

Investigations of Thin Helium Films with Surface-Bound Electrons

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We have measured the mobility of electrons above ^3He and ^4He films condensed on a solid hydrogen substrate. Oscillations in the mobility as a function of film thickness indicate substantial layerwise adsorption of the films up to 9 atomic layers. Superfluid and normal films show similar behavior.

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Thin films of liquid helium are among the most prominent systems for studying the behavior of matter between two and three dimensions. In the numerous investigations dealing with these films, experimental techniques like ellipsometry, third-sound propagation, specific-heat measurements, and neutron scattering have been applied. These studies have led to a good understanding of the properties of helium films, such as, the phase diagram in the monolayer and submonolayer range, the formation of 1 or 2 solid layers next to the substrate due to the van der Waals interaction, and the gradual appearance of the bulk excitations as the film thickness is increased.¹ On the basis of these studies it was expected that films with a thickness larger than about four or five atomic layers would have essentially the structure and the excitations of the bulk liquid. Here we present measurements which appear to be extremely sensitive to the state of the film surface, and which show that the influence of a solid wall on the structure of the film extends out to at least nine layers at low enough temperatures.

Our method is based on the measurement of the mobility of a sheet of electrons floating on top of the helium film. Solid hydrogen was used as a substrate for the film. The only bare substrates which support an electron layer are hydrogen and neon. Our measurements were performed with a setup using the Sommer technique² similar to earlier experiments.³ Three capacitor segments, evaporated on a thin glass platelet, were covered with a 1-mm-thick layer of solid H_2 which was carefully grown from the liquid phase (see Fig. 1). On this substrate a sheet of electrons was produced from a discharge in the H_2 vapor while the capacitor plates were biased with a positive voltage of a few hundred volts. The electron densities were in the range from 10^7 to 10^9 cm^{-2} . When a small ac voltage, superimposed on the bias, is applied to one of the segments, the electron density is locally modulated, giving rise to a pickup signal on another segment. The amplitude and phase of this signal are determined by the number of electrons in the cell and by their mobility.²

With no helium in the cell the electrons float above the bare H_2 surface at a distance of about 25 \AA depending on the external holding field. As a He film is condensed

on the hydrogen, the electrons are "lifted off" from the solid.⁴ Their distance from the liquid surface increases with film thickness because of the smaller dielectric constant of liquid He compared to H_2 .

When He is added to the surface, one might naively expect the mobility of the electrons to increase, since the helium atoms could smooth out defects in the H_2 surface. However, Paalanen and Iye⁵ found that the addition of a small amount of He, on the contrary, always reduces the mobility. This was interpreted as additional scattering of the electrons from two-dimensional density fluctuations of the He layer. Interestingly, the reduction in the mobility was not a monotonic function of the coverage, but displayed pronounced oscillations. It was suggested that these oscillations reflect the atomic-layer structure of the film, and that mobility maxima (minima) occur whenever the uppermost monolayer is completely (half) filled. Although this interpretation is appealing, it is expected to break down for higher coverages, because for a large thickness well defined layers are difficult to imagine in the liquid state. Indeed, in the experiment of Paalanen and Iye the amplitude of the mobility oscillations dropped rapidly with coverage, and beyond four atomic layers no oscillations could be seen.

Our study of these oscillations extends to somewhat lower temperatures than in Ref. 5, a detail which proved

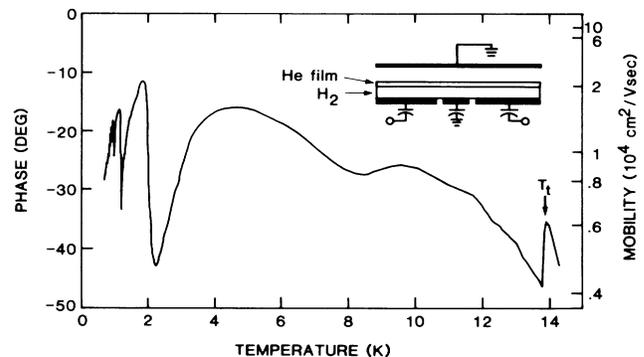


FIG. 1. Phase shift for electrons on solid H_2 covered by a He film with a temperature-dependent thickness. The He gas pressure was 0.35 mbar at 5 K. The electron mobility is given on the right-hand scale. Inset: Sketch of the sample cell.

