Stability of Charged ^4He Films

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We have investigated the stability of electrons above saturated films of superfluid ^4He on both insulating and conducting substrates. Electron densities of up to 10^{11} cm^{-2} have been observed to be stable, nearly 2 orders of magnitude more than on bulk ^4He . Charging the films to such high densities leads to a drastic reduction in film thickness.

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Electrons on liquid helium have proven to be a quite suitable candidate for studying two-dimensional (2D) behavior: They form a particularly clean system characterized by simple forces, their interaction with the substrate is small, and their density—at least below a limit of $n_c = 2.4 \times 10^9 \text{ cm}^{-2}$ —can readily be varied.¹ A number of intriguing phenomena have been observed in this system, the most spectacular probably being the transition to a 2D electron solid.² In the experiments performed to date these electrons behave like a *classical* Coulomb system, because for $n < 2.4 \times 10^9 \text{ cm}^{-2}$ the Fermi energy is much smaller than the Coulomb energy.

It would be most interesting to move towards higher electron densities, where first quantum corrections in the phase transition to the Wigner solid are expected to appear, and at still larger n , when the Fermi energy dominates, melting of the Wigner lattice into a degenerate Fermi gas should occur.³ Unfortunately, on bulk liquid helium this regime is not accessible, because an electrohydrodynamic instability of the charged surface develops at the critical density n_c .⁴⁻⁶ In order to circumvent this restriction it has been suggested to use a thin helium film as a substrate rather than bulk liquid, because a film is additionally stabilized by van der Waals forces,¹ as is seen from the dispersion relation for ripples coupled to the 2D electron layer^{7,8}:

$$\omega^2 = \frac{\rho_s}{\rho} \left[\left(\frac{3\alpha}{\rho d^4} + g \right) k + \frac{\sigma}{\rho} k^3 - \frac{4\pi n^2 e^2}{\rho} k^2 F(k, \epsilon) \right] \tanh(kd). \quad (1)$$

The various terms originate from the van der Waals coupling α , the acceleration due to gravity g , the surface tension σ , and the charge density ne . (Here ω and k are the angular frequency and wave vector of the ripplon, ρ is the helium density, ρ_s/ρ is the superfluid fraction, d is the film thickness, and it is

assumed that the film is charged to saturation so that the electric field above the film vanishes.) The function F takes into account the effect of image charges in the solid substrate; for dielectric materials it is approximately equal to the dielectric constant ϵ , whereas for metals $F = \coth(kd)$. According to Eq. (1) charging a film reduces the ripplon frequency just as it does for bulk liquid ($d = \infty$), but the critical charge density where ω becomes imaginary is increased for a film. For a thickness of $300 \text{ \AA}$ —a typical value for a saturated film at a height $H = 1 \text{ cm}$ above the bath level—it is expected that an electron density $n_c = 3 \times 10^{10} \text{ cm}^{-2}$ can be reached, about 10 times more than on bulk helium. In fact, densities of that order of magnitude have been observed on such substrates as glass, copper,⁹ and solid neon.¹⁰ However, quantitative investigations of the stability of charged films, in particular as a function of the film thickness d , have not yet been reported.

In our experiment¹¹ we have determined the two relevant quantities entering the problem, the electron density and the film thickness, by capacitor measurements and ellipsometry,¹² as sketched in Fig. 1. A small glow discharge served as the source for the electrons. The bottom capacitor plate of the sample cell was covered with the respective material forming the substrate for the helium film. The top plate had a central part which was vibrated vertically so that a pickup signal proportional to the electric field E_u above the electron layer was induced.¹³ From E_u and the knowledge of the applied voltage U the difference between the lower and the upper field, $E_l - E_u$, and hence the electron density $n = (E_l - E_u)/4\pi e$, could be calculated. A decision whether at a certain density all the electrons were located above the film, or whether some amount had penetrated the helium, was obtained by reducing U to zero or a somewhat negative bias. The electrons on the film, which are only weakly bound, then leave the film surface, whereas those charges which have punched the film are localized on the

FIG. 1. Schematic of the experimental setup: S, electron source; G, guard ring; LI, lock-in amplifier. Ellipsometer components: P, polarizer; R, retardation plate; A, analyzer. The spacing between the capacitor plates is 1 cm.

solid substrate and cannot be removed by small negative fields.

Two examples for electron densities determined by this method are shown in Fig. 2. Here the substrate was a 10- μm -thick polymer foil (Hostaphan) covered with a saturated helium film at 1.6 K. As the film was slowly charged by increase of the applied voltage with the electron source switched on, the electron density followed the dashed line (I). Then the source was turned off and the voltage reduced to probe the mobile part of the electrons (II). In Fig. 2(a), where the film was charged with $0.9 \times 10^{11} \text{ e/cm}^2$, n dropped sharply after an initial delay, with a density of $0.25 \times 10^{11} \text{ cm}^{-2}$ remaining at $U = 0$.¹⁴ This latter value could be further reduced continuously by the application of a negative voltage, until at $U = -250 \text{ V}$ all the charges had disappeared. This indicates that in Fig. 2(a) the stability limit had not been exceeded. Conversely, in Fig. 2(b), where n had been increased to $1.4 \times 10^{11} \text{ cm}^{-2}$, only a fraction of $0.95 \times 10^{11} \text{ cm}^{-2}$ could be removed, whereas the rest of $0.45 \times 10^{11} \text{ cm}^{-2}$ was localized and could be eliminated only by neutralization of the foil with positive ions from the glow discharge. Thus we conclude that on Hostaphan covered with a saturated helium film, densities up to $0.95 \times 10^{11} \text{ cm}^{-2}$ are stable; values of the same order of magnitude were found also for other insulators like Kapton and glass; an influence of temperature was not observed in the investigated range $1.3 \leq T \leq 1.9 \text{ K}$.

