Abstract

Charged clusters in liquid helium in an external electric field form a two-dimensional system below the helium surface. This 2D system undergoes a phase transition from a liquid to a Wigner crystal at rather high temperatures. Contrary to the electron Wigner crystal, the Wigner lattice of charged clusters can be detected directly.

1. The properties of charged particles on the surface and in the bulk of liquid helium have been extensively studied for more than 30 years [1]. An electron on the helium surface has very high mobility due to the high purity of helium at low temperatures, when all outside particles quit the quantum liquid. In the bulk of helium an electron has a low mobility since it forms a bubble of a large radius \( \sim 17\text{Å} \). Negative ions as \( \text{H}^- \) or \( \text{O}_2^- \) in the bulk of liquid helium also attracted some attention [2]. In [3] the surface states of negative ions of large radius (as \( \text{Ca}^- \) and \( \text{Ba}^- \)) were predicted and studied. One interesting point is that the negative ions \( \text{Hg}^- \), \( \text{Be}^- \), \( \text{Zn}^- \), \( \text{Cd}^- \) which do not exist in vacuum may exist in liquid helium. The atoms of these elements have a very great polarizability, and an electron may almost form a bound state with these atoms. In helium an electron is already confined so that these bound states realize.

There are several experimental methods to introduce an outer particle into liquid helium. Atoms, molecules and small clusters are created inside helium by the laser ablation. Larger clusters of radius \( R > 50\text{Å} \) are instilled into helium through its surface [4]. At high concentration of these clusters in helium an iceberg is formed. This was studied experimentally in [5] by injecting clusters of \( \sim 10^3 \) molecules into liquid helium. Large clusters of air or hydrogen were observed as a fog, slowly falling on the bottom of the vessel at speed of \( 10^{-2}\text{cm/sec} \). Below the \( \lambda \)-point of helium the clusters coagulate in the flakes of a “snowfall” [6].

In the present paper we study the properties of large charged clusters in liquid helium. We show that in strong electric field one can create a two-dimensional charged system of heavy particles below the surface of the liquid.
This system may give an opportunity to detect directly its Wigner crystallization as the temperatures decreases.

2. Keeping in mind recent experiments with the crystals of $H_2O$ in helium $^5$, we shall simulate the impurity particle as a macroscopic dielectric sphere of radius $R$. As is well known $^6$, the interaction $V_{eB}(r)$ of an electron with such a sphere at large distance $r > R$ has the form

$$V_{eB} = \frac{eE_{ex} r R^3}{r^3} - \frac{e^2 R^3}{2r^4}, \quad (1)$$

where

$$R^3 = \frac{R^3 \varepsilon_B - \varepsilon_h}{\varepsilon_B + 2 \varepsilon_h}. \quad (2)$$

The first term in (1) is the interaction of an electron with the dipole moment of the cluster $\vec{d} = \vec{E}_{ex} R^3$ created by the external field $\vec{E}_{ex}$. The second term in (1) is due to the polarization of the cluster by the electron. As can be seen from (2) the effective size of the cluster $R_*$ is equal to $R$ if its dielectric constant $\varepsilon_B \gg 1$. Note that the dielectric constant of helium is $\varepsilon_h = 1.054$. If in helium there are some electrons and impurities, the interaction (1) leads to the formation of charged clusters with large binding energy $E_{Be} \sim 0.1 - 1 eV$. The electric field $E_{ex}$ presses these heavy charges to the surface of helium, where the clusters organize into a two-dimensional system. The optimal distance $z_B^{opt}$ of these heavy charged particles to the surface is determined by the minimum of the potential energy:

$$V_{cl}(z_B) = eE_{ex} z_B + \frac{e^2}{z_B} 2 \nu_1 - M g z_B + \frac{(\vec{E}_{ex} R^3)^2}{z_B^3} \nu_1 + V_{vdW}, \quad (3)$$

where

$$\nu_1 = \frac{1}{8} \frac{\varepsilon_h - 1}{\varepsilon_h + 1} \approx 1/300.$$

The first term in (3) is due to the external field. The second term is the image potential of a charge near the surface. The third gravitational term is important only for very big clusters of radius $R > 10^4 \text{Å}$; usually, one can neglect the gravitational term. The fourth term in (3) is the image potential of the electric dipole moment of the cluster. Rigorously, the dipole moment of the cluster $\vec{d}$ is generated both by the external field $\vec{E}_{ex}$ and by the field from the attracted electron. However, the image potential due to electron-generated dipole moment of the cluster is always smaller than the second term in (3) by a factor of $[\varepsilon_B - 1)R/(\varepsilon_B + 1)]z_B^2 < 1/2$. Therefore, for the estimation of $z_B$ we shall take into account only the dipole moment $\vec{d} = \vec{E}_{ex} R^3$ generated by the external field. The last term in (3) is the van der Waals potential. This term prevents the cluster to emerge from helium. It is $\propto 1/z_B^3$ and becomes important only when the cluster is very close to the surface.

