ON PHASE TRANSITIONS IN THE SYSTEM OF CHARGED FERMI PARTICLES ABOVE THE LIQUID DIELECTRIC SURFACE

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A consistent theory of equilibrium states of charges and fields above the surface of liquid dielectric film located on a solid substrate is built. The developed approach is used for the description of phase transitions in these systems to the states with spatially-periodic ordering.

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1. INTRODUCTION

As it is well-known [1, 2] in the system of electrons above liquid dielectric surface under certain conditions the spatially periodic structures of two-dimensional crystal type or so-called dimple crystals appear. Theoretical approaches to the description of these structures are based so far on using the model potentials that contain adjustable constants. In addition the existing theoretical works consider a many-particle system of charges near the surface of a liquid dielectric as two-dimensional structure [1], ignoring their distribution above the surface of helium. The question of constructing a general theory without these shortcomings arises.

The present paper is devoted to developing the approach to the description of distributions of the same sign charges and fields above the liquid dielectric surface in terms of considerably generalized by us Thomas-Fermi model [3]. As an application of this approach we consider some phase transitions in such system associated with the transformation of the liquid dielectric surface and the formation of spatially periodic structures.

2. THE BASIC CONCEPTS OF THE VARIATIONAL PRINCIPLE

Let us consider a system of identical particles with charge $Q = -e$ (e is the charge of electron), mass $m$, spin $S_2$, momentum $p$ and energy $\varepsilon_p = \frac{p^2}{2m}$. The charges are located in vacuum above the surface of liquid dielectric film which has thickness $d$ and permittivity $\varepsilon$. We assume that the liquid dielectric film is located on a flat solid substrate with dielectric permittivity $\varepsilon_d \gg \varepsilon$. The surface profile of the liquid dielectric film is described by a function $\xi(\rho) \equiv \xi(x, y)$, where $\rho \equiv \{x, y\}$ is the position vector in the plane $z = 0$ of a Cartesian coordinate system $\{z, x, y\}$. The interface regions “1” - “3” in the direction of the coordinates $\rho \equiv \{x, y\}$ are considered unlimited.

We consider that the charges are attracted by external electric field $E$ directed along the axis $z$. We also assume that there is a potential barrier prohibiting the penetration of charges into the liquid dielectric film. All physical quantities relating to the region $z > \xi(\rho)$ are denoted by the subscript “1”, the physical quantities related to the liquid dielectric film $\xi(\rho) > z > -d$ - index “2”, and the physical quantities related to the solid dielectric substrate $z < -d$ - index “3”. In the region “1” the system is fully described by a distribution function of particles $f_p(r)$, the potential of the electric field $\varphi_i (r)$ created by a system of charged particles, the potential of attracting external electrostatic field $\varphi^{(e)} (r)$ and the surface profile of liquid dielectric $\xi(\rho)$. In the region “2” the characteristics of the system are the surface profile of a liquid dielectric $\xi(\rho)$ and the total electric potential. By the “total” we mean the potential of the external electric field in a liquid dielectric and the field induced by charged particles located in the region “1”. The region “3” is characterized by the total potential of the electric field in a solid substrate.

The basis of the variational principle is the concept of the thermodynamic potential $\Omega$

$$\Omega = -S + Y_0 E + Y_1 P_i + Y_1 N, \quad (1)$$

where $S = S(f, \xi)$ is the entropy of the system

$$S = -\gamma^{-1} \int d^3 r dp \{ \gamma f_p (r) \ln (\gamma f_p (r)) +$$

$$+ (1 - \gamma f_p (r)) \ln (1 - \gamma f_p (r)) \} , \quad \gamma = \frac{g}{(2\pi \hbar)^3}, \quad (2)$$

$P_i = \int d^3 r dp f_p (r) p_i$ is the total momentum of the

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particles, \( N = \int d\textbf{r} d\textbf{p} f_p (\textbf{r}) \) is the total number of particles in the system and \( g = (2\sigma) + 1 \).

