Stability of helium films charged with electrons

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The linear stability of 100-Å films of helium coated with high densities of electrons ($n \ge 10^9$ cm⁻²) is investigated. We find that the van der Waals force stabilizes such films and that such systems offer a new and intriguing experimental system.

I. INTRODUCTION

Recently, there has been a great deal of interest in the system composed of electrons trapped at the free surface of liquid helium.^{1,2} The system is important because it is extremely well defined and displays a wide range of physically interesting phenomena which may be varied by the application of external fields and voltages. The freezing of the electron liquid and the behavior of the surface near its maximum supportable charge density³ are two of the more interesting phenomena.

In the experiments on the Coulomb solidification,² the surface of bulk helium, whose thickness $d \sim 1$ mm, is charged with electrons to densities $n < 10^9$ cm^{-2} . The temperature is then lowered through the transition temperature $T_c(n \propto T_c^2)$. For $n \simeq 10^9$ cm^{-2} , $T_c \simeq 0.3$ K. In this regime of density and temperature, the system is purely classical, i.e., $k_B T_c >> \pi \hbar^2 n / m \equiv E_F \ (E_F \cong 10^{-2} k_B \text{ for } n = 10^9$ cm^{-2}), the Fermi energy of the electron liquid. The evidence so far seems to suggest that such classical melting occurs in a way described by Kosterlitz and Thouless,⁴ i.e., by virtue of the unbinding of pairs of defects. At zero degrees, another kind of freezing (melting) should occur. The so-called Wigner transition, from liquid to solid in the purely quantum regime, occurs as the density of the electron gas is lowered.⁵ Although the value of $r_s = (\pi/n)^{1/2} m e^2/\hbar^2$ at which this occurs is not known precisely, we expect it to occur for $n \ge 10^{11}$ cm⁻², a value far in excess of that which the bulk helium surface can support. Quantum-mechanical melting, i.e., the entire phase diagram⁵ of the two-dimensional (2D) electron liquid would be very interesting to investigate. It requires that we somehow manage to stabilize the helium surface to higher charge densities.

In addition to electron liquid solidification, the instability of the helium surface has been studied.³ It is an interesting phenomenon in its own right, i.e., it gives us a simple variable system for studying many types of hydrodynamic phenomena. Without going into too much detail, it suffices to say that the surface is unstable because the frequency ω of the ripplons (capillary waves) softens as the density of the

electrons is increased. For bulk helium having a mass density ρ with surface tension τ in the limit $(k/2\pi)^2/n \ll 1$ (Refs. 6 and 7):

$$\omega^2 = gk + \frac{\tau}{\rho}k^3 - \frac{4\pi e^2 n^2}{\rho}k^2 \quad . \tag{1}$$

Here, g is the gravitational constant. The wave number at which the instability occurs is given by

$$k_c = (g\rho/\tau)^{1/2} . (2)$$

The corresponding critical density is given by

$$n_c = (\tau g \rho)^{1/4} / (2\pi e^2)^{1/2} \quad . \tag{3}$$

For helium, $\tau/\rho \simeq 2 \text{ dyn cm}^2/\text{g}$, $k_c \simeq 30 \text{ cm}^{-1}$, and $n_c \simeq 10^9 \, {\rm cm}^{-2}$.

In this paper, we would like to analyze the behavior of thin (approximately 100-Å) films of helium when they are charged with electrons. We will show that large electron densities can be expected to be stabilized and that a wide range of new and interesting physical possibilities exist for such systems.

II. SUPERFLUID FILM

When the film of helium is superfluid and at low enough temperatures, the problem has in some sense already been solved. The linearized equations governing the fluid motion are^{7,8}

$$\rho \frac{\partial \nabla}{\partial t} = -\nabla p + \left[\left(\tau \frac{\partial^2 a}{\partial x^2} - en \frac{\partial \phi}{\partial z} \right) \delta(z - a) - \frac{3\alpha}{(z + d)^4} - \rho g \right] \hat{z} \quad , \tag{4}$$

along with the incompressibility condition

. .

 $\vec{\nabla}\cdot\vec{\mathbf{v}}=0~~.$ (5)

Here, \vec{v} is the fluid velocity, p is the pressure, ϕ the electrostatic potential, α is the van der Waals constant characterizing the force between the substrate and the helium, and a is the deviation of the surface from equilibrium position. We have chosen the z

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axis perpendicular and the x axis parallel to the helium surface. When there is no perturbation, the surface is at z = 0. The bottom of the helium film z = -d contacts a solid insulating substrate. At thicknesses of 10^3 Å or less, the bulk force in Eq. (4) is dominated by the van der Waals attraction, $-3\alpha/(z+d)^4$.