to be important. Instead of our taking data at constant temperature, T , and varying the film thickness, n , it was preferable in our case to choose a thermodynamic path where the mobility, μ , is determined as a function of T at fixed amounts of helium in the sample cell. Here n is the film thickness in units of layers. This path, although somewhat more complicated to analyze because T and n vary simultaneously, has the advantage that the film thickness is always close to its equilibrium value, and that the deterioration of the surface resulting from the gradual condensation of minute traces of impurities in the admitted gas is reduced.

Figure 1 shows an example of the phase shift ϕ which, to a good approximation, is related to the mobility by $\tan\phi \propto \mu^{-1}$ over the temperature range investigated. For the amount of He used in this run, corresponding to a film thickness of ten layers at $T=0$, a film starts to condense below about 5 K. Therefore, the structure at the higher temperatures is due to the H_2 substrate. Starting at temperatures above the triple point at $T_t=13.8$ K the substrate is liquid, and ϕ drops as T is lowered because electron scattering from gas atoms decreases. The known value of $\mu=6 \times 10^3$ cm²/V·s on liquid H_2 at T_t ^{6,7} can be used to add an absolute mobility scale to Fig. 1. The jump in ϕ at T_t signals additional scattering from surface defects which appear as the substrate solidifies. On further lowering of T , ϕ gradually decreases as the gas-atom scattering vanishes. Around 8 K we frequently observe a dip in the mobility, which in its detailed structure appears to be sample dependent and which can possibly be attributed to surface melting or roughening.⁸ Eventually, at the lowest temperature pronounced oscillations in the mobility develop as the He film condenses onto the substrate.

These data are plotted on an expanded scale in Fig. 2(a). The arrows indicate those values of T where, according to the relation

$$n^3 = \alpha/T \ln |p/p_{\text{sat}}|, \quad (1)$$

completed atomic layers are expected. Here $p_{\text{sat}}(T)$ is the saturated vapor pressure at the temperature T , $p(T)$ is the actual pressure in the cell, and α is the van der Waals constant, which for He on H_2 ^{5,9} is $\alpha=20 \pm 7$ K (layer)³. The pressure could not be measured reliably at low T , but was calculated from the known number of moles of He in the cell, with the amount condensed in the film taken into account. The cell surface area, which had to be assumed in this calculation in order to obtain consistency with the data, was considerably larger than the geometrical surface area of the cell, probably because the hydrogen did not form flat surfaces on all cell walls. However, this calculated value for the effective surface area was nearly constant from run to run, even if a new H_2 crystal was grown.

What is striking in Fig. 2(a) is the large number of oscillations in μ that can be resolved at low T when n be-

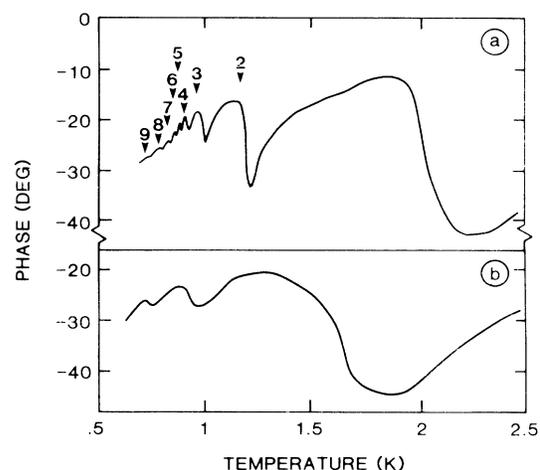


FIG. 2. (a) Data from Fig. 1 below 2.5 K drawn on an expanded scale. (b) Run similar to (a), but with a reduced amount of ^4He in the cell ($p=0.115$ mbar at 5 K).