Thermodynamic parameters \( Y_0, Y_1, Y_4 \) in (1) are associated with temperature, average velocity \( v_i \) and chemical potential \( \mu \) by formulæ \( T = 1/Y_0, -Y_1/Y_0 = v_i, -Y_4/Y_0 = \mu \). We note that in the case of stationary system the velocity \( v_i \) is zero (we shall consider this case further on \( v_i = 0 \)).

The energy of the system \( H \) that functionally depends on the above parameters describing the system \( H = H (f, \varphi, \xi) \) has the following form:

\[
H = H_1 + H_2 + H_3 + H_4,
\]

where \( H_1, H_2, H_3 \) determine the energy of the system in regions “1” - “3”, and \( H_4 \) is the energy of the perturbed liquid dielectric:

\[
H_1 = \int d\rho \int_\xi d\varphi \int d\textbf{r}_p f_p (\textbf{r}_p) \varepsilon_p + \frac{Q}{2} \int d\rho \int d\varphi \int d\textbf{r}_p \varphi_1^{(i)} (\textbf{r}_p) f_p (\textbf{r}_p) + Q \int d\rho \int d\varphi \int d\textbf{r}_p \varphi_1^{(c)} (\textbf{r}_p) f_p (\textbf{r}_p) + \frac{1}{8\pi} \int d\rho \int d\varphi \int_\xi d\textbf{r}_p \left( \nabla \varphi_1^{(c)} (\textbf{r}_p) \right)^2,
\]

\[
H_2 = \frac{\varepsilon_4}{8\pi} \int d\rho \int d\varphi \int d\textbf{r}_p \left( \nabla \varphi_2 (\textbf{r}_p) \right)^2,
\]

\[
H_3 = \frac{\varepsilon_4}{8\pi} \int d\rho \int d\varphi \int d\textbf{r}_p \left( \nabla \varphi_3 (\textbf{r}_p) \right)^2,
\]

\[
\varphi_2 = \varphi_1^{(i)} + \varphi_1^{(c)}, \quad \varphi_3 = \varphi_3^{(i)} + \varphi_3^{(c)},
\]

\[
H_4 = \frac{\alpha}{2} \int d\rho \int d\textbf{r}_p \left( \nabla \rho \xi (\rho) \right)^2 + \kappa^2 \varepsilon_2 (\rho),
\]

\[
dS = d^2 \rho \sqrt{1 + (\nabla \xi (\rho))^2}
\]

The external forces acting on the liquid dielectric film with thickness \( d \) are the gravitational force, as well as van der Waals forces from the solid substrate. Thus (see, e.g., [3]), the constant \( \kappa \) in (4) is given by \( \kappa^2 = \frac{\alpha}{\sigma} (g + f) \), where \( g \) is the acceleration of gravity, \( \alpha \) is the surface tension of liquid dielectric, \( \rho \) - its density and \( f \sim d^{-4} \) is Van der Waals constant, which in the case of a massive liquid dielectric \( d \rightarrow \infty \) can be neglected in comparison with \( g \). In the case of a thin dielectric film the force of gravity acting on the atoms of liquid dielectric become negligibly small comparing to the van der Waals forces.

The variational the problem is reduced to finding the minimum of the thermodynamic potential \( \Omega \) under the condition of the Poisson equation validity,

\[
\Delta \varphi_1^{(i)} (\textbf{r}) + 4\pi Q \int d\textbf{r} f_p (\textbf{r}) = 0,
\]

in the region “1” \((z > \xi (\rho))\). In the regions “2” and “3” there are no charges hence for these regions the equation (5) becomes the well-known Laplace equation

\[
\Delta \varphi_2^{(c)} (\textbf{r}) = 0, \quad \Delta \varphi_3^{(c)} (\textbf{r}) = 0.
\]

We also note that the attractive potential of the external electric field satisfies the same equation in all three regions

\[
\Delta \varphi_1^{(c)} (\textbf{r}) = 0, \quad l = 1, 2, 3.
\]

