Rapid Communication

The Influence of Electrostatic Fields on Films of Liquid Helium

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Prompted by the recent striking experimental results reported by Babkin and Hakonen that appeared to show that liquid helium-II does not wet magnesium fluoride, we have examined the effects that an inhomogeneous electrostatic field has on thin films of liquid helium at temperatures below 0.5K. Our model includes the influence of gravity, surface tension, the electric field and the van der Waals interaction between the helium and its supporting substrate. We show that an inhomogeneous charge on the substrate can produce effects that mimic the surface profiles between wetted and non-wetted areas. The calculations also indicate that some special precautions may be necessary when studying films of liquid or solid helium on insulators.

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1. INTRODUCTION

At a recent workshop on wetting (Gif-sur-Yvette, March 1995) Babkin and Hakonen[1] presented some remarkable video recordings of experiments that seemed to show that liquid helium-II does not wet magnesium fluoride (MgF_2). Their observations, which we will refer to as the BH experiments, were on small cylindrical cells partially filled with liquid helium. The cells, which were used to study liquid-solid coexistence in ^4He, mixtures of ^3He–^4He and phase separation,[2] each comprised glass optical flats forming the top and bottom separated by a ~3mm high metal spacer ring of ~10mm diameter. Helium vapour was put into an initially empty and cold, T~0.5K, cell through a capillary that penetrated the wall of the metal ring. A video camera recorded interference patterns between light reflected from the top surface of the lower optical flat and from the top surface of the

condensed liquid helium. As the cell was filled it seemed as though some areas of the flat were not wetted by the liquid while films 5–10 μm thick grew on adjacent areas. The contact angles of the liquid helium at the edges of the main 'dry' area were small, but varied with position between 2 and 30 mrad. The configuration of dry and wetted areas was very stable and did not change over a period of a day. The phenomenon was not particularly temperature dependent below the lambda point T_λ but there was some evidence that it did not occur for $T > T_\lambda$ and it was suggested that this was due to the long time needed to reach equilibrium when the liquid was not superfluid.

Until recently it was believed that liquid helium wetted *everything* and, although it is now thought that caesium and perhaps rubidium are not wetted,[3,4] a demonstration that a commercial magnesium fluoride anti-reflection coating could resist wetting was astonishing; liquid helium would be expected to wet such a surface at all temperatures.[5]

Making one plausible conjecture but otherwise using only well-established properties of liquid helium, we propose an explanation for these striking results: Assuming that magnesium fluoride is wetted by liquid helium and in the dry areas the film is thinner than the optical resolution of the experiment, $\lambda/10 \sim 50$ nm, a possibility mentioned by Babkin and Hakonen at the workshop, we conjecture that the areas of thick film are associated with a static electric field that draws helium away from the thin regions where the field is smaller. It is unlikely that the static charge causing the fields is distributed completely uniformly over the substrate so variations in the film thickness and 'contact' angle, much like those seen in the experiments, are to be expected.

To explore the consequences of electrostatic charge in the cell we have calculated the surface profile of an idealised, but not unrealistic, case. We have not attempted to reproduce the detail of the BH experiments in this paper but instead will show how a very simple electric field pattern can reproduce their most striking
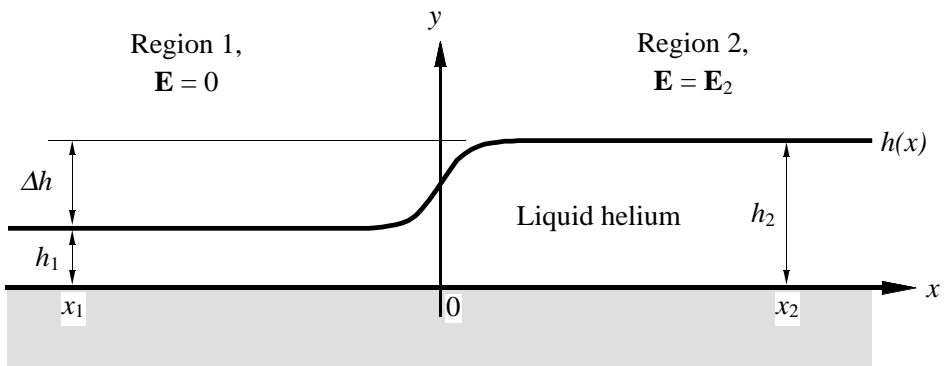