Now we have to separate two different cases. First we consider small clusters when $e/R^2 \gg E_{ex}$ or $R \ll \sqrt{e/E_{ex}} \approx 700 \text{Å}$ for a field $E_{ex} = 3000 V/cm$. The
The gravitational field, the dipole image potential and the van der Waals potential are then much weaker than the first two terms in \( (3) \). Therefore, to find the optimal \( z_B \) we shall consider only these two (pure electronic) terms, and \( z_B \) now means the \( z \)-coordinate of the bound electron. Differentiating \( V_{cl}(z_B) \) with respect to \( z_B \) we find

\[
z_{B}^{opt} = \sqrt{\frac{2e}{\nu_1 E_{ex}}}. \tag{4}
\]

For a field of \( E_{ex} = 3000V/cm \) we get \( z_{B}^{opt} \approx 60\text{Å} \). If \( z_{B}^{opt} < R \) the cluster will be close to the surface but it will not emerge from helium because of the short-range van der Waals attraction forces between cluster and helium atoms.

The opposite case of a large cluster (\( e/R^2 \ll E_{ex} \) or \( R^2 \gg \text{(700Å)}^2 \)) is even more interesting. Now we have to consider also the dipole-dipole image potential of the cluster as it increases as \( R^6 \). The optimal distance \( z_{B}^{opt} \) is now determined by the quadratic equation

\[
\frac{dV_{cl}}{dz_B} = \frac{e E_{ex} - e_2}{z_B^2} - 3 \left( \frac{E_{ex} R_s^3}{\nu_1 e} \right)^2 \nu_1 = 0, \tag{5}
\]

which gives

\[
z_{B}^{opt} = \sqrt{\frac{\nu_1 e}{E_{ex}}} \left[ 1 + \sqrt{1 + \frac{3}{\nu_1 \left( \frac{E_{ex} R_s^2}{e} \right)^3} \right]^{1/2} \tag{6}
\]

At \( E_{ex} R_s^2 / e \gg 1 \) this becomes

\[
z_{B}^{opt} \approx \sqrt{\frac{\nu_1 e}{E_{ex}}} \left( \frac{3}{\nu_1} \right)^{1/4} \left( \frac{E_{ex} R_s^2}{e} \right)^{3/4}. \tag{7}
\]

Now the optimal distance \( z_{B}^{opt} \propto (E_{ex})^{1/4} R_s^{3/2} \), and one can reach \( z_{B}^{opt} > R \) by increasing \( E_{ex} \) and \( R \). At \( z_{B} = 80 \approx \infty \) and \( E_{ex} = 5000V/cm \) this condition is fulfilled only for rather huge clusters \( R \geq 5000\text{Å} = 0.5\mu m \).

Above we have argued the possibility of realization of a 2D system of charged clusters below a helium surface. The dipole moment and electric charge of each cluster allow to fix the cluster at a macroscopic distance \( z_0 > R \) below the helium surface. At low helium temperatures any thermal fluctuations of the positions of these heavy charges are small. Let us now consider the Wigner crystallization of these charges.

The interaction of two charged clusters separated by distance \( a \) is described by the potential energy

\[
V_{int}(a) = \frac{e^2}{a} + \frac{(E_{ex} R_s^3)^2}{a^6} \equiv V_e + V_d. \tag{8}
\]

The dipole-dipole part \( V_d \) of the interaction between clusters becomes dominant \((V_d > V_e)\) if \( ea < E_{ex} R_s^3 \). At \( E_{ex} = 5 \cdot 10^4V/cm, R_s = 5000\text{Å} \) and \( a = 10R_s \) (that corresponds to the average cluster density \( n_s = 1/a^2 = 4 \cdot 10^6\text{cm}^{-2} \)) one gets

\[
V_e = 3K; \quad V_d = 300K. \tag{9}
\]
Accepting the empirical criterion $V(a) > 100k_BT$ of melting of 2D Wigner crystal we come to the conclusion of the possibility to observe the liquid-crystal phase transition at rather high temperature $T \sim 3K$. The oddity of the situation in hand is that the crystal lattice is formed by the short-range dipole forces ($V_d \gg V_e$), and the long-range Coulomb interaction appears in the existence of long-wave plasma oscillations. An observation of these effects should be possible by using the method first applied 50 years ago [7]. The neutral clusters come to helium through its surface and go down slowly to the bottom of the helium vessel where the electron source is maintained. The electron from the injector are drawn up by the electric field. In the bulk of helium some of the clusters form a bound with one electron and form a 2D charged system below the surface. Other clusters glue to the bottom of the vessel by the van der Waals forces. The excess electrons will finally make a quantum tunnel transition through helium surface to the vacuum where the electrons have a lower energy. The energy of a big charged clusters is, on the contrary, lower inside the helium because of the short-range van der Waals attraction of the cluster to the atoms of liquid. This gain of the van der Waals energy is roughly proportional to the surface of cluster.

