Mixed scenario of the charged liquid surface reconstruction
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Abstract

Discussed in the paper is a mixed scenario of the charged liquid surface reconstruction in the situation where the 2D surface charge density is close to its saturation value.

One of the instabilities studied in classical hydrodynamics is the Frenkel-Tonks (FT) instability [1,2] arising in a threshold manner on the charged liquid surface and resulting in its deformation. In contrast to other known instabilities, such as the Rayleigh instability of a cylindrical jet [3], von Karman trace generated by moving cylinder (sphere) [4], Taylor vortex instability of a viscous layer between two co-axial rotating cylinders [5], etc., the FT decomposition process can be halted a new metastable state with finite corrugation depth can be formed (charged liquid surface reconstruction). The surface reconstruction was studied by different authors at different times [6-19]. However, by the present time a consistent comprehensive picture of the instability development is only available for small values of the parameter \( \nu \ll 1 \)

\[
\nu = n_s/n_s^{\text{max}}, \quad n_s^{\text{max}} = \sqrt{\alpha \rho g},
\]

(\( \rho \) and \( \alpha \) are the liquid density and surface tension, \( g \) is the gravity acceleration, \( \kappa = \sqrt{\rho g/\alpha} \) is the capillary length), determining the liquid boundary population by localized charges. Explicitly manifested here is the dual nature of the FT instability development: the spectral scenario where the frequency imaginary part changes its sign as a function of external parameters or, alternatively, or the surface corrugation arises due to fluctuations. Under these conditions it is possible to draw the phase diagram branches in the \((\nu, E^-)\) plane operating with the concepts used in the physics of the 1-st order phase transitions: spinodal, binodal, etc. [20]. Here \( E^- \) is the electric field above the charged liquid surface. At the \( \nu = 1 \) the electric field is \( E^- = 0 \).

For finite values \( \nu \leq 1 \) available in the theory are only isolated facts which do not form any coherent picture. For example, it is easy to extend the the spinodal in the \((\nu, E^-)\)-plane into the \( \nu \leq 1 \) domain. Here the alternative part of the problem, the nucleation-controlled “decomposition” of a flat charged surface (the binodal) is left in the uncertain state (there are no typical scale for the charge involved into formation of a dimple). On the other hand, the experiment unequivocally indicates a periodic reconstruction [18,19]. This difficulty is lifted if one considers the FT instability temporal evolution. Authors of Refs. [21,22] noted that the spinodal scenario of the surface decomposition with a fixed total charge leads to the appearance of a periodic array of neutral spots on charged surface. This behavior obviously means the end of spino- and the beginning of binodal evolution with well defined initial conditions: the charge per arising dimple is known
and all the dimples are identical. We refer to the scenario beginning with the “spino-” regime and continuing with the “bino-” evolution of the charged surface deformation as the mixed one. This scenario reproduces in a qualitatively correct way basic known properties of the FT instability in the $\nu \leq 1$ domain: periodicity of the reconstruction, loss of equipotentiality along the corrugated liquid surface, and jump-like behavior of the corrugation amplitude when the stationary reconstructed liquid boundary is formed.

One should emphasize that the mixed decomposition scenario of two-component systems is a rule rather than exception, the point that has not yet been fully taken advantage of. Consider, for example, the phase diagram in the vicinity of its extremum for a two-component solution [23]. Standard system transition from a “stable” point $a$ to unstable point $b$ by a jump of temperature initiates the onset of the spinodal decomposition in the solution along the $c - b - c_1$ isotherm. In this process the system undergoing decomposition in the “directions” from $b$ to $c, c_1$ approaches the spinodal-binodal boundary from the inside of the spinodal zone rather than from the uniform state which takes place in the standard transitions of the $d - e$ type. Starting from the $c^* - c_1^*$ points the evolution becomes binodal one characterized by the spatially modulated stratified density arising at the end of the spinodal decomposition phase (just like in the above reconstruction problem) rather than by a random nucleation-controlled density.