Fig. 1. The liquid helium film is supported by a solid substrate (shaded). In region 2 the electric field is $\mathbf{E}_0$ 0. (The x- and y-axes are drawn to very different scales.)

features. Even if our suggestion turns out not to be the explanation of the BH experiments, the results of the calculation are interesting because they show that experiments intended to study layers of helium, liquid or solid, on insulating substrates should incorporate checks to ensure that results are not affected by static electric fields.

2. FILM THICKNESS

Before analysing the detailed profile of the surface between two regions of differing electric field we can calculate the film thicknesses in regions of uniform electric field perpendicular to the substrate. Consider the situation sketched in Fig. 1 which shows film of helium supported by an insulating substrate and, on the left of the diagram, in region 1 the electric field $\mathbf{E}$ is $\mathbf{E}_1 = 0$. In region 2, on the right of the diagram there is a uniform field $\mathbf{E}_2 \neq 0$ with an abrupt jump in $\mathbf{E}$ between the two regions at $x=0$. Well away from $x=0$ the thickness of the helium is constant and represented by h_1 and h_2 in the two regions respectively. The difference in these two heights h has also been marked on the diagram because it will turn out to be a convenient order parameter. The relative permittivity χ of the liquid helium is[6]

$$\chi = 1 - \varepsilon_{\mathrm{r}} = 0.0572 \tag{1}$$

so we shall ignore the small effect of the polarised helium film on $\mathbf{E}$ until section 4. This is a useful approximation as it means that the results do not depend on the direction of the electric field, only its magnitude at the liquid surface. The situation we have in mind is at temperatures $T < 0.5\,\mathrm{K}$ so the vapour pressure is negligible.

The chemical potentials in the two regions must be equal so

$$\frac{-\gamma(h_1)}{h_1^3} + mgh_1 = \frac{-\gamma(h_2)}{h_2^3} + mgh_2 - \frac{m}{2\rho}\chi\varepsilon_0 E_2^2 = \Delta\mu \tag{2}$$

where $g = 9.81\,\mathrm{ms}^{-2}$, ε_0 is the permittivity of free space, $\rho = 145\,\mathrm{kgm}^{-3}$ is the liquid density,[7] m is the mass of the helium atom and

$$\gamma(h) = \gamma_0\left(1 + 1.64\left(h/d_{1/2}\right)^{1.4}\right)^{-1/1.4}. \tag{3}$$

The strength of the van de Waals interaction between the helium and the substrate[10] is governed by the empirical function $\gamma(h)$ suggested by Cheng and Cole[8] which, to a good approximation, is equal to the constant commonly referred to as ΔC_3 $\left(= C_3^{\mathrm{He-substrate}} - C_3^{\mathrm{He-He}}\right)$ in the literature for $h < 10\,\mathrm{nm}$ and is proportional to h^{-1} when $h > 100\,\mathrm{nm}$. Tabulated values of the parameters γ_0 and $d_{1/2}$ do not vary much between one fluoride material and another;[8,9] to represent the magnesium fluoride substrate of the BH experiment we have used the nominal values $d_{1/2} = 18\,\mathrm{nm}$ and

$\gamma_0 = 1.38 \times 10^{-50}\,\text{Jm}^3$ ($= 1000\,\text{K\AA}^3$) based on values for similar fluorides given in the literature.[9] The chemical potential of the film μ is less than that of bulk liquid μ_0 and

$$\Delta\mu = \mu - \mu_0 = k_{\text{B}} T \ln\left(\frac{P}{P_0}\right) \quad (4)$$

where P_0 and P are the vapour pressure above bulk liquid and a thin film respectively,[10] k_{B} is the Boltzmann constant and the cell is isothermal at temperature T.