comes large. Up to nine maxima were observed, which we take as an indication that the order induced in the He film by the solid H_2 wall extends over as much as nine atomic distances. The amplitude of the mobility oscillations decays nearly exponentially with a characteristic length ξ of 1.6–2.1 layers. That the oscillations are really related to the film thickness and are not due to some other temperature-dependent property of the 2D electron gas is evident from a comparison with runs with smaller amounts of He [Fig. 2(b)].

The modulation of μ , in particular at the larger values of n , becomes more pronounced at lower temperatures. For example, in order to observe the maximum corresponding to the completion of the fifth layer the temperature must be below 1 K. If a thickness of five layers is reached at higher T because of a larger amount of He in the cell, the structure is washed out by thermal excitations.

Electron scattering from the first and second helium layers, which are not superfluid, appears to be well understood on the basis of electron scattering from two-dimensional density fluctuations in the uppermost layer.⁵ In the case of a superfluid film the electrons must scatter from excitations in the superfluid phase. These excitations have traditionally been treated as quantized surface (third-sound) waves on an incompressible continuum superfluid. The coupling between electrons and third-sound excitations occurs because the electrons gain energy in the holding field by moving into the troughs of the waves.

To further clarify the scattering mechanisms, we investigated normal films of ^3He . The results, presented in Fig. 3, show that $\mu(T)$ for ^3He films displays qualitatively the same modulated structure as for ^4He . For the amount of ^3He in the cell, approximately 8 times that of

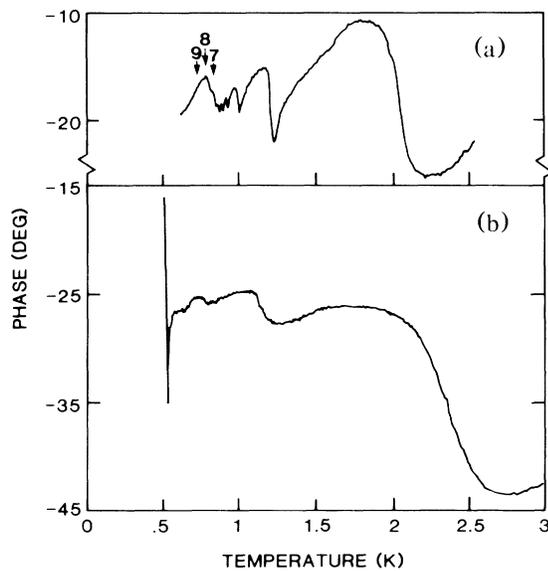


FIG. 3. Phase shift for (a) ^4He and (b) ^3He films. The hydrogen crystal was the same in both runs. The ^4He filling was 0.45 mbar and the ^3He filling was 3.5 mbar at 5 K. The peak in (a) for a thickness $n > 6$ layers, not present in the data of Fig. 2(a), was sample dependent. Superimposed on it are the mobility maxima 7 to 9.

^4He because of the higher vapor pressure of ^3He , the saturated vapor pressure is reached at about 0.55 K. The rapid drop and subsequent steep rise in the mobility at $T \sim 0.5$ K are a result of a rapid condensation of a thick film. When we compare the same temperature range, the structure is more affected by thermal broadening in ^3He , probably because of the smaller binding energy. From a comparison of Figs. 3(a) and 3(b) we conclude that excitations characteristic of only the superfluid state do not dominate the electron mobility of films.