Turning back to the charged liquid surface, let us formulate a number of arising problems. First of all, it is highly desirable to make sure that the stationary “dimpled” state in energetically more favorable than the unstable flat state (experimentally, this is quite obvious since no deformation would otherwise be observed). one should also estimate the charge localization degree in the sense of the inequality

$$\kappa R \leq 1,$$

where $R$ is the single charged spot radius.

1. In the standard flat capacitor geometry with the distance $h$ between the metal electrodes and the liquid film of thickness $d$ covered by a charged 2D system characterized by the number density $n_s$ the problem of finding the surface properties is assumed to be “bulk-like” if $h > d >> a$, where $a = \kappa^{-1}$ is the so-called capillary length. In addition, we are only interested in the saturation case where the electric field $E_-$ above the liquid is zero while the density $n_s$ has its maximum possible value, i.e. $\nu = 1$. Under these conditions the electric field $E_{+}^\text{max}$ in the liquid film also reached its maximum value equal to

$$E_{+}^\text{max} = 4\pi e n_s^\text{max}$$

where $\nu$ is taken from (1).

It is convenient to compare the system energy in different states (flat $\bar{W}$ and corrugated $\tilde{W}$) by first considering the collective effects and then calculating the contribution of a single Wigner-Seitz cell. There are two collective effects. The first one is the gain in Coulomb energy $\Delta w_c$ due to charge ordering. This contribution plays the major role in Coulomb crystallization on flat substrates and is estimated to be [24]

$$\Delta w_c \simeq -1.1 Q^2 / a$$

where $Q$ is the elementary charge.
where $Q$ is the single dimple charge and $a$ is the lattice spacing.

The second effect is specific to the charged liquid surface. A continuous electron disk of radius $R$, pressed by the electric field $E_{\text{max}}$ to the liquid surface squeezes some liquid into the neutral neighborhood of its perimeter. This effect raises the net electron system energy by $\Delta W_\xi$ (see Appendix). The same disk driven to the lattice state displaces a different amount of liquid (actually, it will be shown below that in that case no liquid is displaced at all). Therefore, the surplus energy $\Delta W_\xi$ favors the charged surface reconstruction.

In terms of $\Delta w_c$ and $\Delta W_\xi$ the general requirement

$$\bar{W} > \tilde{W}$$

is written as

$$\bar{w}_c + \Delta W_\xi > \tilde{w}_c + n\tilde{w}_l, \quad \bar{w}_c - \tilde{w}_c = n\Delta w_c, \quad \Delta W_\xi = n\Delta w_\xi. \quad (5)$$

Here $\bar{w}_c$ is the total Coulomb energy of a capacitor with a flat charged liquid layer, $\tilde{w}_c$ is the same energy in the presence of a cluster lattice, $w_l$ is the total energy of a single multiply charged dimple, $n > 0$ is the total number of such dimples arising in the reconstruction process. When writing Eq. (5) we have assumed that in the mixed reconstruction the corrugation possesses the following property: each cell of the arising structure contains a single multiply charged dimple with a charged core of radius $R$ such that $a/R \gg 1$ and the array of dimples has a spatial period comparable to the capillary length. This means that the inequality (5) we are interested in is satisfied if

$$\tilde{w}_l < \Delta w_c + \Delta w_\xi. \quad (6)$$

To estimate $w_l$ one could employ the available results [22] on the properties of individual dimples. In that case the energy $w_l$ becomes zero at

$$E_{\text{crit}} = 1.146 E_{\text{max}}$$

which is qualitatively quite acceptable. However, the product $\kappa R$ in that case yields $\kappa R \simeq 1.5 > 1$, making the single dimple approximation completely meaningless in the calculations of $w_l$ for mixed reconstruction. Here the problem should be reformulated from the very beginning taking into account the finite size of the cell containing a single charged spot.