Fig. 2 shows the solutions of Eq. (2) for a range of electrostatic fields and film thicknesses. The curves resemble the P–V isotherms of a gas–liquid system, the two phases being the thick (Δh 0) and thin ($\Delta h = 0$) films and the analogue of temperature is the reciprocal of the electric field. As h_1, the film thickness in the zero-field region, is increased an electric field greater than the critical value $E_{\text{c}} \sim 50\,\text{Vmm}^{-1}$ has almost no effect, *i.e.* $\Delta h \sim 0$, until a critical thickness h_{c} is reached. At this point h_2 increases very rapidly and the film thickens in the region of the field, eventually reaching a point when h becomes independent of further increases in h_1. For fields below E_{c} there is no sudden jump in h. It can be seen that height differences between the films of $\sim 10\,\mu\text{m}$ can be created by fields of only $\sim 200\,\text{Vmm}^{-1}$.
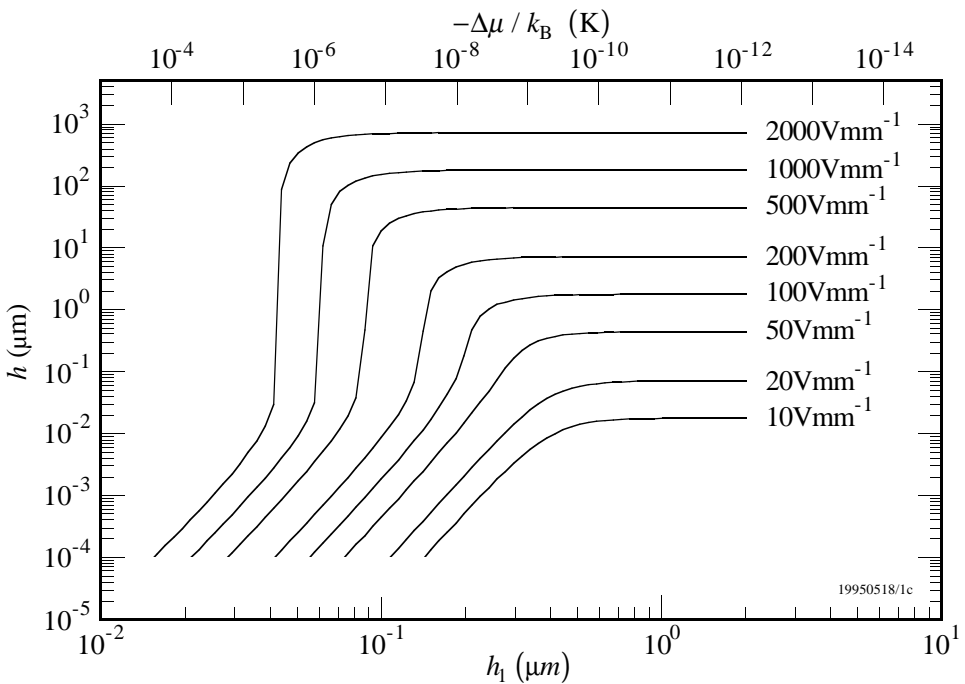

Fig. 2. Step height Δh in the surface profile due to fields of various strengths in region 2 as the thickness h_1 in region 1 (zero field) is varied.

Consider a cell where regions 1 and 2 of the substrate have areas A_1 and A_2 respectively and fields, as shown in Fig. 1, $E_1=0$ and $E_2>0$. The total volume of liquid is then

$$V = A_1 h_1 + A_2 h_2 \tag{5}$$

If a fixed volume of liquid helium is put into the cell then there are three types of behaviour. If the volume is small we are on the low Δh side of the near-vertical step in Fig. 2 and $\Delta h \sim 0$. If the volume is large then we are on the horizontal part of the curve then $\Delta h \gg 0$ and is independent of h_1. For intermediate volumes the system will be on the near vertical step so h_1 is essentially constant. The value of h_2 and hence the exact position on the step will be determined by the amount of liquid in the cell and Eq. (5). It is worth noting that, for a fixed amount of helium in the cell, at a given temperature the vapour pressure is a function of the electric field in region 2.

