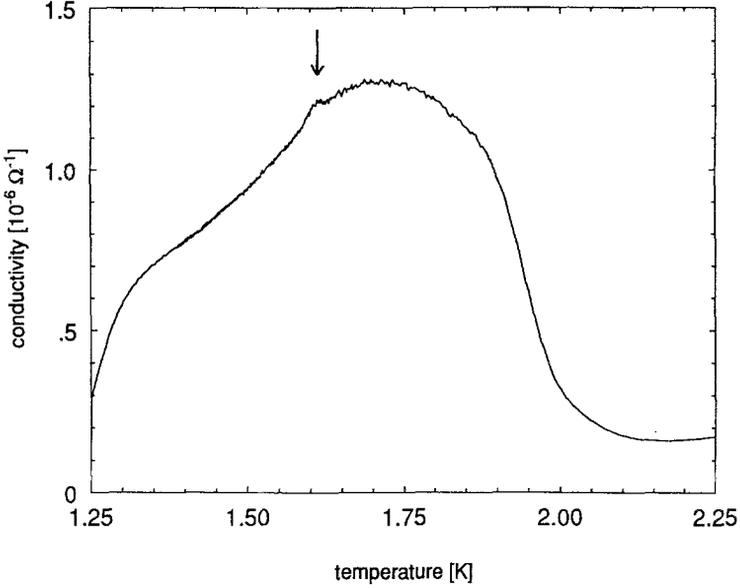


Fig. 2. Conductivity maximum corresponding to the completion of the first monolayer at low He gas density ( $n_{\text{He}} = 4.4 \times 10^{17} \text{ cm}^{-3}$ ). Amplitude and shape of the curve differ from Fig. 1. Note the additional feature marked by the arrow.

observed on the low temperature side of the main maximum, i.e. at a coverage somewhat above one monolayer.

At first sight, one might think of a superfluid transition as an explanation for this anomaly. Assuming a He coverage  $\rho_{\text{total}}^{2\text{D}} = 1.1 \text{ layers} \times \rho_{\text{bulk}}^{2/3} = 0.086 \text{ \AA}^{-2}$  at the transition, one can derive from the Kosterlitz-Thouless criterion

$$\frac{\rho_s(T_{\text{KT}})}{T_{\text{KT}}} = \frac{2}{\pi} k_{\text{B}} \frac{m_4}{\hbar^2} = 0.053 \text{ atoms/\AA}^2\text{K} \quad (4)$$

a critical temperature  $T_{\text{KT}} = 1.62 \text{ K}$  which is remarkably close to the observed value  $T_c = 1.61 \text{ K}$ . (Here  $\rho_s(T_{\text{KT}})$  is the superfluid density at the Kosterlitz-Thouless transition temperature  $T_{\text{KT}}$  and  $m_4$  the mass of a He atom.) However,  $T_{\text{KT}}$  has to be corrected for thermal excitations in the film ( $\rho_s(T_{\text{KT}}) \approx 0.8\rho_s(0)^5$ ) as well as for the unknown reduction of the superfluid density due to the van der Waals interaction with the substrate ( $\rho_s(0) < \rho_{\text{total}}^{2\text{D}}(0)$ ). Furthermore, in none of the experiments carried out so far has superfluidity been observed to noticeably influence the SSE mobility. For example, the curve in Fig. 1 where the superfluid onset

should occur at some  $T_{KT}$  between 1.6 K and 2.17 K does not display any comparable structure. For these reasons a superfluid transition appears very unlikely as the origin of the anomaly.

We suggest here another explanation, for which we recall the—so far rather qualitative—interpretation of the SSE conductivity curves as a whole. In the coverage range below one monolayer the mobility is described as resulting from the scattering of the electrons from 2D-density fluctuations, which are the more pronounced the larger the 2D compressibility.<sup>8</sup> In this way one can understand the mobility drop upon adding He to the bare  $H_2$  surface. As soon as the first layer is completed, it is close-packed. Hence its compressibility is small again and the mobility approximately reaches the starting value on the bare substrate. As the film gets thicker than one layer this picture remains qualitatively valid, due to the layerwise growth of the He film,<sup>8,9</sup> but now the density fluctuations are no longer those of a 2D fluid. Eventually, for very thick films the electron scattering from surface excitations can be described in terms of electron-ripplon scattering.<sup>19</sup>

Within this scheme the anomaly in the mobility can be understood in the following way: Since the value of the mobility around the shoulder is above the background given by the beginning of the build-up of the second layer, the compressibility in this region is obviously reduced. It is known from experiments<sup>3</sup> on graphite substrates as well as from calculations,<sup>20</sup> that the build-up of a second layer increases the density of the first one. This compression can induce a phase transition from liquid to solid in the first layer. Furthermore, registration of the first He layer on a  $H_2$  substrate, as suggested by Wagner and Ceperley,<sup>18</sup> may enhance the tendency to form a solid. Since the compressibility in the solid phase is reduced, this would lead to the observed increase in the SSE mobility. We note, however, that in the calculations of Wagner and Ceperley the first layer is always found to be liquid.