As a result, we obtain the final form of self-consistency equations relating the parameters describing the system under consideration. The equations for the potentials of the electric field, both external and induced by charges in all three regions of the system have the form (5) - (7). The self-consistency equation for \( \xi (\rho) \) has the following form:

\[
Y_0^{-1} \gamma^{-1} \int d\textbf{r} \ln (1 - \gamma f_p (\textbf{r}))|_{z = \xi (\rho)} = \frac{\varepsilon}{8\pi} \left( \nabla \varphi_2 (\textbf{r}) \right)^2 - \left( \nabla \varphi_2^{(c)} (\textbf{r}) \right)^2 \right)_{z = \xi (\rho)} + \alpha^2 \xi (\rho) \sqrt{1 + (\nabla \xi (\rho))^2} - \alpha \nabla \left( \frac{\nabla \xi (\rho) \left( 1 + \kappa^2 \xi^2 (\rho) + \frac{3}{2} (\nabla \xi (\rho))^2 \right)}{\sqrt{1 + (\nabla \xi (\rho))^2}} \right),
\]

and the distribution function \( f_p (\textbf{r}) \) in (5) and (8) is given by the expression

\[
f_p (\textbf{r}) = \theta (z - \xi (\rho)) \gamma^{-1} \times \{1 + \exp Y_0 (\varepsilon_p - \mu + Q \varphi)\}^{-1}.
\]

It is easy to see that the obtained system of self-consistency equations is closed. However it must be supplemented by the boundary conditions for the potentials of electric fields and their tensions at the boundaries \( z = \xi (\rho) \) and \( z = -d \) accounting the limitation of fields at infinity. To simplify the calculations we consider that the surface charges at the interface regions are absent (for details see [3]).

3. THE MAIN EQUATIONS FOR THE PARAMETERS DESCRIBING THE SYSTEM IN ASYMMETRIC PHASE

The scenario of the phase transition leading to the transformation of the liquid dielectric surface film is assumed as follows. As already mentioned the external electric field attracting the charges to the surface leads to the subsidence of the surface of flat liquid dielectric film in the region of the field acting. And the bottom of the deflection remains flat. Hence the deformation of the surface of liquid dielectric leaving the bottom of the deflection flat can be characterized by a single parameter \( \xi < 0 \) (the subsidence depth). With further increasing of the external electric field the depth \( \xi < 0 \) increases and the surface of the deformation bottom remains flat up to a certain critical
value of the total electric field $E_0$ on the dielectric surface. We note that at the same time the condition $|\tilde{\xi}| < d$ must be satisfied if the liquid dielectric is a film with thickness $d$ located on the solid substrate.

With further increasing of attracting electric field the surface profile of the bottom of the formed deflection may deform and acquire a periodic structure. Hence it follows that the phase transition to a spatially periodic structures in the considered system occurs on a background of the flat structure of a liquid dielectric.

According to (5) - (9) the above scenario of the phase transition the surface profile of liquid dielectric in a phase with lower symmetry can be written as follows:

$$\xi(\mathbf{r}) = \bar{\xi}(\mathbf{r}) + \tilde{\xi}(\mathbf{r}), \quad |\tilde{\xi}| \gg |\bar{\xi}(\mathbf{r})|,$$  \hspace{1cm} (10)

where $\tilde{\xi}(\mathbf{r})$ is a spatially inhomogeneous surface profile resulting from the phase transition on the background of a flat bottom surface $z = \bar{\xi}$ of the described above homogeneous deformation. Thus, the surface profile $\xi(\mathbf{r})$ is the order parameter of the phase transition turning into zero at the critical point.

According to (10) we can expect that distribution of charges and fields in the system will be little different from the distributions taking place in the case of a flat dielectric surface $z = \bar{\xi}$. Then the field potentials $\varphi_l(z, \mathbf{r})$, $l = 1, 2, 3$ can be written as

$$\varphi_l(z, \mathbf{r}) = \tilde{\varphi}_l(z) + \tilde{\varphi}_l(z, \mathbf{r}), \quad |\tilde{\varphi}_l(z)| \gg |\tilde{\varphi}_l(z, \mathbf{r})|,$$  \hspace{1cm} (11)