The appropriate approach is given by the Wigner-Seitz (WS) approximation with the external cell radius of the order of $a$ and the charged spot radius (actually, there exist two alternative distributions differing by whether charged or empty spots occupy the central part of the WS cell; qualitative estimates given in the Appendix favor the charged central spots). In that case the following set of equations is to be solved:

$$eE_{\perp}(r) + 2\pi e^2 \int \frac{n(r')r'dr'}{|r-r'|} = \lambda_e \quad (7)$$

$$\Delta \xi - \kappa^2 \xi - P_{el}(r)/\alpha = \lambda_\xi, \quad P_{el}(r) = eE_{\perp} n(r) \quad (8)$$
\[ \frac{\partial \xi}{\partial r} \bigg|_{r=a} = 0, \quad 2\pi \int_0^a r dr \xi(r) = 0 \]  
\[ 2\pi \int n(r) r dr = N, \quad N = \pi a^2 n_s \]  
(9)

Here \( \xi(r) \) is the self-consistent helium surface deformation, \( \kappa = \sqrt{\rho g/\alpha} \) is the liquid capillary length, \( P_e(r) = eE_\perp n(r) \) is the electron pressure on the helium surface, \( E_\perp \) is the electric field pressing charges towards the liquid surface (in our case, according to Eq. (3), \( E_\perp = E_\perp = 4\pi e n_s^{\text{max}} \)), \( \lambda_e \) is the Lagrange multiplier accounting for the fixed total charge (10). Physically, this multiplier is equivalent to \( \tilde{w} \) (6). \( \lambda_\xi \) is another Lagrange multiplier ensuring the total liquid volume conservation (9).

Assuming the electron spot radius \( R \) to be sufficiently small, \( (\kappa R < 1) \), it is convenient to represent the surface deformation \( \xi(r) \) in the central part of the cell as a series

\[ \xi(r) \approx \xi(0) + \frac{1}{2} \xi''(0) r^2 + \ldots \]  
(11)

In that case, by analogy with the contact Hertz problem [25]

\[ n(r) = \frac{3N}{2\pi R^2} \left( 1 - \frac{r^2}{R^2} \right)^{1/2} \]  
(12)

\[ E_\perp \xi''(0) = \frac{3}{2} Ne \int_0^\infty \frac{ds}{(R^2 + s)^2 \sqrt{s}} \]  
(13)

\[ \lambda_e - eE_\perp \xi(0) = \frac{3}{4} Ne^2 \int_0^\infty \frac{ds}{(R^2 + s)^{3/2}} \]  
(14)

In addition, in the central part of the cell the Eq. (8) becomes

\[ \xi''(0) - \kappa^2 \xi(0) - \lambda_\xi = 3QE_\perp / (4\pi R^2 \alpha), \]  
(15)

Relations (13) and (15) contain there dimple characteristics: \( \xi''(0), \, R, \, \xi(0) \). To close this set of equations, one should add a definition of \( \xi(0) \), i.e. the solution of (8) which can be obtained by expanding \( \xi(r) \) into a series in appropriate Bessel functions. By writing

\[ \xi(r) = \tilde{\xi} + \tilde{\xi}(r), \quad \tilde{\xi} = -\lambda_\xi, \]  
(16)

and choosing the functions

\[ J_0(\mu_n r/a), \quad \frac{\partial J_0(\mu_n r/a)}{\partial r} \bigg|_{r=a} = 0 \]  

as a basis set so that to automatically satisfy the first boundary condition in (9) one can obtain to solution to Eq. (8) as a series

\[ \xi(r) = \bar{\xi} + \sum_{n=1}^\infty \xi_n J_0(\mu_n r/a), \]  
(17)
\[ \tilde{\xi}_n = -\frac{Af(\mu_n, R/a)}{1 + \mu_n^2 J_0^2(\mu_n)}, \quad f(x) = \frac{\sin x - x \cos x}{x^3}, \quad A = \frac{3QE_\perp}{\pi \alpha}. \]

The quantity \( \tilde{\xi} \) here is obtained from the second boundary condition (9) and proves to be zero.

Analysis of the equation set (13), (15), (17) allows one to suggest (see arguments cited in the Appendix) that at the initial stage of the dimple reconstruction development the quantity \( \xi(0) \) does not play any significant role in these equations. Hence, the relationships between \( \xi''(0) \) and \( R \) immediately yield a closed equation set from which they can be easily found:

\[ E_\perp \xi''(0) = \frac{3\pi Q}{4R^3}, \quad R = \frac{\pi^2 \alpha}{E_\perp}. \]  

(18)

Here the quantity \( \xi(0) \) is obtained from Eq. (17) with \( R \) calculated from Eq. (18).