In order to check whether one is really dealing with a physical phenomenon, and to exclude experimental artefacts, we have repeated the experiment at different amounts of He gas in the cell. Results are presented in Fig. 3a: In all three examples the anomaly is clearly discernible. As expected, the transition shifts to lower temperature as the amount of He is decreased (like the main maximum). Even more revealing is a plot of the mobility as a function of the chemical potential (Fig. 3b). In all the cases the anomaly appears around  $-\Delta\mu = (6.8 \pm 0.5)$  K. In addition, this figure also shows the mobility maximum corresponding to the completion of the first monolayer at  $-\Delta\mu = (7.8 \pm 0.3)$  K mentioned above.

We would like to add that an indication of the anomaly reported here can be distinguish already in the data of an earlier investigation of He films on  $H_2$ ,<sup>9,21</sup> although there the feature was not quite as pronounced as in the

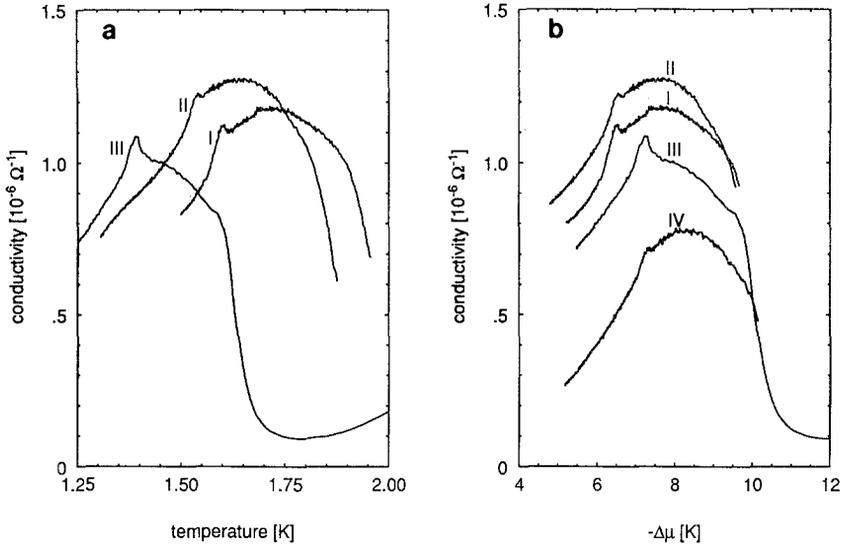


Fig. 3. Conductivity vs. temperature (a) and conductivity vs. chemical potential (b) around the completion of the first monolayer for different He densities:  $n_{\text{He}} = 5.9 \times 10^{17} \text{ cm}^{-3}$  (I),  $3.9 \times 10^{17} \text{ cm}^{-3}$  (II) and  $0.8 \times 10^{17} \text{ cm}^{-3}$  (III). In 3b also the data of Fig. 2 are included (IV), shifted for clarity by  $5 \times 10^{-7} \Omega^{-1}$ .

present work and therefore was not further taken into account. The value of the chemical potential  $-\Delta\mu$  at the transition derived from those data is also 7 K and thus agrees within experimental error with the one obtained here. The fact that a similar behavior was observed in a completely independent experiment corroborates that one is dealing with a real effect.

In summary, we have presented evidence for a phase transition in  ${}^4\text{He}$  films on solid  $H_2$  slightly beyond the completion of the first monolayer. As a possible interpretation it is suggested that a liquid-solid transition takes place in this layer, mediated by the pressure of atoms in the second layer. Since the SSE used as a probe here only provide sensitive information about the scattering by the film, but not about its possibly crystalline structure, other transitions, like a growth instability of the film connected with the formation of 2-dimensional liquid clusters,<sup>20</sup> cannot be ruled out at present. Additional characterization of the system with complementary techniques therefore appears highly desirable.

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