The finite compressibility of the helium film with respect to coverage was neglected in the above discussion. We give here a qualitative argument¹⁰ which accounts for the shape of the mobility oscillations based on scattering from density fluctuations. It is known from adsorption isotherms of various gases on graphite that the film thickness does not vary smoothly as described by the van der Waals relation, Eq. (1); rather, stepwise growth occurs whenever the chemical potential $\phi = k_B T \ln(p/p_{\text{sat}})$ reaches the value characteristic for the respective layer. Figure 4(a) shows the variation of the ^4He coverage versus temperature expected on this basis under the experimental conditions specified for Fig. 2(a). The widths of the steps will be finite as a result of imperfections of the substrate, thermal excitations, and because along our thermodynamic path with a fixed amount of He in the cell the gas pressure drops noticeably during the building up of a new layer. In the region

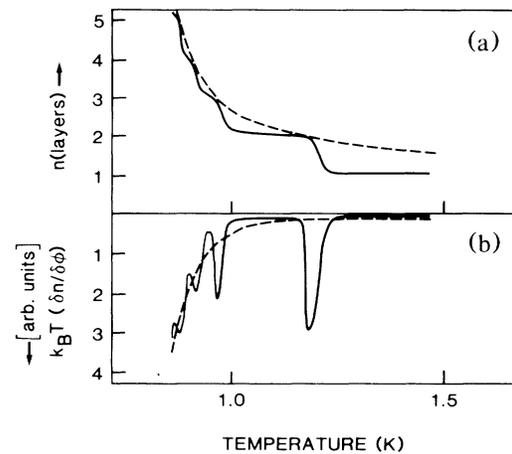


FIG. 4. (a) Schematic variation of the film thickness n and (b) the quantity $k_B T \partial n / \partial T$ derived from (a) vs temperature. The solid curves show the stepwise growth known from adsorption measurements on graphite (Thomy, Duval, and Regnier, Ref. 11); the dashed lines represent the thickness given by Eq. (1).

of a step, where dn/dT is large, the compressibility of the film is much higher than in the plateaus. Consequently, thermally excited fluctuations in the coverage will also be strongest in the step region. As pointed out by Paalanen and Iye,⁵ the scattering rate of electrons from two-dimensional density fluctuations in the uppermost monolayers should be given by $\tau^{-1} \propto k_B T \partial n / \partial \phi$. This quantity is sketched in Fig. 4(b). The qualitative similarity between Figs. 2(a) and 4(b) lends strong support to this interpretation.

Our data suggest that density fluctuations in the surface layer are excitations of both the normal and superfluid phases. Electrons at 1 K have thermal wavelengths of the order of 1000 Å and therefore probe surface density fluctuations on this scale. Fluctuations of a larger range may be influenced by surface irregularities. This may explain why surface electrons are a much more sensitive probe of the uppermost layer than third-sound experiments which probe density fluctuations on a centimeter scale.

Indications for the layering of the He films have also been found in specific-heat¹² and third-sound¹³ measurements of ^4He on graphite substrates. Up to seven layers could be resolved in specific-heat experiments, and two oscillations, corresponding to the fourth and fifth layer (second and third fluid layer) completion, were observed in the third-sound experiments. A theoretical study of thin helium films on various substrates by Krotscheck¹⁴ gave quite substantial layering as well. The decay length ξ found in computer simulations of a Lennard-Jones solid-liquid interface is of the same order as observed in our experiments.¹⁵

In summary, we have shown that surface electrons can

be used as an ultrasensitive probe for studying thin He films. The experimental results obtained here provide evidence that the layerwise adsorption of He films extends up to considerable thicknesses on a hydrogen substrate. Further theoretical efforts are required to describe quantitatively the interaction of the electrons with the film.

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⁷The absolute value of μ can, in principle, be determined from ϕ and the known cell geometry, charge density, and frequency. In our measurements ϕ was also influenced by the trapped charge on the glass plate underneath the H₂ substrate. This should not affect the scale in μ derived from the triple-point value or the results at lower T .

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