The final stage in the estimation of the mixed scenario feasibility is the estimation of parameters which are most critical for its realization. By setting in the general formulas (7)-(18) \( E_\perp = E_+ = 4\pi en_s^{\text{max}} \), one has

\[ \frac{R_{\text{crit}}}{a} \simeq 0.4 < 1, \quad \frac{\xi_{\text{crit}}(0)}{a} \simeq 0.3 < 1, \quad \lambda_{\text{crit}} \equiv \tilde{w}_l^{\text{crit}} < \Delta w_c^{\text{crit}} + \Delta w_\xi^{\text{crit}} \]  

(19)

Here \( n_s^{\text{max}} \) is taken from (1) and \( \tilde{w}_l \) from (6).

Estimates (19) reveal that the mixed scenario of the charged liquid helium surface reconstruction seems to be rather acceptable in the vicinity of the filling factors (1) close to unity. In the present paper, it is impossible to obtain more definite conclusions since the employed approximation (B-3) cannot claim any quantitatively correct statements. We do not see any other approaches (different from the mixed scenario) which could proved at least qualitative explanation of the observed periodic reconstruction of the liquid helium surface in the range of \( \nu \leq 1 \).

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Appendix

1. Let $R_s$ be the charged area radius and $L > R_s$ the radius of entire liquid surface between vertical walls. The equilibrium of the cell as a whole is defined by the following equations:

\[
\rho g \xi_0 + P_{el} = \rho g \xi_1, \quad (P1)
\]

\[
R_s^2 \xi_0 + (L^2 - R_s^2) \xi_1 = 0, \quad L > R_s. \quad (P2)
\]

Here $\xi_0$ is the liquid surface deformation in the charged area, $a$, $\xi_1$ is the the liquid surface deformation outside the electron disk, and $P_{el}$ is the effective pressure in the charged area. Finding from $(P2)$ the quantity $\xi_1$

\[
\xi_1 = -\xi_0 R_s^2/(L^2 - R_s^2)
\]
and substituting it into (P1) one has
\[
\xi_0 = -P_{el}/g^*, \quad g^* = g(1 + \frac{R^2}{L^2 - R^*}) \quad (P3)
\]

The additional energy \(\Delta W_\xi\) due to deformation \(\xi_0\) has the scale
\[
\Delta W_\xi \simeq \pi R^2 P_{el} \xi_0 \quad (P4)
\]

Combining all the necessary definitions one obtains from (P4)
\[
\Delta W_\xi = n\Delta w_\xi, \quad \Delta w_\xi = \frac{4\pi \alpha a^2}{f(R_*, L)}, \quad f(R_*, L) = (1 + \frac{R^2}{L^2 - R^*}). \quad (P4)
\]

This expression is used for estimates in the main text of the paper.

2. Within the alternative including either charged or empty spots, it is convenient to start from qualitative estimates. Suppose we consider a cylindrical Wigner-Seitz model with the spot external radius of the order of \(a\), charged spot radius \(R < a\), and charge-free ring with area \(\pi(a^2 - R^2)\). If the last formula is interpreted as an integral relation, one can estimate within the cylindrical model the probability of scenario described in Ref. [21] where the initial stage of the reconstruction is assumed to be represented by a periodic array of empty (charge-free) spots whose area is different from zero to the extent of positive supercriticality \(\delta E = (E - E_c^-)\),
\[
\delta E = (E_- - E_c^-). \quad (P5)
\]

However, this is most probably not so because of the following reasons.

Within the WS cell both the local equilibrium conditions and liquid volume conservation should be satisfied, just as in (P1). Then, by similar arguments one has
\[
\xi_0 = -P_{el}/g^*, \quad g^* = g(1 + \frac{R^2}{a^2 - R^2}) \quad (P6)
\]

According to (P6), development of a stationary corrugation under the conditions \(\pi(a^2 - R^2) \to 0\) (empty spots with low area) is unlikely since in that domain the effective value of \(g^* \to \infty\). In that case the appearance of corrugation, i.e. growth of \(\xi_0\) (P6) is hindered.