3. SURFACE PROFILE

To analyse the surface profile we will minimise the total energy. The grand potential $\delta\Phi$ of a thin column, base area δA, above the substrate at position x where the liquid height is h and the vapour pressure negligible is

$$\delta\Phi = \frac{\rho}{m} f_0 h \delta A + \tfrac{1}{2}\rho g h^2 \delta A + \sigma\left(1+\dot{h}^2\right)^{1/2} \delta A$$
$$- \tfrac{1}{2}\chi\varepsilon_0 E^2 h \delta A + \frac{\rho}{m}V(h)\delta A - \frac{\rho}{m}\mu h \delta A. \tag{6}$$

In the first term f_0 is the Helmholtz free energy per atom of bulk liquid helium. The next three terms are the gravitational, surface and electrostatic contributions to the potential. The dot denotes differentiation with respect to x. The liquid density is taken to be independent of the film thickness and the surface tension will also be assumed to be constant and equal to its low temperature value[11] $\sigma=3.54\times10^{-4}\,\mathrm{Jm}^{-2}$. The penultimate term is the van der Waals interaction between liquid and the substrate, obtained by integrating Eq. (3),

$$V(h) = \int_a^h \frac{-\gamma(y)}{y^3}\,\mathrm{d}y \tag{7}$$

where a is a small positive constant which is usually taken to be the equilibrium atom spacing of the adsorbate and the substrate. In the final term μ is the chemical potential per helium atom. When the system is in equilibrium its grand potential, which is the integral of Eq. (6) over the substrate plane, is minimised with respect to variations in the surface profile $h(x)$, *i.e.*
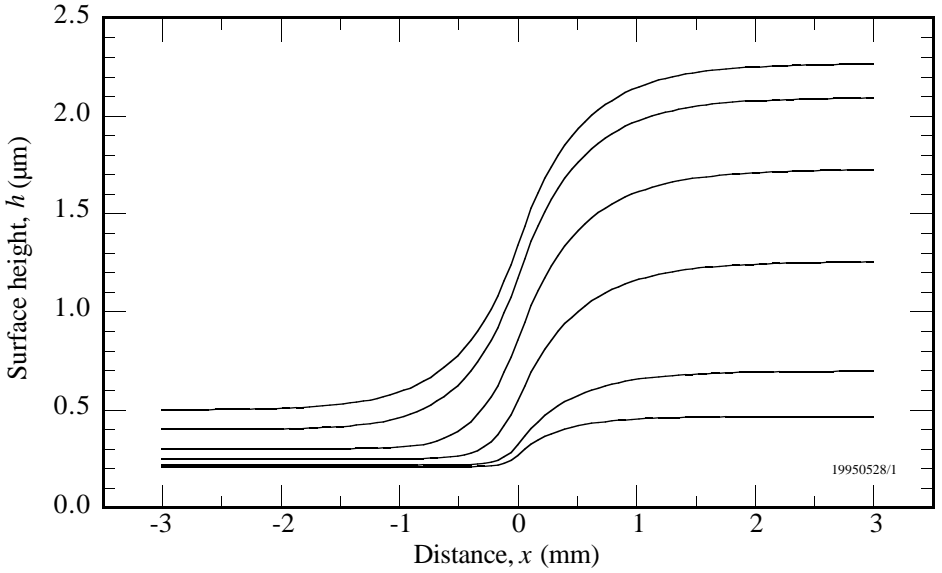
Fig. 3. Film profiles as more helium is put onto the cell when the field in region 2, *i.e.* $x>0$, is $100\,\text{Vmm}^{-1}$ and region 1, *i.e.* $x<0$, is field-free.

$$\delta \int \phi \, dx \, dz = 0 \quad \text{where} \quad \phi = \phi(x,h,\dot{h}) = \lim_{\delta A \to 0}\left(\frac{\delta \Phi}{\delta A}\right) \tag{8}$$

and this will be the case when $h(x)$ satisfies[12]

$$\frac{d}{dx}\left(\frac{\partial \phi}{\partial \dot{h}}\right) = \left(\frac{\partial \phi}{\partial h}\right) \tag{9}$$

which is a nonlinear ordinary second-order differential equation

$$\frac{m\sigma \ddot{h}}{\rho\left(1+\dot{h}^2\right)^{3/2}} - \left(mgh - \gamma(h)h^{-3} - \frac{m}{2\rho}\chi\varepsilon_0 E^2 - \Delta\mu\right) = 0 \tag{10}$$