We examine a surface perturbation of the form $\exp(ikx - i\omega t)$ by employing the methods described in Refs. 7 and 8. Using a boundary condition $\nabla \cdot \hat{z} = 0$ at z = -d, one obtains the dispersion relation

$$\omega^2 = \omega_0^2 \tanh(kd) \quad , \tag{6}$$

where

$$\omega_0^2 = \left(\frac{3\alpha}{\rho d^4} + g\right)k + \frac{\tau}{\rho}k^3 - \frac{4\pi e^2 n^2}{\rho}k^2 F(k)$$
(7)

and

$$F(k) = \frac{1 + \epsilon - (1 - \epsilon)e^{-2kd}}{1 + \epsilon + (1 - \epsilon)e^{-2kd}}$$

when the surface is fully charged by the electrons. Hereafter, we consider thin films $(kd \ll 1)$, so that $F(k) = \epsilon$, the dielectric constant of the insulating substrate. The function F takes into account the effect of image charges in the insulating substrate. All image charges, because they reduce the Coulomb repulsion between electrons, tend to destabilize the surface. In addition to insulator substrate, a metal electrode is required. It is the conductor to which the voltage is applied which binds the electrons to the surface. The image charges on this metal electrode would also tend to destabilize the surface, i.e., produce an additional factor analogous to F. However, if the conducting electrode is at a distance far from the surface, (kd' >> 1), then there is no effect from such image charges.

It is important to note that for typical materials like glass $\alpha = 9.5 \times 10^{-15}$ in cgs unit $3\alpha/(\rho d^4g) \simeq 2 \times 10^8$ for d = 100 Å. The van der Waals attraction to the substrate acts to stabilize the surface. The maximum supportable electron density limited by the hydrodynamic instability is now given by

$$n_{c} = (3\alpha\tau)^{1/4} / (2\pi e^{2}\epsilon)^{1/2} d$$
(8)

and instability occurs at a wave vector

$$k_c = (3\alpha/\tau)^{1/2}/d^2$$
 (9)

when $n > n_c$. Both $n_c \epsilon^{1/2}$ and k_c are plotted in Fig. 1 as a function of d for a glass substrate.

In deriving Eqs. (6) and (7), the electrostatic term in Eq. (4) is taken into account by assuming that the electric field is continuous and that it instantaneously forms an equipotential surface. For linear disturbances, such an assumption is valid when the frequencies of the mode at the wave number is small compared to the two-dimensional plasmon frequency



FIG. 1. The maximum supportable density n_c and the critical wave number k_c as a function of film thickness.

 $\omega_p^2(k) = (2\pi ne^2/m)k$, and that the electron fluid is continuous, i.e., that $(k_c/2\pi)^2 \ll n$. The first condition is well satisfied for density $n > 10^5$ cm⁻². To examine the second condition, $(k_c/2\pi)^2$ is plotted as a function of *d*. The continuous fluid approximation is valid for densities above this line. Thus down to thickness of 100 Å, the fully charged system is described by our approximation and is hydrodynamically stable.

Although the hydrodynamic analysis indicates that $n \approx 10^{11}$ cm⁻² is supportable, the electric field at this density becomes as high as 180 kV/cm. At this high field, the surface *may* become unstable to tunneling of the electrons through the 1-eV barrier at the surface into the liquid.⁹ As long as the tunneling does not occur, the thin film provides a hydrodynamically stable situation for high density (or electric field).

III. STABILITY WITH VISCOSITY

For the case of viscous helium films, for example, ³He or He I, the linearized capillary wave problem for arbitrarily thick films, to our knowledge, has not been solved. This regime is interesting for at least two reasons.

The dynamics, i.e., the growth rate of unstable capillary waves will be dominated by viscosity and the ultimate nonlinear evolution of these waves will be quite different from the zero-viscosity case. In addition, it is important to know the hydrodynamic dispersion relation of ripplons near $k = (2\pi n)^{1/2}$ if one is to do experiments analogous to the Grimes-Adams experiment at temperatures higher than the lambda point. In this section, we will analyze the linear stability problem of a viscous He film charged with electrons.