where $\tilde{\varphi}_l(z, \mathbf{r})$, $l = 1, 2, 3$ are the total electric field potentials in all regions of the system described above (but not on the surface interface!) in the case when the surface of the liquid dielectric is flat $z = \bar{\xi}$. Potentials $\tilde{\varphi}_l(z, \mathbf{r})$ describe small distortions of the field in all these regions due to inhomogeneous surface profile $\xi(\mathbf{r})$. We also note that the representation similar to (11) will be considered valid for the potentials of the external electric field in all three regions

$$\tilde{\varphi}_l^e(z, \mathbf{r}) = \tilde{\varphi}_l^e(z) + \tilde{\varphi}_l^e(z, \mathbf{r}), \quad |\tilde{\varphi}_l^e(z)| \gg |\tilde{\varphi}_l^e(z, \mathbf{r})|.$$  \hspace{1cm} (12)

The method of solving equations (5) - (9) accounting boundary conditions in the case of non-degenerate gas of charges above a plane dielectric surface is described in detail in [3]. Here we present only the final results.

The dependence of the electric field $E_l(z) = -\partial \mathcal{V}(z)/\partial z$ and charge density on coordinate above the liquid dielectric surface $z = \bar{\xi}$ is given by the expressions

$$E_l(z) = \frac{1 + \chi(z)}{1 - \chi(z)} \cdot n(z) = \beta \frac{E_0^2}{\varepsilon} \cdot \frac{4\chi(z)}{8\pi (1 - \chi(z))^2} \cdot \chi(z) \equiv \frac{E_0 - E}{E_0 + E} \cdot \exp \left\{ - (z - \bar{\xi})/z_0 \right\}.$$  \hspace{1cm} (13)

where $z_0 \equiv (\beta e E)^{-1}$, $\beta^{-1} = T$, $E_0 - E = 4\pi e n_s$ and $n_s$ - the number of volume charges per unit of a plane dielectric surface. Equation (8) allows us to identify the subsidence of the flat surface level $\xi$

$$\bar{\xi} = -\varepsilon + \frac{1}{8\pi \varepsilon \alpha x_c} (E_0^2 - E^2).$$  \hspace{1cm} (14)

Let us note that in the absence of charges the value of $\bar{\xi}$ is zero as expected.

4. CRITICAL PARAMETERS OF THE PHASE TRANSITION TO A SPATIALLY PERIODIC PROFILE OF THE LIQUID DIELECTRIC SURFACE

As a result of rather cumbersome calculations the equation for the Fourier transform $\tilde{\varphi}(\mathbf{q})$ of the spatially periodic surface profile $\xi(\mathbf{r})$ in the linear approximation of perturbation theory (see (10), (11)) can be reduced to the form:

$$\Phi(\mathbf{q}_0) \tilde{\varphi}(\mathbf{q}_0) = 0, \quad \Phi(\mathbf{q}_0) =$$

$$= \frac{q_0 (E_0^2 - E^2)}{4\pi \varepsilon} \cdot \frac{C^2 f_1 + C f_2 + f_3}{C^2 g_1 + C g_2 + g_3} - \alpha (\kappa^2 + q_0^2),$$

where we introduce the following notations

$$f_1 \equiv (\varepsilon - 1) \cdot \left( - (\varepsilon - 1) (2x)^2 + 2x + (\varepsilon + 1) y \right),$$

$$f_2 \equiv 2 \varepsilon \cdot \left( - (\varepsilon - 1) (2x)^2 + \varepsilon (2x + 1) + y \right),$$

$$f_3 \equiv - (\varepsilon^2 - 1) (2x)^2 + 2x (2x^2 + 3x + 1) + 2\varepsilon (\varepsilon + 2) - y (\varepsilon^2 + 2x - 1),$$

$$g_1 \equiv (\varepsilon - 1) \cdot (\varepsilon - 1) (2x (2x + 1) - y),$$

$$g_2 \equiv 2 \cdot ((\varepsilon^2 - 1) 2x (2x + 1) - y),$$

$$g_3 \equiv (1 + \varepsilon) \cdot (1 + \varepsilon) 2x (2x + 1) + y,$$

$$C = (\varepsilon_0 - \varepsilon) (\varepsilon_0 + \varepsilon)^{-1} \exp \left( -2q (d + \bar{\xi}) \right),$$

$$x \equiv q_0 T/\varepsilon E_0, \quad y \equiv (E_0^2 - E^2)/E_0^2.$$  \hspace{1cm} (15)