and we have used the fact that f_0 differs negligibly from μ_0 at low pressures to write the expression in terms of $\Delta\mu$. Both $\dot{h}$ and $\ddot{h}$ are zero at the points where $h_1 = h(x_1)$ and $h_2 = h(x_2)$ are defined so these heights, which will be employed as boundary conditions when Eq. (9) is solved, can be found by satisfying

$$mg(h_2 - h_1) + \gamma(h_1)h_1^{-3} - \gamma(h_2)h_2^{-3} = \frac{m}{2\rho}\chi\varepsilon_0 E_2^2 \tag{11a}$$

the result that was anticipated in Eq. (2), or in terms of the order parameter h defined earlier

$$mg\Delta h + \frac{\gamma(h_1)}{h_1^3} - \frac{\gamma(h_1 + \Delta h)}{(h_1 + \Delta h)^3} = \frac{m}{2\rho}\chi\varepsilon_0 E_2^2. \tag{11b}$$

Eq. (10) was solved by iterating an equivalent finite-difference equation using h_1 and h_2 as boundary conditions. The grid spacing was reduced near the step at $x=0$ to handle the cases when the electric field was large. Fig. 3 shows the film profile changing as the amount of helium in the cell is increased while the field in region 2 fixed at $100\,\mathrm{Vmm}^{-1}$. Films thicker than about $0.3\,\mu\mathrm{m}$ are almost uninfluenced by the substrate their shape being governed by gravity but, as the amount of helium is reduced, the van der Waals term in the potential starts to dominate the behaviour and there is a noticeable change in the shape for the thinner films. The charateristic length of the transition between regions of different heights is approximately proportional to the surface tension and therefore relatively insensitive to temperature in the range interest.

Fig. 4 is included to make comparison with the BH experiments easier; the amount of helium in the model system has been adjusted so that the film thickness is always $5\,\mu\mathrm{m}$ in region 2 and each curve is the surface profile obtained for a different electric field. At very low fields the film in region 1 is relatively thick and the step

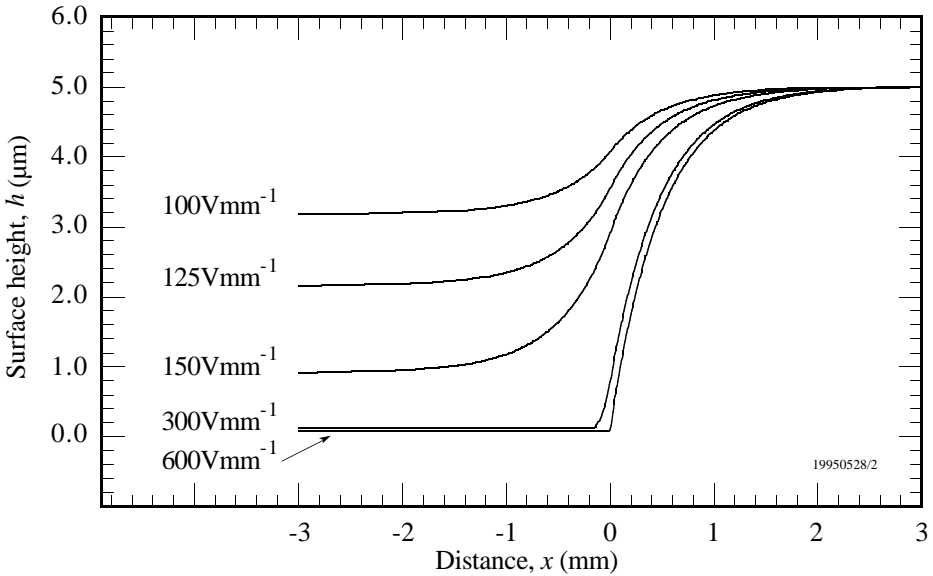Fig. 4. Film profiles when the field in region 1, *i.e.* $x<0$, is zero, but with various values of field in region 2, *i.e.* $x>0$. The amount of helium is adjusted to keep the film height in region 2 constant with $h_2=5\,\mu\mathrm{m}$. The surface heights in the thin region for the 300 and $600\,\mathrm{Vmm}^{-1}$ cases are $h_1=129$ and $85\,\mathrm{nm}$ respectively.