The equations governing the fluid are the Navier-Stokes equations⁸ with viscosity μ , i.e.,

$$\rho \frac{\partial v_x}{\partial t} = \mu \left(\frac{\partial^2 v_x}{\partial x^2} + \frac{\partial^2 v_x}{\partial z^2} \right)$$
$$-\mu \left(\frac{\partial v_x}{\partial z} + \frac{\partial v_x}{\partial x} \right) \delta(z - a) - \frac{\partial p}{\partial x} ,$$
$$\rho \frac{\partial v_z}{\partial t} = \mu \left(\frac{\partial^2 v_z}{\partial x^2} + \frac{\partial^2 v_z}{\partial z^2} \right) \qquad (10)$$
$$+ \left(-2\mu \frac{\partial v_z}{\partial z} + \tau \frac{\partial^2 a}{\partial x^2} - en \frac{\partial \phi}{\partial z} \right) \delta(z - a)$$
$$- \frac{\partial p}{\partial z} - \rho g - \frac{3\alpha}{(z + d)^4} ,$$

and

$$\vec{\nabla} \cdot \vec{\mathbf{v}} = 0 \tag{11}$$

for the incompressible fluid.

The solution which depends on x and t as $exp(ikx - i\omega t)$ is of the form,

$$v_{x} = (Ae^{kz} + Be^{-kz} + Ce^{mz} + De^{-mz})e^{ikx - i\omega t} ,$$

$$v_{z} = i \left[-Ae^{kz} + Be^{-kz} - \frac{k}{m}Ce^{mz} + \frac{k}{m}De^{-mz} \right]e^{ikx - i\omega t} ,$$

$$p = \frac{\omega}{k}\rho (Ae^{kz} + Be^{-kz})e^{ikx - i\omega t} - \rho gz - \frac{\alpha}{(z+d)^{3}} ,$$
(12)

where

$$m = (k^2 - i\omega/\nu)^{1/2}, \quad \nu = \mu/\rho$$

The four coefficients A, B, C, D are determined by four boundary conditions. Two boundary conditions at the liquid surface, z = a, are obtained by integrating Eqs. (10) across the surface, i.e., no shear,

$$\frac{\partial v_x}{\partial z} + \frac{\partial v_z}{\partial x} = 0 \quad , \tag{13}$$

and by setting

$$\frac{\partial}{\partial t} \left(p - 2\mu \frac{\partial v_z}{\partial z} + \tau \frac{\partial^2 a}{\partial x^2} - en \frac{\partial \phi}{\partial z} \right) = 0 \quad . \tag{14}$$

The conditions at z = -d are simply

$$v_x = 0, v_z = 0$$
 (15)

Following a prescription described in Ref. 7, one can write *n* and $\partial \phi / \partial z$ in Eq. (14) in terms of *a*. On the liquid surface, *z* in Eq. (12) equals *a*. Eliminating *p* from Eqs. (12) and (14) and using $\partial a / \partial t = v_z$ at z = a, we obtain an equation for v_x and v_z . The determinant of the four boundary conditions gives us

a dispersion relation

$$\Delta(\omega,k) = \begin{vmatrix} e^{kd} & e^{-kd} & e^{md} & e^{-md} \\ e^{kd} & e^{-kd} & \frac{k}{m}e^{md} & -\frac{k}{m}e^{-md} \\ -1 & +1 & -\frac{(m^2+k^2)}{2mk} & \frac{(m^2+k^2)}{2mk} \\ C_1 & C_2 & C_3 & C_4 \end{vmatrix} = 0 ,$$
where

$$C_{1} = \frac{\omega^{2}}{\omega_{0}^{2}} + \frac{2i\omega\nu k^{2}}{\omega_{0}^{2}}, \quad C_{2} = \frac{\omega^{2}}{\omega_{0}^{2}} + \frac{2i\omega\nu k^{2}}{\omega_{0}^{2}} - 1 \quad ,$$
$$C_{3} = \frac{2i\omega\nu k^{2}}{\omega_{0}^{2}} + \frac{k}{m}, \quad C_{4} = \frac{2i\omega\nu k^{2}}{\omega_{0}^{2}} - \frac{k}{m} \quad ,$$

and $v = \mu/\rho$.

