

# Properties of a self-localized charged layer on the surface of liquid helium

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It is shown that the interaction between electrons significantly affects the structure of a charged layer on the surface of liquid helium. The stability of this layer is also investigated. At low temperatures, the stability is lost jumpwise with increasing electron density on the surface.

## 1. INTRODUCTION AND STATEMENT OF THE PROBLEM

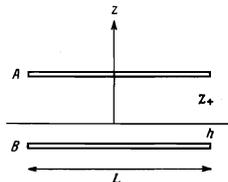
It has been shown in the papers of Shikin<sup>[1]</sup> and Cole and Cohen<sup>[2]</sup> that the surface of helium is capable of holding electrons near it. The corresponding potential well has the character of a barrier on the liquid helium side with energy  $\sim 1.4$  eV, due to the negative affinity of the electrons; at the same time, the effect of image forces causes their attraction. The binding energy amounts to  $\sim 10^\circ\text{K}$ ; however, as Crandall and Williams have pointed out,<sup>[3]</sup> the Coulomb nature of the spectrum of a single electron near the surface leads to virtual ionization and withdrawal of the electrons into the volume of the liquid at sufficiently high temperatures. Furthermore, a finite surface density of electrons  $n_S$  can be obtained only at the expense of the imposition of an electric field intensity:  $4\pi en_S \lesssim E$ . In the experiments of<sup>[3-5]</sup>, we have densities  $n_S \sim 10^8 - 10^9 \text{ cm}^{-2}$ , which correspond to fields up to  $10^3 \text{ V/cm}$ .

Under such conditions, the surface electrons on the boundary of liquid helium are actually observed,<sup>[3-5]</sup> but their properties are naturally quite different from those of the model considered in<sup>[1,2]</sup>. For sufficiently strong fields, the first difference is that the effective thickness of the layer  $z$ , while remaining macroscopic, is determined by the value of the impressed electric field. Thus, in the limit of high electric fields,<sup>[6]</sup>  $\bar{z} \sim (\hbar^2/m_e)^{1/3} (e/E)^{1/3}$ . Assuming that  $E \sim en_S = er_S^2$ , we get

$$\bar{z} \sim a_0^{1/2} r_S^{3/4},$$

where  $a_0$  is the Bohr radius and  $r_S$  is the mean distance between the electrons on the surface. The value of  $\bar{z}$  is comparable with the characteristic diameter of localization of the state<sup>[1,2]</sup> for  $n_S \sim 10^8 - 10^9 \text{ cm}^{-2}$  ( $E \sim 10^2 - 10^3 \text{ V/cm}$ ).

It is still more important that the motion of the electrons cannot as a rule be regarded as free, because of the long-range nature of the Coulomb interaction. The interaction of the electrons with one another essentially determines the structure of the charged layer on the surface of the liquid helium. Finally, as has been noted above, ionization at finite temperatures removes a significant fraction of the electrons to points far from the surface.



In what follows, we shall have in mind the usual experimental situation shown in the figure. A potential difference is applied between the metal plates B ( $z = -h$ ) and A ( $z = z_+$ ). The helium surface with the electrons corresponds to  $z = 0$ . Sufficient dimensions  $L$  of the lower plate in comparison with its distance  $h$  from the helium surface guarantees good homogeneity of the charge distribution and field in this "capacitor." Usually, the surface charge cancels out the field above the helium surface (i.e., for  $z > 0$ ).

The effect of the factors enumerated above on the structure of the electron layer on the helium surface is studied below. The interelectronic Coulomb interaction is taken into account in two limiting cases: a) when the interaction between the electrons is small in comparison with the kinetic energy, b) in the opposite case, when the Coulomb energy of interaction predominates. The first case corresponds to sufficiently low densities and high temperatures, and the second to sufficiently low temperatures. In the latter case, as Crandall and Williams have pointed out,<sup>[7]</sup> it can be assumed that the conditions for the existence of a two-dimensional Wigner lattice are in the main satisfied.