At first glance our results are at variance with the stability limit derived from Eq. (1): Not only did the instability threshold appear to be independent of the thickness d_0 of the saturated helium films in the investigated range, $200 \text{ \AA} < d_0 < 600 \text{ \AA}$, but

FIG. 2. Electron density n vs the voltage U applied to the bottom capacitor plate. The substrate for the saturated He film ($H = 1 \text{ cm}$) was a 10- μm -thick Hostaphan foil ($\epsilon = 3.4$). (I), charging, electron source on; (II), discharging, electron source off. The dotted line in (b) indicates the amount of charge localized on the solid substrate.

also the experimental absolute value of the maximum charge density was distinctly larger than anticipated for these films. This apparent discrepancy is resolved when the electronic pressure p_{el} exerted on the film is taken into account. Since p_{el} adds to the gravitational and van der Waals terms in the chemical potential, the equilibrium thickness of a charged film will be reduced to^{11,15}

$$d = d_0 (1 + 2\pi n^2 e^2 / \rho g H)^{-1/3}. \quad (2)$$

Here $d_0 = (\alpha / \rho g H)^{1/3}$ is the thickness of the film before charging. The value of d given by Eq. (2) is an average, around which the local film thickness is expected to vary because of the formation of a dimple underneath each electron.¹⁵⁻¹⁸ As a result of the reduction of d , the instability threshold, calculated from Eq. (1), is raised, the film thus stabilizes itself, and a higher charge density can be obtained.

The change in film thickness on charging was confirmed experimentally by an ellipsometric determination of d , as plotted in Fig. 3 for two values of d_0 . The agreement between the data and the solid lines representing Eq. (2) appears to be quite satis-

FIG. 3. Thickness d of charged saturated ^4He films wetting a glass substrate ($T = 1.6$ K) vs the electron density of the films. The thickness of d_0 of the uncharged films was 220 and 420 Å, respectively. (The relatively low electron density reached in this run was not limited by an instability of the charges on the film, but by electric breakdown in the leads.)

factory. Since for high electron densities the curves converge, it is not surprising that the observed stability limit is not significantly influenced by the initial film thickness.

Whereas the data presented so far refer to insulating substrates, we have extended the experiments also to charged helium films on conductors, for which somewhat controversial predictions regarding their stability have been made.^{1,3,15} An ellipsometric measurement of a film supported by a conducting doped silicon sample cut from a commercial wafer is shown in Fig. 4. Like for the insulators, charging led to a reduction of the film thickness, which implies that also for conducting substrates stable electron states above the helium film exist.¹⁹ With use of Eq. (2) the electron density is estimated to be $0.7 \times 10^{11} \text{ cm}^{-2}$ at the smallest thickness reached in Fig. 4, $d = 0.18d_0$. (A determination of the charge density with the vibrating capacitor plate was not possible here because for conducting substrates the sensitivity of this method is strongly reduced.)

Although the maximum charge density on insulators and conductors is of the same order of magnitude, a significant difference between the two classes of substrates is observed, which hints at an additional possible origin for the loss of charge from the film surface: On our silicon sample the film thickness starts to increase as soon as the electron source is turned off, and a gradual relaxation back

FIG. 4. Thickness of a saturated ^4He film on silicon vs time. Charging started at A ; at B the electron source was switched off, but the holding voltage ($U = 10$ V) was still applied.

to a value only slightly below that of the uncharged film occurs (cf. Fig. 4). In contrast, insulating substrates showed no such variation on a comparable time scale. One might interpret this behavior as due to tunneling of the electrons through the helium film at some rough structures of the surface, where the equilibrium film thickness probably is substantially reduced.²⁰ For an insulating substrate such centers will be charged already by the very first electrons to tunnel, so that the majority of the surface electrons is repelled from these spots and hence prevented from tunneling. On the surface of conductors, on the other hand, the electrons do not accumulate, and therefore this “healing” of weak spots cannot become effective and the charges leak away.

In conclusion, our experiments have shown that electron densities up to about 10^{11} cm^{-2} —the highest value reported so far—are stable on saturated helium films. The substrate material appears to be of minor importance, but nonideal surface quality may favor loss mechanisms such as tunneling over the collective electrohydrodynamic instability, which so far has been mostly considered as the limiting factor.^{1,3,7} In order to reach the high densities it is not necessary to adjust the film thickness carefully, because d is essentially determined by the electronic pressure. With these systems a density range has become accessible where quantum effects in the 2D electron system ought to be relevant, although these electrons will probably deviate in their behavior from the ideal system considered earlier³ because of their trapping in a polaron state.¹⁸ Furthermore, the influence of the high electron densities on the ripplon spectrum, as predicted by Eq. (1), appears as an interesting phenomenon to

study.

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