Theory of the stability of multielectron bubbles in liquid helium

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Abstract.
We have performed numerical calculations to investigate the stability of multi-electron bubbles in liquid helium-4. In our calculations, for each bubble shape considered we find the equilibrium distribution of the electron gas on the inside surface of the bubble. For a stationary bubble, we find that whenever the pressure in the liquid is positive the bubble is unstable against fission into two bubbles. We find that a moving bubble can be stable within a certain region of the pressure-velocity plane. At the high pressure boundary of this region, the bubble undergoes fission, and on the low pressure boundary the bubble explodes.

Electrons above the free surface of a bath of liquid helium will remain outside the helium because of a 1 eV potential barrier [1]. If a positive voltage is applied to an electrode immersed inside the liquid, when the field reaches a critical value, the surface of the liquid becomes unstable and a large number of electrons enter into the liquid through the formation of multi-electron bubbles (MEB). Each of these bubbles typically contain \(10^7 \sim 10^8\) electrons with radii about 100 \(\mu\text{m}\) [2].

There have been several theoretical investigations of the stability of MEB [3, 4]. In the simplest model, the electrons are treated as classical particles with Coulomb repulsion and so are localized at the inner surface of the bubble in a layer of zero thickness. The distribution of electrons is such that the electric field is everywhere exactly normal to the surface. This ensures that the charge distribution is in equilibrium. The stable bubble shape is attained by minimizing the total energy

\[
E = E_S + E_V + E_C.
\] (1)

Here \(E_S = \sigma S\) is the surface energy with \(S\) the surface area and \(\sigma\) the surface tension (0.36 erg cm\(^{-2}\) at 1.3 K), \(E_V = PV\) the volume energy with \(P\) the applied pressure and \(V\) the bubble volume. \(E_C\) is the Coulomb energy given by \(E_C = \int (\epsilon E^2/8\pi) \text{d}V\), where \(\epsilon\) is the dielectric constant (1.0573 at low temperature), and the electric field \(E\) is nonzero only outside the bubble. For a spherical bubble of \(Z\) electrons, the radius that gives the minimum value of the energy is the solution of the equation \(R_0(8\pi \epsilon PR_0 + 16\pi \epsilon \sigma) = Z^2 e^2\).

To consider whether the spherical shape is stable, we introduce the shape parameters \(\eta_{lm}\) and write

\[
R(\theta, \phi) = R_0 \left\{ 1 + \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \eta_{lm} Y_{lm}(\theta, \phi) \right\}.
\] (2)

\(\eta_{l,m} = \eta_{lm}^*\)
If the $\eta_{lm}$ are small, one can show that to the second order, the total energy is

$$E = 12\pi \sigma R_0^3 + \frac{16\pi}{3} PR_0^3 + \sum_{l=0}^{\infty} \frac{1}{2} \kappa_l \sum_{m=-l}^{l} |\eta_{lm}|^2,$$

where the spring coefficients $\kappa_l$ are given by

$$\kappa_l = \begin{cases} 
6\sigma R_0^2 + 4PR_0^3, & (l = 0) \\
(l-2)(l-1)\sigma R_0^2 - 2(l-1)PR_0^3, & (l \geq 1)
\end{cases}$$

From this one can see that the bubble is stable against spherically symmetric perturbations provided that $6\sigma R_0^2 + 4PR_0^3 > 0$. This leads to the condition $P > P_c = \left(\frac{27\pi e^4}{4Z^2e^2}\right)^{1/3}$. For $l = 1$, the spring coefficient $\kappa_1$ is zero; this is to be expected since a perturbation of the form $\eta_{1m}Y_{1m}(\theta, \phi)$ corresponds to a simple translation of the bubble in some direction. For $l = 2$ the spring constant $\kappa_2$ is zero if the pressure is zero, and negative if the pressure is (arbitrarily small) positive. This means that a spherical bubble within this pressure range is unstable against an $l = 2$ shape change. The higher $l$ spring constants are all positive at zero pressure but each becomes negative if the pressure is increased to a sufficiently positive value. In addition, if a negative pressure is applied (but not negative with respect to $P_c$), all of the spring constants will be positive (excluding $\kappa_1$ which is always zero) and so the bubble should be stable [4].