profile smooth. However, for electric fields above about $300\,\mathrm{Vmm^{-1}}$ the surface profile, with its well-defined contact angle, bears a striking resemblance to pre-wetting behaviour. Inside a charged insulating cell it is difficult to imagine that electrostaic field will be particularly uniform and Fig. 5 shows that even relatively small changes in the electrostatic field, can have quite substantial effects on the helium film thickness. There is nothing particularly critical about the fields chosen to prepare Fig. 5 – for example the $1250\,\mathrm{Vmm^{-1}}$ field in the central thin region does affect the film, but only very slightly; the thickness is increased by 10nm compared with 52nm when there is no field.

4. DISCUSSION AND CONCLUSIONS

It is well known that electric fields created with electrodes distort the free surface of liquid helium. However, it is surprising to find that fields of quite moderate strength, arising from free or induced static charge, such as might be created during assembly of the cell, can in principle affect the surface profiles in a way that mimics the behaviour of helium on a non-wetting substrate.

We have shown that between regions of differing electric field the surface profile of liquid helium can be very asymmetric with a rapid change of slope
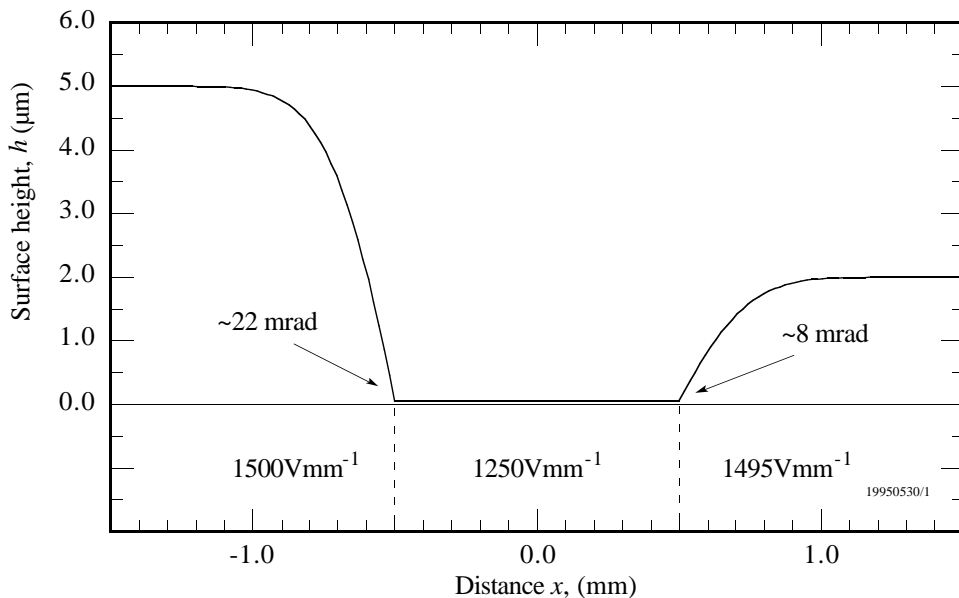

Fig. 5. A film profile shown in a region where the field changes abruptly, but by fairly small amounts. It can be seen that thick films of different heights and contact angles, can coexist with thin films under such circumstances.

between thin and thick film regions resembling the behaviour of a liquid on a non-wetted surface. Moreover, there can be a wide range of, albeit small, angles created by the fields. The range of possible contact angles and surface profiles could be increased by considering more complicated fields and surface charge densities than the simple cases we have discussed in this paper. We did not include the effect of the liquid helium on the **E**-field in our calculations but its influence would be to introduce a certain amount of hysteresis into sensitive situations like that depicted in Fig. 5 so we expect the detailed configuration of helium in the cell would be sensitive to the precise cell-filling conditions and not reproducible in detail between fills.

Although we believe that electrostatic fields could explain the results reported by BH, further experiments will be needed to establish whether this is the correct explanation. Whatever the outcome in this particular case, as much work on thin films of liquid helium involves insulating materials this work suggests that it would be prudent in future experiments to ensure that there are no unwanted electrostatic fields distorting the film thickness.

ACKNOWLEDGMENTS

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