In this paper, we shall be interested in the structure of the charged layer under different conditions, and in its stability. The presence of the electrons has an appreciable effect on the long-wave portion of the spectrum of surface oscillations. It will be shown below that the structure of the layer is significant. The acoustic and plasma branches of the spectrum in the high density case were recently obtained by Crandall.<sup>[8]</sup> In the case of high densities, as noted in<sup>[6]</sup>, there exists a limit of stability of the charged layer due to the competition of the stretching electrical forces and the action of the surface tension and the force of gravity, which tend to preserve the plane shape of the boundary. This problem is also discussed below.

## 2. THE CHARGED LAYER IN THE CLASSICAL LIMITING CASE

At the very boundary of the helium, the electrons experience the effect of image forces. Therefore, in the case of weak compressing fields, there are, of course, localized levels. Their population, however, in spite of the rather large binding energy, can be low, inasmuch as the electrons are generally capable of penetrating into the volume under the helium at finite temperature, as has been pointed out. This case is also considered in the present work. The interactions between the electrons lead to a change in the form and screening of the compressing field, and, in the final analysis, as we shall see, guarantee the self-localization of electrons at distances that are large in comparison with the dimensions of the deep near-surface levels.

The considered case corresponds to the classical limit of a rarefied plasma. We begin with the determination of the structure of this layer of electrons. We assume that the electrons are located in a potential well near  $z = 0$ , formed by a self-consistent field. In other words, we assume that the volume density  $n$  depends only on the  $z$  coordinate:

$$n = n_0 e^{e\varphi/T},$$

where the potential  $\varphi$  satisfies the Poisson equation. We neglect the image forces at these distances. The conditions under which this is possible are given below.

We use the notation  $y = e\varphi/T$  and  $c^2 = 4\pi e^2 n_0 / T$ . The one-dimensional Poisson's equation takes the form

$$y'' = c^2 e^y. \quad (1)$$

We can obtain the potential distribution from (1). The first integral of (1) is

$$(y')^2 = 2c^2 e^y + D^2.$$

The choice of the root

$$y' = -(2c^2 e^y + D^2)^{1/2} \quad (1')$$

is determined by the geometry of the drawing; the potential energy of the electrons keeps them around  $z = 0$ . The quantity  $-(2c^2 + D^2)^{1/2}$  is proportional to the field intensity at the upper plate:

$$y'|_{z=z_+} = -(2c^2 + D^2)^{1/2}.$$

Integration of (1') gives

$$\frac{1}{D} \ln \frac{(2c^2 + D^2)^{1/2} - D}{(2c^2 + D^2)^{1/2} + D} - \frac{1}{D} \ln \frac{(2c^2 e^y + D^2)^{1/2} - D}{(2c^2 e^y + D^2)^{1/2} + D} = z - z_+ \quad (2)$$

where  $z_+$  is the position of the upper plate. The potential is thus calculated from the potential of the plate A. The case  $c^2 e^y \ll D^2$  corresponds to the static electric field between A and B in the absence of electrons:

$$y = -D(z - z_+).$$

In the general case, it is convenient to rewrite (2) in the form

$$y = -2 \ln \left\{ \frac{\sqrt{2c^2} \operatorname{sh} \left[ \frac{D}{2}(z + \kappa) \right]}{D} \right\}, \quad y' = -D \operatorname{cth} \left[ \frac{D}{2}(z + \kappa) \right], \quad (3)$$

$$\kappa = -z_+ - \frac{1}{D} \ln \frac{(2c^2 + D^2)^{1/2} - D}{(2c^2 + D^2)^{1/2} + D}. \quad (3')$$

In the liquid helium ( $z < 0$ ) there are no electrons and the field distribution is determined by the Laplace equation  $\nabla^2 \varphi = 0$ , i.e.,  $\varphi = \alpha - \beta z$ . On the surface, the condition of continuity of the potential and its derivative (the normal component of the field) connect the new constants with the solution (3). The boundary condition on the plate B amounts to furnishing the potential difference between A and B:  $y|_z = -h = eV/T$ .