The stability of the bubble at zero or small positive pressure is of especial importance since in the experiments that have been performed so far there has been no applied pressure apart from the very small hydrostatic pressure due to the distance the bubble is below the free surface. Although the above perturbation analysis shows the instability of a spherical bubble against the $l = 2$ mode, it does not rule out the possible existence of a non-spherical stable shape (energy local minimum) at some finite $\eta_{lm}$. So we have performed numerical calculations by the finite element method [5] without restricting the parameters $\eta_{lm}$ to being small. For each shape we allow the electrons to redistribute themselves so as to minimize the energy and to make the electric field inside the bubble zero.

The result of this investigation is that for all positive pressures there is no barrier to fission, whereas for negative pressures there is a barrier. This result holds for all values of $Z$. To illustrate the path to fission, we describe results obtained for a simplified calculation in which only $l = 0, 2$ and $m = 0$ contributions are retained. Thus we write

$$R(\theta, \phi) = a_0 + a_2(3\cos^2\theta - 1).$$

Within this simplified model, fission occurs when $a_2 = a_0$; and the bubble develops a hole along the z-axis, i.e., takes on a donut shape, when $a_2 = -a_0/2$. In Fig. 1 we show examples of contour plots of the energy in the $a_0$-$a_2$ plane for $Z = 10^6$. For a pressure of $-0.03$ mbar (Fig. 1(a)), there is a stable minimum with $a_2$ equal to zero, i.e., the bubble is spherical. When the pressure is zero (Fig. 1(b)), there is still a point in the plane at which the energy of the bubble is stationary with respect to both $a_0$ (at 23.8 $\mu$m) and $a_2$ (at 0), but it is now possible to reach the fission line from this point without passing over any energy barrier. Note that along this path there is, of course, an increase in the value of $a_2$ but also a substantial decrease in $a_0$. Once the pressure becomes positive (Fig. 1(c)), there is no point in the $a_0$-$a_2$ plane where the energy is stationary.

These results can be compared with the earlier calculations by Tempere et al. [4], who also investigated the stability against fission. To simplify the calculation, they made the approximation that the charge density was uniform over the surface of the bubble and concluded there exists an energy barrier which prevents fission, whereas we find no barrier. To understand
Figure 1. Contour lines of constant energy for an MEB containing $10^6$ electrons for three different pressures. The energy spacing between contour lines is 0.05 eV.

In order to find the shape of moving bubbles and the range of velocity and pressure for which they are stable, we have performed computer simulations by balancing the surface forces due to the potential flow and the electron repulsion. For an MEB with $Z = 10^6$ the stable shapes for three velocities at zero $P_0$ are shown in Fig. 2. In Fig. 3 we show a plot of the region in the pressure-velocity plane in which the bubble is stable. This region is bounded by two lines. For small velocities there is a critical positive pressure at which the bubble undergoes fission.
Figure 2. The shape of an MEB containing $10^6$ electrons for three different velocities. The pressure far away from the bubble is zero.

Figure 3. Plot of the stable region in the pressure-velocity plane for an MEB containing $10^6$ electrons. It is bounded by the lines on which the two different types of instability occur. Along the dashed line the bubble becomes concave at the poles and the numerical calculations become inaccurate.

At negative pressures the bubble becomes unstable against expansion. For zero velocity this expansion is isotropic. We are only able to perform the numerical calculation until the bubble becomes concave at the poles (shown by the dashed line).

We note that in the above calculation we have treated the liquid as inviscid although, of course, helium above the lambda point has a finite viscosity and below the lambda point the liquid still has a normal fluid component. At sufficiently low temperatures the density of the normal fluid becomes very small and, in addition, the mean free path of the excitations making up the normal fluid becomes comparable to the radius of an MEB. Under these conditions, it appears that the only effect of the normal fluid is to determine the mobility of an MEB and there should be no effect on the shape change or the stability. At high temperatures where the helium is in the normal state, the situation is not so clear. It is known that when the Reynolds number is large (but not so large that the flow becomes turbulent) the viscosity results in a thin boundary layer on the surface of the bubble and the pressure at the bubble surface is close to the value that would result from potential flow [6]. This general idea would suggest that the inviscid approximation should give reliable results for the stability of MEB over a wide range of Reynolds number.

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