To study the stability of the 2D charged system of clusters one has to consider also their van der Waals attraction to each other at the mean distance $a$. At large distance, the van der Waals potential energy of two atoms is

$$V_{vdW}(a) = \frac{23hc}{4\pi a^7} \beta^2$$

where $\beta$ is the atomic polarizability of clusters. It is related to the dielectric constant $\varepsilon_B = 1 + 4\pi \beta n$, where $n$ is the density of the medium of the clusters. Integration of (10) over the cluster volume gives

$$V_{BB}(a) = \frac{23hcR^6}{36\pi a^7} (\varepsilon_B - \varepsilon_H)^2$$

For a cluster consisting of the molecules of $H_2O$ with $\varepsilon_B \approx 80$, for the parameters $R = 5000\text{Å}$ and $a = 10R$ (the same as in [9]), from (11), we get an estimate of $V_{BB}(a) = 0.6K$. Since $V_{BB}(a) \ll V_d(a)$, there is a wide range of parameters where the 2D system under study is stable against coagulation (sticking together) of clusters.

It is much easier to detect the Wigner lattice of huge charged clusters than that of electrons. The Wigner lattice of electrons allows only an indirect method for experimental studies [9]. On the other hand, the Wigner crystallization of charged clusters can be detected directly by neutron or X-ray scattering. If the charged clusters are very huge ($R \gg 5000\text{Å}$) they can be visually observed.

In closing we note that the considered effects are not very sensitive to the type of quantum liquid. They could also be observed in liquid hydrogen, where $\varepsilon_H = 1.28$ and the parameter $\nu_1 = 1.5 \cdot 10^{-2}$.

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Appendix.
In this appendix we study the structure of the charged cluster in more detail. The attraction of electron to the cluster (at large distance given by eq. (3)) is quite strong so that the binding energy of the electron is large compared to the potential energy (3). The electron is localized at one side of the cluster (if the electron was uniformly distributed along the surface of cluster it would not gain any polarization energy). The mean distance of the electron from the cluster is always much smaller than the cluster radius: $z_e \ll R$.

To estimate the binding energy of the electron and the dipole moment of the charged cluster we shall consider the surface of the cluster near the electron to be flat (the problem of a point-like charge near dielectric sphere cannot be solved in finite form [6]). Then the polarization of the cluster by the electron can be taken into account via the image potential

$$V_{eB} = \frac{e^2 \varepsilon_B - 1}{8z_e \varepsilon_B + 1}$$

which determines the average distance $\overline{z_e}$. This is a one-dimensional Coulomb potential similar to that of an electron above helium surface without external field. Above the helium surface the electron levitates on a distance of $z^h_{he} = 114\AA$ [1]. In the case of an electron near the dielectric cluster we have

$$\overline{z_e} = \frac{\varepsilon_h - 1}{\varepsilon_h + 1} \left( \frac{\varepsilon_B - 1}{\varepsilon_B + 1} \right)^{-1} \overline{z^h_e}.$$ 

For $\varepsilon_B = 80 \approx \infty$, $\overline{z_e} = 0.027\overline{z^h_e} \approx 3\AA \ll R_B$. The corresponding electron-generated dipole moment of the cluster is $d_{e-g} \approx R_B e (\varepsilon_B - 1)/(\varepsilon_B + 1) = eR^3/R^2$. It becomes equal to the dipole moment $d_{ex} = E_{ex} R^3$ generated by the external field at $E_{ex} = e/R^2 \approx 1500V/cm$ for $R = 1000\AA$.

The coordinate $z^{opt}_B$ determined from the potential (3) is actually the $z$-coordinate of the center of charge (not the center of the cluster). $z^{opt}_B$ coincides with the center of cluster only for $\varepsilon_B \to \infty$. If $z^{opt}_B$ obtained from (3) is equal or less than the cluster radius $R$, one has to take into account the short-range van der Waals potential, which prevents the cluster from emerging from helium.

References

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Figure caption: The schematic view of the charged clusters below helium surface.
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