Thus, only a single combination is determined from the two constants  $c$  and  $D$  in (3) and (3'). The remaining arbitrariness is connected with the fact that the volume change between the plates can be arbitrary. It is seen from the Eqs. (3) that this charge is, generally speaking, by no means concentrated at the surface of the helium. Self-localization of the charge arises only when the total charge in the volume is sufficient for almost complete screening of the field. This case is of fundamental interest.<sup>1)</sup>

For complete screening of the field in the space above the helium, the quantity  $(2c^2 + D^2)^{1/2}$  in (2), which

is proportional to the field intensity at  $z = z_+$ , is small. Letting  $D \rightarrow -$  in (3), we obtain

$$n(z) = T/2\pi e^2(z + \kappa)^2. \quad (4)$$

The total surface density  $n_S = T/2\pi e^2 \kappa$  is connected with the total potential difference, which now turns out to be applied to the gap between the helium surface and the lower plate:

$$4\pi e^2 n_S = eV/h. \quad (5)$$

Thus  $\kappa = T/2\pi e^2 n_S$  has the meaning of the effective thickness of the surface layer of localized electrons. The volume density in the layer is

$$n = n_S / \kappa = 2\pi e^2 n_S^2 / T. \quad (6)$$

We now discuss the conditions of applicability of Eq. (4). It is clear that, in order to speak of the surface layer,  $\kappa$  must be small in comparison with the basic dimensions in the problem. The latter are usually of the order of 0.0–1 cm.<sup>[3]</sup> More significant limitations arise from the use of the self-consistent field approximation (1). The condition of smallness of the Coulomb terms in comparison with the kinetic energy of the electrons takes the form

$$e^2 n_S^{3/2} \ll T. \quad (7)$$

The presence of an electric field at the surface leads (departing from the image forces) also to discrete levels  $\Delta \varepsilon \sim \hbar^{2/3} m^{-1/3} (T/\kappa)^{2/3}$ . The condition of their smallness in comparison with the temperature is conveniently written in the form

$$\frac{\Delta \varepsilon}{T} \sim \left( \frac{e^2 n_S^{3/2}}{T} \right)^{2/3} \left( \frac{\hbar^2 n_S}{mT} \right)^{1/3}.$$

The second factor is also small in the case of satisfaction of (7).

Furthermore, we shall assume that the interaction of the charge with its image on the surface of the helium does not play a significant role for the layer as a whole. For this, the characteristic dimension  $d$  of the bound state<sup>[1,2]</sup> should be small in comparison with  $\kappa$ . Finally, the population of the levels should be small.<sup>[1,2]</sup> For sufficiently high temperatures, the statistical weight connected with almost free motion at distances of the order of  $\kappa$  should be of the order of  $\kappa (mT)^{1/2} / \hbar$  in the logarithmic potential (3). Therefore, the fraction of atoms  $\nu$  located at the level with binding energy  $\varepsilon_0 = 7.5^\circ \text{K}$  is, from<sup>[9]</sup>

$$\nu = \frac{n_0}{n_s} \sim \frac{\hbar}{\kappa \sqrt{mT}} e^{\varepsilon_0/T} \sim \frac{e^2 n_s^{1/2}}{T} \left( \frac{\hbar^2 n_s}{mT} \right)^{1/2} e^{\varepsilon_0/T}. \quad (7')$$

The factor  $(\hbar^2 n_S / mT)^{1/2}$  for  $n_S \approx 10^5 - 10^6 \text{ cm}^{-2}$  is of the order of  $10^{-3}$ . Thus, the population of the localized states is small for  $T > 1.1^\circ \text{K}$ .

### 3. LOW-FREQUENCY OSCILLATION SPECTRUM

The presence of electrons on the liquid surface affects the dispersion law of the surface waves. We shall assume the inequality  $kh \gg 1$  so as to disregard the boundary conditions on the metallic plate. Moreover, it is assumed that the frequencies are small in comparison with the plasma frequencies, as a consequence of which we can use the Boltzmann expression for the particle number density  $n = n_0 e^{e\varphi/T}$ . Therefore, it is appropriate to solve the linearized Poisson equation over the helium surface and to use the hydrodynamic equations in the helium itself.