It follows from (15) that since $\bar{\xi}(\mathbf{q}_0) \neq 0$ (in the opposite case there are no spatially periodic solutions), it must be $\Phi(\mathbf{q}_0) = 0$

$$\frac{q_0 (E_0^2 - E^2)}{4\pi \varepsilon} \cdot \frac{C^2 f_1 + C f_2 + f_3}{C^2 g_1 + C g_2 + g_3} = \alpha (\kappa^2 + q_0^2).$$  \hspace{1cm} (16)

This equation determines the length of vector $\mathbf{q}_0$ as a function of physical parameters of the problem: the temperature $T$, the number of electrons $n_s$ per unit of the flat liquid dielectric surface, the density of the dielectric $\rho$, its surface tension $\alpha$ and permittivity $\varepsilon$ and the permittivity of the solid substrate $\varepsilon_d$. The vector $\mathbf{q}_0$ characterizes the reciprocal lattice of two-dimensional periodic structure inside its unit cell exp(i$\mathbf{q}_0 \mathbf{r}) = \cos (q_0 x) + \sin (q_0 y)$. The condition for the appearance of a new phase - the periodic structures in a system of charged fermions above the surface of the liquid dielectric film is reciprocal lattice vector equal to zero $\mathbf{q}_0 = 0$ at the critical point. If $q_0 = 0$ at the critical point, (17) takes the form:

$$T = \frac{(\varepsilon + 1) e E_0}{8\pi \alpha x_c^2 \kappa^2} (E_0^2 - E^2).$$  \hspace{1cm} (18)
Equation (18) is the equation of the critical surface of the phase transition in three dimensional space.

Near the critical value of the external electric field $E_c$ defined by equation (18), (but $E > E_c$), the value of the vector $q_0$ must be small and can be obtained analytically from (14) assuming $q_0d \ll 1, \varepsilon \ll \varepsilon_d$,

$$
q_0 \approx \kappa \left( \frac{2(E - E_c)}{E_0c} \right)^{\frac{1}{2}} \left( 1 + \frac{(\varepsilon + 1)\pi e^3 n_e^2 E_0c}{\varepsilon^2 a c^2 T_c} \right)^{\frac{1}{2}} \times \left( 1 + \frac{\varepsilon - 1}{\varepsilon + 1} \left( \frac{2T_c^c}{eE_0c} \right)^2 \right)^{-\frac{1}{2}}.
$$

(19)

The index "c" in some of the values in the formula (19) means that these values lie on the surface of the phase transition given by the expression (18).

Far from the surface of the phase transition (17) can be solved only numerically. We make these numerical calculations basing on the experimental data published in [4]. In this paper authors establish the clear observation of the two-dimensional hexagonal hole lattice on the surface of liquid $^4$He with a period $a \approx 1.76$ nm ($a = 2\pi/q_0$), the order of the electron number in each hole about $10^7$, thicknesses of helium film $d$ from 0.2 to 1.9 cm, external field $E = 1820$V/cm and temperature $T = 4.2$ K. We obtain the satisfactory agreement between the calculated and experimental values of the system parameters at $n_e \approx 0.84 \cdot 10^9$ cm$^{-2}$.

Let us also note that in the case of a system of charges above a massive liquid dielectric ($\varepsilon = \varepsilon_d, d \to \infty$) the described phase transition in the investigated system is impossible.

5. CONCLUSIONS

Thus, we constructed a consequent theory of charge systems above the surface of a liquid dielectric film. This theory is based on the model of self-consistent field in Thomas-Fermi approach generalized applying to the considered system. The developed theory allows obtaining the self-consistency equations for the parameters describing the system: the distribution function of charges above the liquid dielectric surface, the electrostatic field potentials in all regions of the system and the surface profile of liquid dielectric. An essential fact is that the theory contains no adjustable constants determined from the experiment. The obtained self-consistency equations allow us to describe the phase transition in such system to a spatially periodic surface profile of a liquid dielectric.

References