In some limiting cases, it is possible to write down approximate expressions for this dispersion relation. In the thick limit, $e^{-kd} = 0$ and $e^{-md} = 0$, Eq. (16) reduces to

$$\left(2 - i\frac{\omega}{\nu k^2}\right)^2 + \frac{\omega_0^2}{\nu^2 k^4} - 4\left(1 - i\frac{\omega}{\nu k^2}\right)^{1/2} = 0 \quad , \quad (17)$$

where ω_0^2 is given in Eq. (7). Equation (17) is the result given by Landau and Lifshitz⁸ slightly modified by the electrostatic, surface tension, and van der Waals effects. If $\omega_0^2/\nu^2 k^4 >> 1$, one obtains

$$\omega \simeq \pm (\omega_0^2)^{1/2} - i 2\nu k^2 \quad . \tag{18}$$

We have purely damped (or growing) modes

$$\omega \simeq -i \frac{\omega_0^2}{2\nu k^2}, \quad -i\nu k^2 \quad , \tag{19}$$

when $\omega_0^2/\nu^2 k^4 \ll 1$. The stability, i.e., the static equilibrium is totally determined by the sign of ω_0^2 as it is in the case of no viscosity. However, the growth rate is dependent on the viscosity.

In the thin limit $kd \ll 1$, it is possible to approximate the determinant when $|\omega/k\nu^2| \ll 1$ and obtain

$$\omega = -i \frac{\omega_0^2}{3\nu k^2} (kd)^3 \quad . \tag{20}$$

The stability is again determined by the sign of ω_0^2 .

The dispersion relation can be solved numerically for a realistic case. If we employ a glass substrate elevated from a bulk helium surface by 1 cm, a film with d = 400 Å is formed. We adopt parameters of He I; $\nu = 2 \times 10^{-4}$ P cm³/g, $\alpha = 9.5 \times 10^{-15}$, and $\tau/\rho = 2$ in cgs units. Under these conditions, $k_c = 1.9 \times 10^4$ cm⁻¹ and $n_c = 6.3 \times 10^{10} e^{-1/2}$ cm⁻². Figure 2 shows the dispersion relation when the electrons are almost fully charged, i.e., when $n = 6.2 \times 10^{10} e^{-1/2}$ cm⁻². When $k \ge 8.4 \times 10^4$ cm⁻¹, we find two roots ω_1 and ω_2 where Re($\omega_1 + \omega_2$) = 0, Im($\omega_1 - \omega_2$) = 0 as in Eq. (18). However, as shown in Fig. 2 by dotted lines, Eq. (18) is not a good approximation because the condition $\omega_0^2/\nu^2 k^4 \gg 1$ is not satisfied very well for k shown in the figure.



FIG. 2. The dispersion relation of ripplons when d = 400 Å, $\nu = 2 \times 10^{-4}$, $\alpha = 9.5 \times 10^{-15}$, $\tau/\rho = 2$, and $n = 6.2 \times 10^{10} \epsilon^{1/2}$ in cgs units.

Grimes and Adams employed plasmon-ripplon coupling in order to detect the Wigner lattice. This technique works when the ripplon resonance is sharp, i.e., when $\omega_r/\omega_i >> 1$ at $k = (2\pi n)^{1/2}$. For $n \approx 10^{10}$ cm⁻² (see scale shown in Fig. 2), $\omega_r/\omega_i \approx 6$, indicating that plasmon-ripplon coupling could be observed even at temperatures above the lambda point.

More details of the dispersion relation at smaller wave numbers are shown in expanded Fig. 3. For $k < 8.4 \times 10^4$ cm⁻¹, ω_r vanishes and two purely imaginary roots appear. One has a large negative value and very insensitive to *n*. The other one is expressed by Eq. (20) within an error of a few percent for *k* around k_c , which is 1.9×10^4 cm⁻¹. This is the branch which determined the system's hydrodynamic stability. For these 400 Å films, the growth (or damping) time $t_0 \simeq 10 \ \mu$ sec. We will make no at-

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FIG. 3. The dispersion relation near the critical wave number. The parameters are the same as those employed in Fig. 2.

tempt to analyze the nonlinear evolution of the unstable waves in this system.

IV. CONCLUSION

The simple analysis presented here shows that thin 100-Å films of helium offer intriguing possibilities for stabilizing high densities of mobile electrons in order to observe quantum-mechanical effects in the 2D solidification of the electron liquid. The presence of a number of variable dielectric substrates also allows one to think about dipolar gases, effective electron charges, and polaronic effects which could be physically very interesting.

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