We now consider the surface waves briefly with account of the surface layer of electrons. The equations for the dispersion law are obtained by equating the forces acting on the helium surface (see, for example, [6] or [10]). The pressure in the electron gas is  $P_e = nT$ , the corrections to this equation are small by virtue of (7). Therefore, the nontrivial point is that the density in the electron layer is inhomogeneous and takes the form (4). Selecting the perturbed electron number density in the form

$$n_0 + \delta n = n_0(1 + y), \quad y = f(z) \exp[i(kr - \omega t)],$$

we obtain from the Poisson equation the following equation for the determination of  $f$ :

$$f'' - k^2 f = 2f / (z + \kappa)^2. \quad (8)$$

A solution of (8), which falls for as  $z \rightarrow +\infty$ , is

$$f(z) = Ck[1 + 1/k(z + \kappa)] \exp[-k(z + \kappa)]. \quad (8')$$

Solving also the equation of potential flow in the liquid, and using the continuity of the electric field potential and the field directly on the helium surface (as is done, for example, in [6]), we obtain the following dispersion relation for the surface oscillations:

$$\omega^2 = \frac{k}{\rho} \left\{ \rho g + \alpha k^2 - 4\pi e^2 n_s^2 k \frac{1 + 2k\kappa}{1 + 2k\kappa + 2(k\kappa)^2} \right\}. \quad (9)$$

In the limit  $k\kappa \ll 1$  (long waves) the expression (9) coincides with the oscillation spectrum on the surface of a charged conductor. [10] In the general case, the third term in brackets in (9) leads to a nontrivial dispersion spectrum of the surface wave oscillations. In the region of applicability of our formulas, which is given by the inequality (7), the third term in (9) for the helium represents a very small correction. However, for neon, where the condition (7) is broader, it can reach several dozen percent.

Along with oscillations of the type considered, there also exists a single low-lying branch which corresponds to oscillations of the charge in the plane. The latter, generally speaking, are also connected with the bending of the surface; however, if the considered frequencies are sufficiently large in comparison with (9), then we have in this region (see [8] and the next section)

$$\omega = (2\pi e^2 n_s k / m)^{1/2}. \quad (10)$$

In ordinary three-dimensional Langmuir oscillations, the dispersion law takes the form

$$\omega^2 = \omega_p^2 [1 + 3/2 (r_D k)^2], \quad (11)$$

where  $\omega_p = (4\pi e^2 n / m)^{1/2}$  is the plasma frequency and  $r_D = (T / 4\pi e^2 n)^{1/2}$  is the screening radius. In the oscillations described by the dispersion law (10), the charge distribution along the  $z$  axis does not change. Therefore, complete screening is lacking in the two-dimensional Coulomb problem. At the same time, it is almost certain that plasma oscillations are possible in an inhomogeneous charged layer (4), corresponding to discrete  $k_z \sim 1/\kappa$  in the dispersion law (11) (according to (6),  $r_D \approx \kappa$ ). Accurate calculation is made difficult by the necessity of solving simultaneously the kinetic equation for the distribution function and the self-consistent Poisson equation of the inhomogeneous problem (4). However, the general form of the plasma branches

$$\omega_n^2 = \omega_{0n}^2 [1 + \text{const} \cdot (\kappa k_n)^2] \quad (12)$$

(if they exist) follows from (11). In (12), we have for  $\omega_{0n}$

$$\omega_{0n} \sim \kappa^{-1} \sqrt{T/m} \sim 10^3 n_s / \sqrt{T} \text{ [deg]}.$$

#### 4. LAYER STABILITY (HIGH DENSITIES)

In the limiting case of low temperatures. (the condition is  $T \ll e^2 n^{1/3}$ ) the Coulomb energy of interaction predominates and, as was shown by Crandall and Williams, [7] conditions are created for the Wigner crystallization of the electrons in the near-surface layer. The forces acting on the electrons are determined by the potential  $\Phi$ , which is equal to [6]

$$\Phi = \frac{e^2}{2} \sum_{i \neq j} \frac{3((r_j - r_i)(u_j - u_i))^2 + (r_j - r_i)^2 (u_j - u_i)^2}{|r_j - r_i|^5}, \quad (13)$$

where  $u_i$  is the deviation of the electron from the equilibrium position at the point  $i$  of the lattice. The spectrum of oscillations of the electrons in the plane  $z = 0$  is determined by the equations

$$m \ddot{u}_i = -\partial \Phi / \partial u_i,$$

which, according to (13), take the form

$$m \ddot{u}_i = e^2 \sum_{i \neq j} \frac{3((r_j - r_i)(u_j - u_i))(r_j - r_i) - (r_j - r_i)^2 (u_j - u_i)}{|r_j - r_i|^5}. \quad (14)$$

For long-wave oscillations,  $kr_s \ll 1$ , and, in the continuum approximation we get two oscillation branches from (14) by replacing the summation by an integral:

$$\begin{aligned} \omega &= (2\pi e^2 n_s k / m)^{1/2} - \text{longitudinal oscillations (see (10))}, \\ \omega &= 0 - \text{transverse oscillations.} \end{aligned}$$

Account of the elastic forces in the electron lattice leads to the result that in place of  $\omega = 0$  a branch of elastic oscillations develops with a frequency proportional to the first power of  $k$ . The corresponding sound velocities naturally depend on the direction of propagation in the lattice and were found in [8]. It was also shown that the condition for stability of the lattice relative to thermal fluctuations has the form

$$\frac{\langle u \rangle^2}{r_s^2} \sim \frac{T \ln(L \sqrt{n_s})}{\pi e^2 \sqrt{n_s}} \sim \frac{10^2}{\sqrt{n_s}} T \text{ [deg]} \ln \frac{L}{r_s} \ll 1, \quad (15)$$

where  $L$  is the dimension of the system in the figure. The estimate (15) leads to reasonable temperatures.

In conclusion, we make a few remarks concerning the problem touched on in [6] regarding the maximum possible electron concentration on the surface. As has already been noted, the presence of a charged layer reveals itself in the dispersion law of the surface oscillations (in the region of wavelengths of the order of the capillary constant  $k \sim (\rho g / \alpha)^{1/2}$ ). The spectrum for arbitrary  $kh$  was found in [6]:

$$\omega^2 = k\rho^{-1} \text{th}(kh) \{ \rho g + \alpha k^2 - 2\pi e^2 n_s^2 k [1 + \text{cth}(2kh)] [1 + 2e^{-2kh}] \}. \quad (16)$$

In particular, for  $kh \gg 1$ , it transforms into the well-known expression for the frequency of electro-capillary waves [10]

$$\omega^2 = k\rho^{-1} (\rho g + \alpha k^2 - 4\pi e^2 n_s^2 k).$$

The latter can be written in the form

$$\omega^2 = \frac{\alpha}{\rho} k \left\{ (k - k_0)^2 - 2kk_0 \frac{n_s^2 - n_s^2 \alpha}{n_s^2 \alpha} \right\}, \quad (17)$$

where

$$2\pi e^2 n_s^2 \alpha = (\rho g \alpha)^{1/2}, \quad k_0 = (\rho g / \alpha)^{1/2}.$$

In other words, the charged surface of the helium turns out to be absolutely unstable for  $n_S > n_{S cr}$ . This property is characteristic for the general expression (16).

The supposition was put forth in<sup>[6]</sup> that a superstructure with dimensions of the order of the capillary constant develops on the surface of the helium, with gradual increase of  $n_S$  to the value  $n_{S cr}$ . The assumption made there on the smoothness of such a transition is in error. In order to understand the situation, we attempted to construct a solution for the distorted surface in the neighborhood  $|n_S - n_{S cr}| \ll n_{S cr}$ , assuming this distortion  $\eta(x, y)$  to be small, and the period of the structure to be close to  $2\pi/k_0$ . The macroscopic approach<sup>[11]</sup> can be applied to the problem. Assuming, as above, that the potential difference between the charged helium surface and the metal plate B (see the figure) is given, one must solve the Laplace equation

$$\nabla^2 \varphi = 0$$

in the gap  $-h < z < 0$ . Under the assumption  $k_0 h \gg 1$ , we obtain the general expression in the form

$$\varphi(x, y, z) = 4\pi e n_s z + 4\pi e n_s \sum_k \varphi_k \exp(ikr + kz). \quad (18)$$

The boundary condition that the potential be constant along the surface,

$$\eta + \sum_k \varphi_k \exp(ikr + k\eta) = 0 \quad (19)$$

allows us to express  $\varphi_k$  in terms of the Fourier component of the shape of the surface  $\eta_k$ . The helium pressure is equal to

$$P = -\rho g \eta + \alpha \frac{\nabla^2 \eta + [(\nabla \eta) \nabla]^2 \eta}{[1 + (\nabla \eta)^2]^{3/2}} - \frac{E^2}{8\pi} \quad (20)$$

In order to find the amplitude of the structure if, as is assumed in<sup>[6]</sup>, the latter develops continuously in the region  $|n_S - n_{S cr}| \ll n_{S cr}$ , we must carry out an expansion in (18) and (19) in terms of third order in  $\eta_k$ . However, it can easily be established that (20) contains nonvanishing terms of second order:

$$P = -\rho g \eta + \alpha \nabla^2 \eta + 4\pi e^2 n_s^2 k_0 \eta - \sum_{\mathbf{k}_1, \mathbf{k}_2} [2k_0 |\mathbf{k}_1 + \mathbf{k}_2| - k_0^2 - \mathbf{k}_1 \mathbf{k}_2] \eta_{\mathbf{k}_1} \eta_{\mathbf{k}_2} \exp[i(\mathbf{k}_1 + \mathbf{k}_2) \mathbf{r}] - O(\eta^3). \quad (21)$$

(Everywhere  $\eta$  is measured from the level of the liquid in the presence of a surface charge).

For example, we assume that the developing instability has the form

$$\eta = \eta_0 F(x, y), \\ F(x, y) = \cos k_0 y + \cos [k_0 (\sqrt{1/2} y + \sqrt{1/2} \sqrt{3} x)] + \cos [k_0 (\sqrt{1/2} y - \sqrt{1/2} \sqrt{3} x)] \quad (22)$$

(the symmetry of a hexagonal prism). Substituting (22) in (21), we get

$$P = 4\pi e^2 n_s^2 \alpha F(x, y) \left\{ (n_s^2 - n_{S cr}^2) k_0 \eta_0 / n_{S cr}^2 - \sqrt{1/2} (k_0 \eta_0)^2 - O(k_0 \eta_0)^3 \right\}.$$

Therefore, the equation  $P = 0$  always has small solutions ( $k_0 \eta_0 \ll 1$ ) but, similar to the case of first-order phase transitions, there is physical meaning only if  $\eta_0 = 0$  or  $k_0 \eta_0 \sim 1$ . Thus, the stability of the plane surface is disrupted by a jump at  $n_S$  on the order of  $n_{S cr}$  (17). In order to find the form of the produced state, the considerations given above are inadequate. It is possible that for  $n_S$  larger than some  $n_{S cr}$ , a periodic shape of the surface is actually produced jumpwise. However, another possibility would be that part of the electrons withdraw into the volume. This would correspond to the existence of a certain limiting charge, above which it is impossible to charge the helium surface. Experiment here is as yet indecisive.

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<sup>1</sup>In the usual experimental setup, [<sup>3-5</sup>] the electrons reach the surface from the gas discharge over helium as a consequence of the potential difference. It is understood that as soon as this potential difference is compensated, the transport of electrons ceases.

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<sup>11</sup>L. D. Landau and E. M. Lifshitz, *Mekhanika sploshnykh sred* (Mechanics of Continuous Media) Gostekhizdat, 1954.

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